LEVEL III

FIRST QUARTERLY PROGRESS REPORT

A061572

LABORATORY APPARATUS FOR THE GENERATION OF RAPIDLY QUENCHED METAL POWDERS.

Quarterly progress rept. no. 1,
1 May to 31 July 1977,

Prepared for:

OFFICE OF NAVAL RESEARCH

ARLINGTON, VIRGINIA 22217

Contract No. N00014-77-C-0373, ARPA Order 3363
Start: 01 May 1977
Expiration Date: 30 April 1978

ARPA Order No. 3363

Program Code No. NR-039-141

4 - Ref. Not for DDC
5 - A061572

31 Jul 77

Principal Investigator: Julius Perel,
John F. Mahoney
Bernard Kalensher
(213) 793-3191

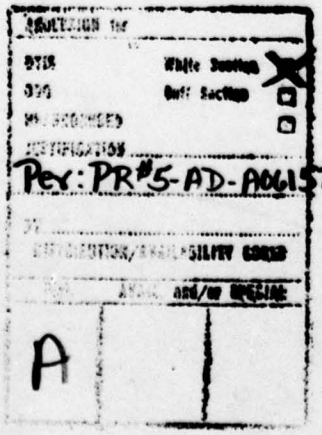
17p.

Prepared by:

391701 PHRASOR TECHNOLOGY
110 South Euclid Avenue
Pasadena, California 91101
Duarte,

CONTENTS

ILLUSTRATIONS



SECTION 1

SUMMARY

The objective of this program is to develop a table top rapid solidification powder generator utilizing electrohydrodynamic (EHD) techniques. EHD is a method of producing very small droplets from a liquid using intense electric fields. The intense field is attained by the application of nominal voltages (several kilovolts) to an emitter having small radius of curvature. When the voltage is applied, the intense field provides sufficient forces upon the liquid surface to form a liquid jet from which droplets are produced. If small droplets are formed from a liquid metal, at the melting point, the droplets will solidify because of heat radiation at rates related to the material properties and the droplet size. The droplet sizes can be varied through a combination of varying the applied voltage and the material mass flow rate. The equipment required to produce metal powders using EHD techniques includes a vacuum system, a droplet source, a particle collector, and the appropriate electronics and test instruments.

During this report period, the vacuum system was assembled for the operation of these specific experiments. The droplet source was fabricated and assembled as was the particle collector and the material feed system. The diagnostic instrumentation and the safety control system are partially complete. A theoretical analysis was initiated to study the variation of the cooling rate with particle size, optical absorption coefficient, and index of refraction. The aim of the analysis is to anticipate cooling rates that are very different from that predicted by the Stefan-Boltzmann law.

SECTION 2

PROGRESS DURING REPORT PERIOD

2.1 EXPERIMENTATION

2.1.1 INTRODUCTION

The experimental facility, including the testing and diagnostic apparatus necessary to initiate experiments using a Pb-Sn alloy is nearly complete. Droplet generation experiments will be conducted during the early portion of the next report period. The experimental task status is indicated in Table I.

TABLE I

EXPERIMENTAL TASK STATUS

Task	Completion Status (%)
Test Chamber (Operational)	100
Droplet Source	100
Collector System	100
Source Feed System	100
Time-of-Flight Diagnostics	80
Test Interlocks and Controls	50

A brief description of each experimental task is included in the following subsections.

2.1.2 TEST CHAMBER

The microparticle test chamber, illustrated in Figure 1, has been assembled and consists of a stainless steel vacuum chamber enclosure 1 foot in diameter by 3 feet in length. The chamber body is outfitted with seven ports that provide ample access for test probing and viewing. In addition, the chamber is equipped with a copper liner which can be chilled with liquid nitrogen, as necessary, to improve or maintain vacuum. Pumping is provided by a 6-inch diffusion pump rated at 1800 liters/sec and is augmented by a cryogenic baffler system. The system is capable of achieving a vacuum level in the 10^{-7} torr range.

