GENERALIZATION OF INTERNAL CENTRIFUGAL
ZONE GROWTH OF METAL-CERAMIC COMPOSITES

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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 and extend present ICZG systems. In a previous year, we developed one-dimensional models for (hypothetical) infinitely large samples heated by long RF induction coils and predicted an S-curve instability phenomenon associated with spontaneous coupling of the RF field to materials whose electrical conductivity increases significantly with temperature. In a succeeding year, this model was extended to two dimensions to account for finite samples and coils. During the present year, we have returned to the one-dimensional modeling and have predicted additional instabilities (S shaped with a cusp) on melting which are "piggy-back" with the S-curve instabilities at lower temperatures. At CMU, the low temperature S instability has been demonstrated for silicon; whereas, in cooperation with Oak Ridge, the predictive capacities of the modeling have been used to engineer around instabilities in the CrO_3 -Mo systems and to extend ICZG to a number of other materials.

PROGRESS REPORT

A. Technical Progress

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; 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 was reported in Progress Report C00-2407-1 and Technical Report C00-2407-2. The most significant result was the prediction of an S-curve instability phenomenon whereby poor electrical conductors whose conductivity increases significantly with temperature can couple strongly and spontaneously to an RF field.

During the second year, as summarized in Progress Report C00-2407-3, boundary conditions were developed for electromagnetic fields in finite samples heated by finite induction coils and computational algorithms were formulated for two-dimensional temperature profiles. Apparatus was also constructed at CMU in order to try to observe the S-curve instability phenomenon predicted by the one-dimensional modeling.

During the first nine months of the current contract year, we have returned to the one-dimensional modeling because its tractability has enabled us to model instabilities and transient phenomena associated with

melting. Details are contained in an extensive technical report, C00-2407-4, which accompanies. Some noteworthy highlights of this and other work follows:

1. Melting Instability

Our modeling predicts an instability phenomenon associated with the change of electrical conductivity on melting. Instead of an S-curve instability as modeled previously for solid samples, one finds an instability wherein a kink replaces one smooth knee of the S-curve (see point D of Figure (3-5)^{*}). This additional instability phenomenon is of considerable technological significance since it leads lack of control and must be coped with or eliminated in ICZG and related processes (such as Skull Melting).

2. "Piggy-back" Instabilities

As evident from Figure (3-5)^{*}, the instability that occurs on melting is "piggy-back" with the S-curve instability which occurs in the solid at lower temperatures. In Figure (3-5)^{*} there are five numbered steady states, all of which are metastable except number 3 which is unstable (see Figure (3-14)^{*} which pertains to the stability analysis).

3. Relation to Catastrophe Theory¹

We have studied these instabilities as a function of continuous variation of parameters such as skin depth and activation temperature (parameter associated with activated thermal conductivity) and are beginning to understand the resulting behavior in light of Catastrophe Theory. Note especially the series of Figures (3-15)^{*} to

* Starred figure numbers refer to the Technical Report C00-2407-4.

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

(3-24)* which could be used to generate a "catastrophe surface."

4. Transient Melting

All of our previous modeling has dealt with steady states. We have now developed transient solutions (see Figure (3-29)*) which show that melting first begins in a zone intermediate between the axis of the sample and the outer radius. To the extent that this result can be generalized to two-dimensional models, it predicts the sort of toroidal zone shapes which formerly mystified experimentalists.

5. Experimental Demonstration of S-curve Instability

We have demonstrated experimentally the existence of the S-curve instability for induction heating of a one-inch diameter cylinder of silicon at 450 kHz. The sample was heated by means of a susceptor to near its melting point where it was coupled directly to the RF field. RF power was then gradually decreased, resulting in a smooth drop in temperature to about 900°C where the temperature suddenly crashed to below 700°C. RF power was then increased but cooling continued, thus demonstrating the hysteresis associated with the S-curve. (Although melting did not occur, the crash in temperature is analogous to a crash from B to C in Figure (3-5)*; restoration of state C would then necessitate further preheating or increase of coil current beyond point A to get off the lower branch of the S-curve.)

6. Liaison with Oak Ridge - Practical Implications

As during previous contract periods, we have maintained close contact with scientists at Oak Ridge. This time, Dr. Clark and

Mr. Holder of Oak Ridge visited our laboratory (instead of the reverse) and telephone contact has been frequent.