2.1.3 LIQUID METAL DROPLET SOURCE

The liquid metal droplet source to be used with initial tests involving a Pb-Sn alloy has been constructed during the report period. The microparticle source consists of a stainless steel reservoir and delivery tube, as shown in Figure 2. The microparticle emitter is a tungsten capillary structure with an orifice dimension of 0.003 inch. Two metal sheathed ceramic-compact heaters provide adequate heat to keep the alloy in a molten state (approximately 300°C). An extractor electrode containing an aperture concentric with the emitter aids in forming the high extraction fields. Also mounted on the microparticle source is a second electrode (not shown) designed to prevent electrons from bombarding the droplet source. Two thermocouples, one mounted near the droplet nozzle and one attached to the droplet reservoir, monitor the temperature. Although the droplet source and electrodes are fabricated from stainless steel, this should provide ample resistance to any corrosive effects of the Pb-Sn alloy.

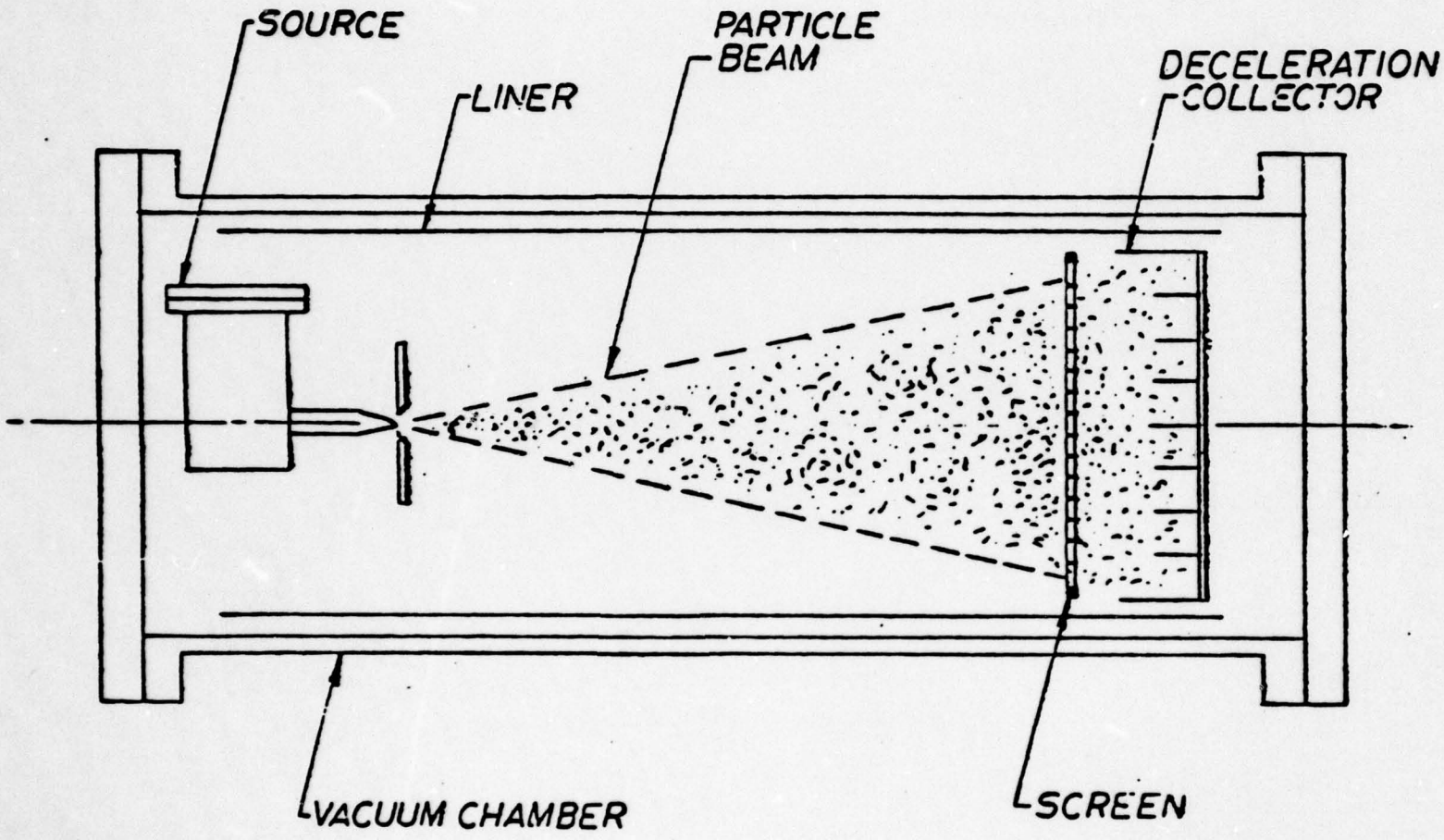


Figure 1. EHD Particle Generation System

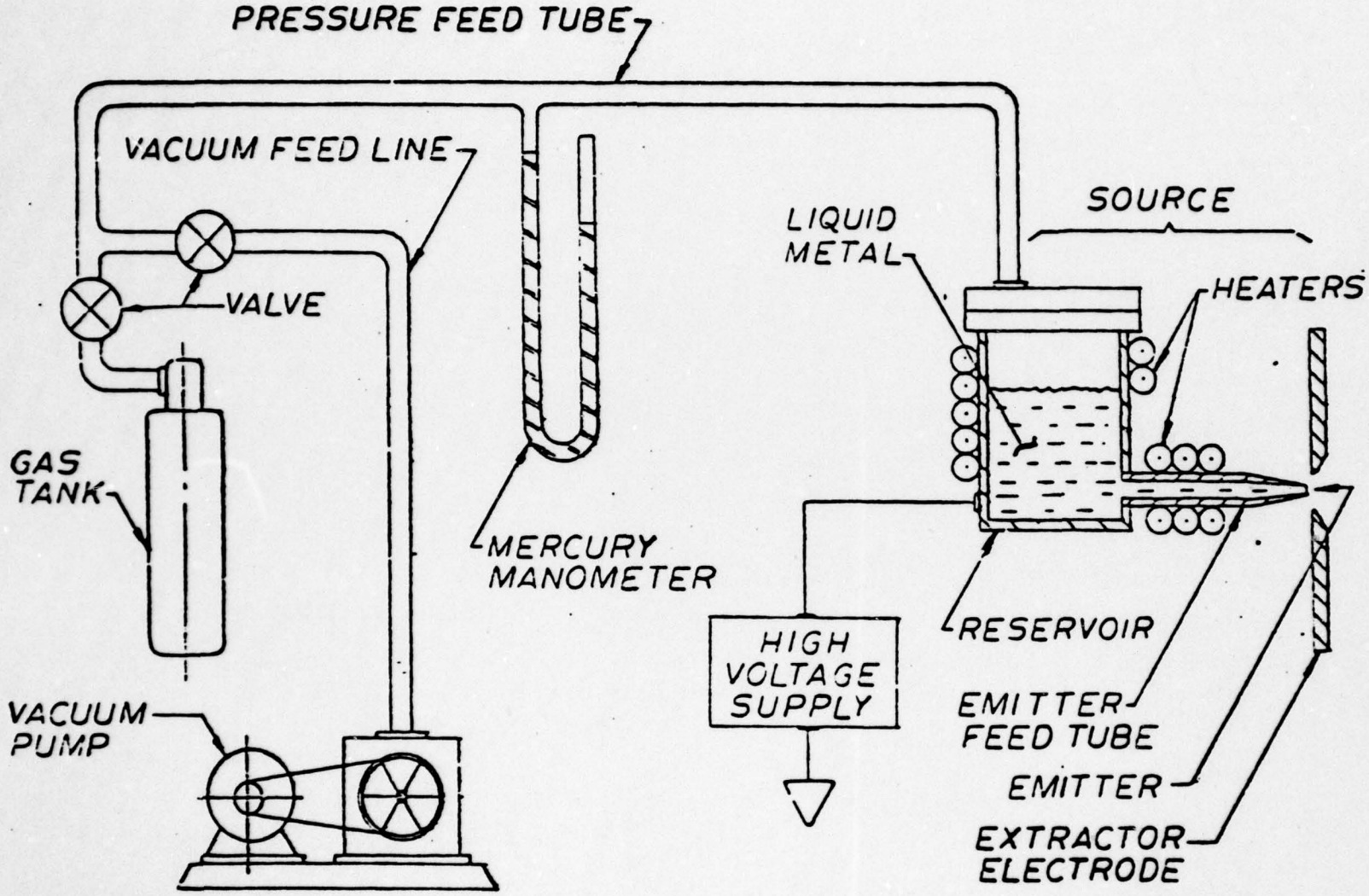


Figure 2. Source and Feed System

2.1.4 COLLECTOR SYSTEM

The droplet collector fabricated for initial testing consists of a hemispherical copper surface. A grid structure, shown in Figure 1, is mounted directly in front of the copper collector and electrically isolated from the collector. The grid is used to suppress any secondary electrons that leave the collector and thereby prevent any false current readings. The dc collector current is monitored with an oscilloscope used to measure the current during time-of-flight testing.

2.1.5 SOURCE FEED SYSTEM

A feed system for controlling the mass flow rate of the liquid metal during operation of the source has been constructed. The feed system also serves to outgas the liquid metal reservoir during the melting phase. An easily controllable valving system allows the reservoir to be evacuated, or to be pressured with an inert gas such as argon.

2.1.6 TIME-OF-FLIGHT DIAGNOSTICS

The shielded collector described previously is used to obtain the time-of-flight waveforms necessary to evaluate the charge-to-mass ratio of the particles. This parameter is useful in evaluating the sizes of the generated particles. The droplet distribution can also be determined from information contained in the time-of-flight trace which displays the collected current as a function of time. The collector current is returned to ground through a low impedance load resistor. The voltage across the load resistor is monitored with an oscilloscope.

A high voltage switch has been fabricated and is used to initiate the time-of-flight data taking process. Switching the high voltage to ground momentarily disrupts the beam to produce the time-of-flight

trace which is photographed with a Polaroid camera. The oscilloscope is triggered simultaneously with the shorting action of the switch. The shorting mechanism was designed with a switching time of approximately 2 microseconds. This task is 80 percent complete. After the TOF preamplifier requirements have been selected (dc to several MHz) and the unit itself acquired and tested, the time-of-flight system will be complete.

2.2 THEORETICAL

The most important quantity to be measured in the rapid quenching of droplets is the cooling rate. In order to reduce the uncertainty in the measurement of this quantity, it is desirable to know what cooling rates one can expect to obtain during the course of the experiment. A preliminary theoretical analysis has been made of the cooling rate, and the results of this analysis are now presented.

The cooling rate of the droplet is given by

$$\frac{dT}{dt} = \frac{-P}{mC_p} \quad , \quad {}^{o}K/sec \tag{1}$$

where, P = power (watts) radiated by the droplet, m and C_p are the mass (kg) and specific heat (joules/kg ^{o}K), respectively, of the droplet, T = droplet temperature (^{o}K), and t denotes time. The radiated power is given by the Stefan-Boltzmann law,

$$P = 4\pi a^2 \epsilon \sigma T^4 \tag{2}$$

where, a = radius of droplet, σ = Stefan-Boltzmann constant = 5.67×10^{-8} watts/meter^2 $^{o}K^4$, and ϵ is the total emissivity of the droplet. In general, the total emissivity is a function of the droplet temperature and radius. Values of ϵ are usually determined by experiment. However, these

experimental values generally apply to large bodies ($a \geq 1$ cm) with well defined surface characteristics (i.e., smooth, rough, oxidized, etc.). It is very difficult to talk about the surface characteristics of a droplet whose radius is of the order of 1 micron ($= 10^{-6}$ meters). Thus, it would be advantageous to have at ones disposal an expression for ϵ which would be valid for very small ($a \approx 1$ micron) bodies. Such an expression has been derived; however, it is only approximate and does not explicitly contain the temperature.

Consider a homogeneous, spherically shaped, metallic liquid droplet which is isotropic to internally generated thermal radiation. The thermal energy originating in a differential volume element within the droplet radiates uniformly in all directions and eventually reaches the surface of the droplet. As the energy passes through the droplet, part of it is absorbed by the medium. If it is assumed that none of the radiation within the droplet undergoes scattering by the medium, and that none of the radiation reaching the surface of the droplet is reflected back into the interior, then, by summing up the contributions to the total radiation from the differential elements of the droplet, one obtains the following approximate expression for the total emissivity.

$$\epsilon = n^2 \left[2 - \frac{4}{\alpha a} + \frac{7}{\alpha^2 a^2} - \frac{6}{\alpha^3 a^3} + e^{-2\alpha a} \left(\frac{2}{\alpha a} + \frac{5}{\alpha^2 a^2} + \frac{6}{\alpha^3 a^3} \right) \right] \qquad (3)$$

Here, n = average index of refraction of the droplet, and α = average optical absorption coefficient (meter^{-1}) of the droplet. Both n and α are functions of the temperature and electrical conductivity of the droplet. Typical values of n for metals are

$$0.1 \leq n \leq 1000$$

and for α,

$$10^5 \leq \alpha \leq 10^8 \text{ meter}^{-1}.$$

It is to be noted in Eq. (3) that ϵ becomes zero when $\alpha a = 0$. Figure 3 shows a plot of ϵ/n^2 versus αa. Note further that in the region of $\alpha a = 1$ the emissivity changes in nature.