In order to optimize exchange of information, it was finally decided to employ Mr. Hartzell of CMU at the Nuclear Division of Union Carbide at Oak Ridge for about two months during the summer of 1976. The following are examples of the practical implications which stem from such liaison:

a) Application of Modeling to the Cr_2O_3 - Mo System

If the frequency is not well-matched to the size and the electrical conductivity of the sample, the instability at melting can cause a large increase in surface temperature, rupturing the wall containing the melt and causing the melt to spill. The surface temperature increase can occur very rapidly, even within a matter of minutes. Violent melting of this nature has been observed in Cr_2O_3 - Mo by Dr. Clark's group at Oak Ridge National Laboratory. For more than a year, the true cause was unknown and solutions that were tried proved to be counter-productive. Dr. Clark's group reasoned that since the melt was not being contained, the skin depth of the electric field was too small; but our modeling has shown that this behavior is, in fact, a manifestation of the heating instability that can be eliminated by a decrease in skin depth. Our analysis has subsequently been proven correct.

We suggested that the ratio of skin depth to sample radius should be decreased through an increase in either sample size, RF frequency, or electrical conductivity. Subsequent experimentation has shown that doubling the sample diameter mitigates the

instability, but the engineering of a higher conductivity through increased concentration of molybdenum was seen as a way to eliminate the instability entirely.

In order to select the proper concentration, the electrical conductivity of hot-pressed samples of Cr_2O_3 - Mo were measured at Carnegie-Mellon University via a four-point probe technique. The results appeared consistent with expectation and with what little data is public. Based on these measurements, there was little doubt that the skin depths of the samples being induction-heated at Oak Ridge were too large and were, indeed, the root cause of the nearly uncontrollable instability. Increasing the molybdenum concentration from 20 mole percent to the 30 mole percent range completely eliminated the instability. Intermediate concentrations near 25 mole percent cause the surface temperature actually to decrease at melting, a result which is well illustrated by our modeling in a series of Figures (3-15)* through (3-21)*, which show the instability going through a transition, as the skin depth is decreased, from a large increase in surface temperature at melting to a small decrease in surface temperature before the instability vanishes entirely.

b) Predictive Capability

One of the objectives originally proposed for this research was to use the predictive capabilities of the modeling to determine which material systems can be centrally melted and under what conditions (e.g., sample size, RF frequency, induction coil current, generator power).

Solution of the coupled electric field and temperature differential equations, for a particular set of material parameters, yields information concerning possible instability problems, minimum pre-heat temperature, surface temperature as a function of solid wall thickness (once melting begins), and much more. This type of analysis is really too extensive for an initial appraisal of a material system. Since we intend to evaluate many dozens and possibly hundreds of materials, a less sophisticated level of analysis is in order.

We have constructed a number of criteria, derived principally from the results of previous modeling, which we believe are essential for central melting. In particular, we have established formulas for the following, which are recommended for successful ICZG:

- i) A minimum sample size (in order to get a sufficient surface to center temperature difference)
- ii) A minimum frequency for RF heating (to get a skin depth sufficiently small to couple with the sample)
- iii) An RF skin depth of about one half the sample radius (for skin depth too small, one gets surface melting; for skin depth too large, one gets undesirable instability phenomena)
- iv) A minimum RF current (needed to sustain the sample near its melting point)

Details will be presented in a forthcoming technical report;

however, a few generalizations are noteworthy. Materials with large electrical conductivities (e.g., non-oxides) generally require very large sample sizes and RF coil currents, perhaps impractical with the possible exception of boron nitride and silicon carbide. On the other hand, oxides require very high frequencies with the possible exception of the dioxides of thorium, hafnium, uranium and zirconium. Success with other oxides usually requires the admixture of metals such as with the Cr_2O_3 - Mo system discussed above. With the aid of these guidelines, ICZG was extended to several new materials at Oak Ridge during the summer of 1976; results are now being prepared for publication.

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		
	1975						1976			1976		
	D	J	F	M	A	M	J	J	A	S	O	N
Prof. Sekerka (Principal Invest.)	← 10% →							45%		← 10% →		
Mr. Hartzell (Graduate Student)	← 100% →											
	(July and August at Oak Ridge)											

During the remaining three months of this contract period, emphasis will be refocused on the two-dimensional modeling, concomitant electromagnetic problems, and on extension of the experimental work at CMU to other materials.

C. Reports

Progress Report C00-2407-1	}	Submitted with first renewal
Technical Report C00-2407-2		
Progress Report C00-2407-3		Submitted with second renewal
Technical Report C00-2407-4	}	Submitted herewith
Progress Report C00-2407-5		

Note: Technical Report C00-2407-4 contains information for several journal articles which are being prepared for submission to the Journal of Crystal Growth.

D. Meetings

AIME White House Conference on "U. S. Energy & Mineral Supplies: Meeting National Needs." Attended by Principal Investigator May 1976.

First European Conference on Crystal Growth (ECCG-1) Zurich, Switzerland, September 12-18, 1976. The Principal Investigator is an invited speaker (abstract attached).