In principle, the total emissivity must never exceed unity. If Eq. (3) were accurate and if the values assigned to α and n were physically correct, then the calculated value of ϵ would satisfy, $\epsilon \leq 1$. Eq. (3) is useful in that it shows the approximate functional dependency of ϵ on the product, αa.

By combining Eqs. (1), (2), and (3) and writing

$$m = \frac{4}{3} \pi a^3 \rho ,$$

where, ρ = density (kg/meters^3) of droplet, the cooling rate of the droplet can be written

$$\frac{dT}{dt} = Kn^2 Q(\alpha,a), \quad {}^{o}K/sec \tag{4}$$

where,

$$K = \frac{T^4}{\rho C_p} , \quad Q(\alpha,a) = \frac{3\sigma}{a} \cdot \frac{\epsilon}{n^2}$$

Figure 4 shows a plot of $Q(\alpha,a)$ or $(dT/dt)\,(Kn^2)^{-1}$ versus the droplet radius a, for the following values of absorption coefficient:

$$\alpha = 10^5, 10^6, 10^7 \text{ meters}^{-1}.$$

An interesting feature of the graph is that the cooling rate increases with decreasing values of the radius, but reaches a plateau for sufficiently small values of a in the region of $\alpha a = 1$. As the absorption

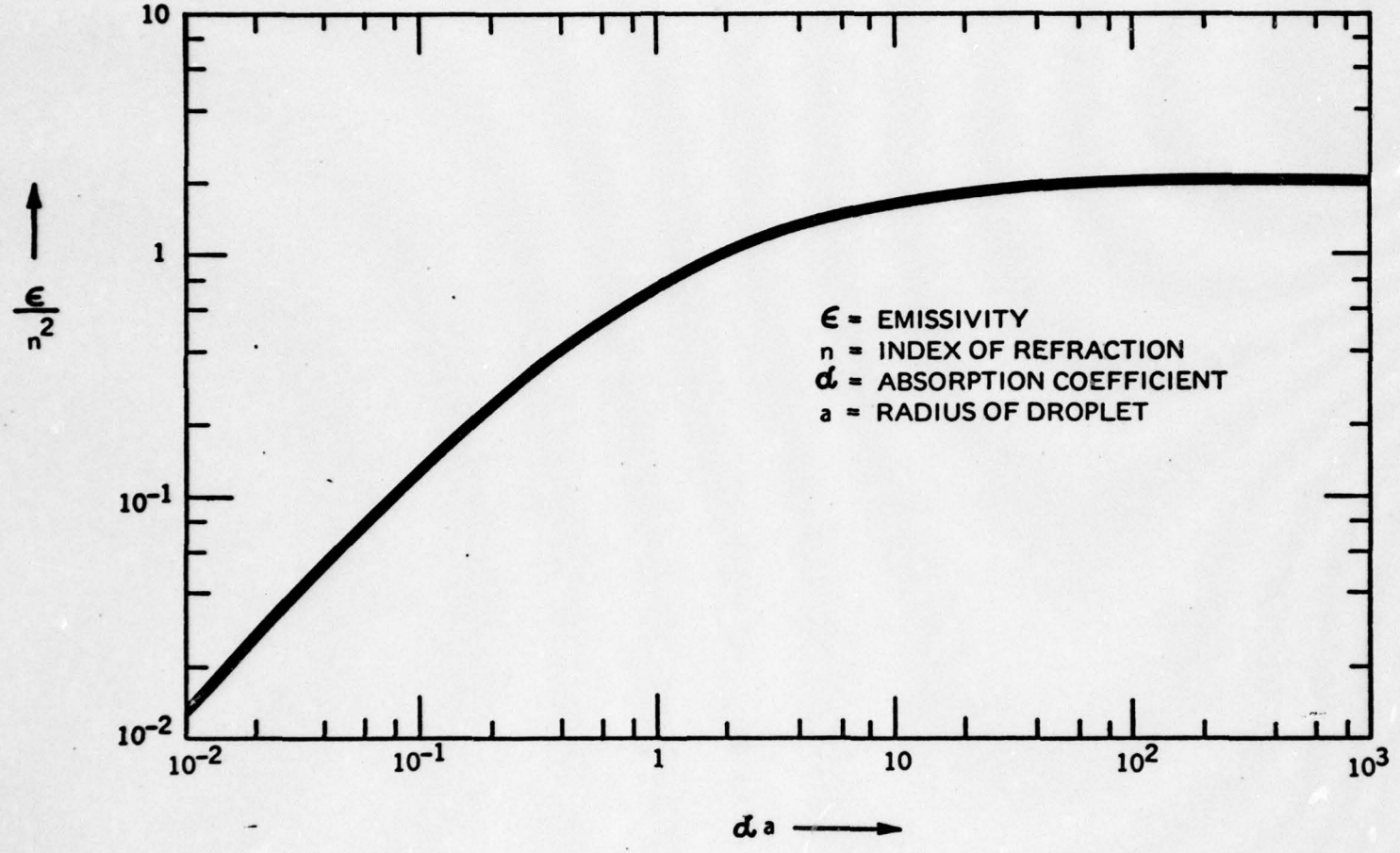


Figure 3. Emissivity versus αa

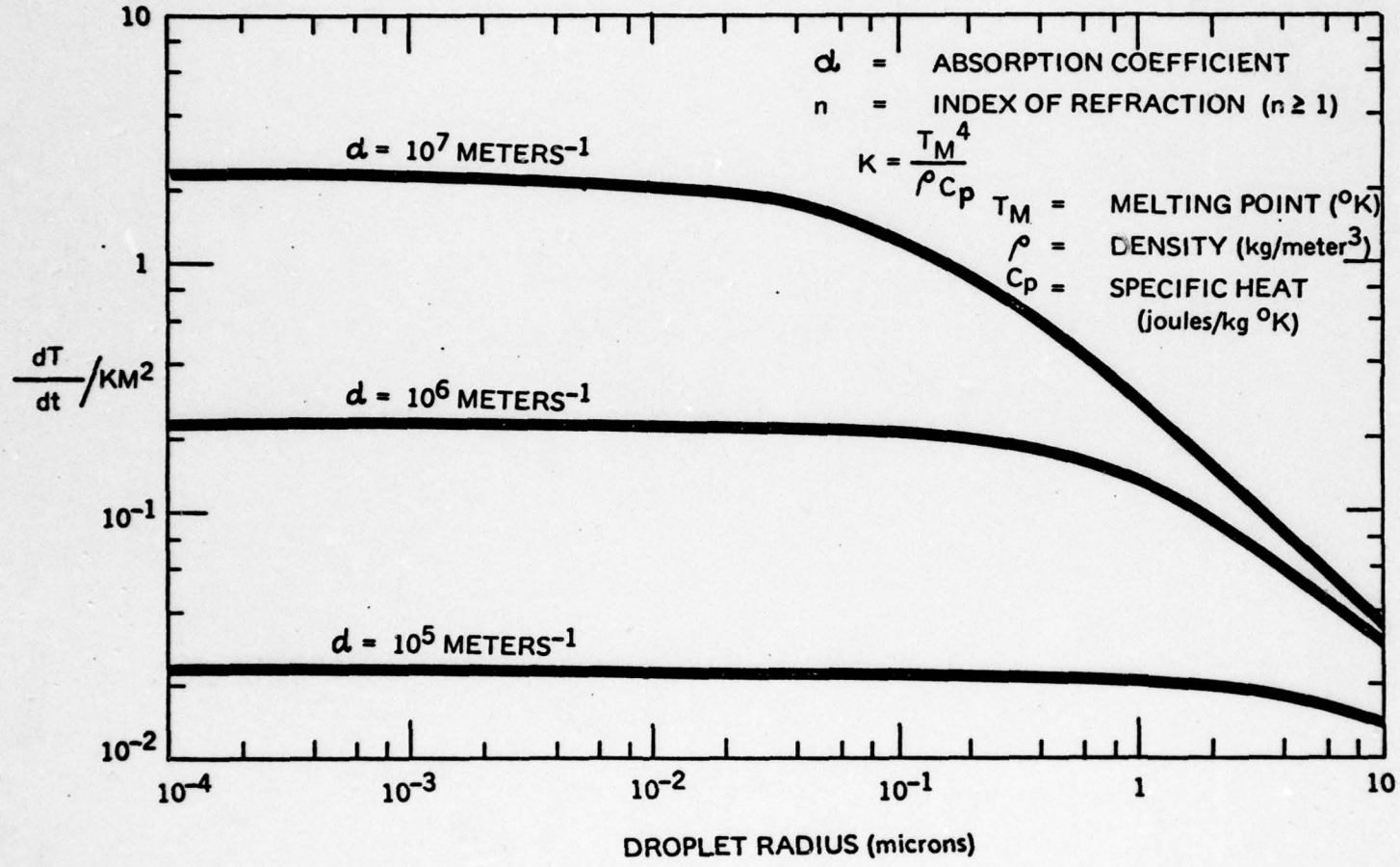


Figure 4. Cooling Rate of Droplet versus Droplet Radius

coefficient increases, this plateau is reached for smaller and smaller values of the radius. It would thus appear that, for a given absorption coefficient, no advantage is gained by reducing the droplet radius below a specified value.

Table II shows values of the constant K for several different metals. Here, the temperature T represents the melting temperature of the metal. Suitable values of n for these metals have not yet been found in the literature.

In the derivation of Eq. (3), it was assumed that the index of refraction, n, and the absorption coefficient, α, were constants. Actually, they both are functions of the frequency of radiation, and this functional dependency must be taken into account if an accurate expression for total emissivity is to be derived. This generalization of the radiation problem will be discussed in the next quarterly report. At that time, it is expected that a more accurate description of the cooling rate can be presented.

The above studies are relevant to the mechanism of radiative cooling. After this analysis has been completed, a theoretical study will be made of the properties of the materials and the particle sizes that affect the cooling rates. The entire EHD process will be analyzed in terms of its application to rapid cooling of metal powders.

TABLE II

VALUES OF $K = \frac{T_m^4}{\rho C_p}$ FOR SEVERAL METALS

Metal	$\frac{T_m^4}{\rho C_p}$ (= K), $\frac{\text{meters}^3\ {}^oK^5}{\text{joules}}$
Aluminum	3.035×10^5
Antimony	4.740×10^5
Beryllium	9.558×10^5
Cobalt	1.634×10^6
Copper	8.329×10^5
Gold	1.144×10^6
Iron	1.851×10^6
Lead	8.394×10^4
Nickel	1.391×10^6
Palladium	3.10×10^6
Silver	8.147×10^5
Tin	2.928×10^4
Zinc	8.476×10^4

SECTION 3

CONFERENCES ATTENDED

John Mahoney and Julius Perel attended the Material Research Council Meeting on Rapid Solidification Technologies held in La Jolla on 11-13 July 1977. The background technology to this program and the progress to date was presented by Julius Perel under the title "Rapid Solidification by Electrohydrodynamics." The use of electrohydrodynamics to produce rapidly quenched metal powders was a new technique presented at the conference.

SECTION 4

PROBLEMS

No significant problems have been encounted that could affect the ultimate completion of the program goals and the effort required to achieve the program objectives appears sufficient at the present time.

SECTION 5

CONCLUSION

The program for the next report period includes the initial testing of a Pb-Sn alloy. This test is anticipated at the beginning of the period, as soon as the component parts such as the EHD source, particle collector, and test electronics are made operational simultaneously. The performance parameters of the source will be examined and powder production will be characterized in terms of source voltage, sample feed rate, beam current, etc. Particle sizes and size distributions will be examined using time-of-flight techniques and will be verified by optical techniques. A source for the production of Aℓ powder will be designed and testing will probably be initiated toward the end of the report period. The analytical study on cooling rate will be continued, and the application of the EHD process to powder production investigated further. Preparations will be made to attend the "International Conference on Rapid Solidification Processing Principles and Technologies" to be held on 13-16 November 1977 in Reston, Virginia. A talk will be prepared, based upon work on the program, under the title, "Electrohydrodynamics Generation of Submicron Particles for Rapid Solidification."