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RADIATIVE AND GAS COOLING OF FALLING MOLTEN DROPS

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16. ABSTRACT

This report calculates the supercooling rate and solidification time for molten drops of niobium, copper, and lead. Calculations for both radiation and helium gas cooling are presented in order to estimate the influence that the presence of helium gas would have upon the cooling rate of falling drops in the Marshall Space Flight Center space processing drop tube.

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NOMENCLATURE

Symbol	Defintion
A	Surface area of drop (cm^2)
C	Specific heat (J/g-K)
D	Diameter of drop (cm)
h	Heat transfer coefficient ($J/s\text{-}cm^2\text{-}K$)
H_f	Latent heat of fusion (J/g)
K_f	Thermal conductivity at T_R (J/s-cm-K)
m	Mass of drop (g)
t	Time (s)
t_c	Time required for drop to cool from T_i to T_f (s)
t_s	Time required for complete solidification of drop (s)
T	Temperature of drop (K)
T_i	Initial temperature of drop (K)
T_f	Final temperature of drop (K)
T_m	Melting temperature (K)
T_o	Ambient temperature (K)
T_R	Thin film temperature (K)
T_s	Surface temperature of drop (K)

NOMENCLATURE (Concluded)

Symbol	Definition
T_∞	Free stream temperature (K)
U_∞	Free stream velocity (cm/s)
Greek	
ϵ	Emissivity
σ	Boltzman's constant ($J/s\text{-}cm^2\text{-}K^4$)
ν_f	Kinematic viscosity at T_R (cm^2/s)

RADIATIVE AND GAS COOLING OF FALLING MOLTEN DROPS

INTRODUCTION

Molten drops of various sizes of selected materials can be cooled and solidified within the 2.5 s of free-fall time available in the Marshall Space Flight Center (MSFC) drop tube facility. For materials with a high melting temperature, above approximately 1800 K, radiation cooling is sufficient to completely solidify the drops of small diameter (1 to 5 mm). However, for drops of materials with lower melting points, heat loss by radiation cooling alone will not allow complete solidification within the 2.5 s time limit. Therefore, it becomes necessary to back-fill the drop tube with known pressures of a cooling gas to increase the cooling rate of a falling drop. Helium gas would seem to be a good candidate since this gas is inert, readily obtained, and has a high value of thermal conductivity relative to other gases. The purpose of this report is to provide a good estimate of the effect that the presence of helium gas in the drop tube would have upon the rate at which heat will be lost by a falling drop. This effect should be evidenced by a change in the time required to solidify a falling drop and by a change in the supercooling rate of the molten drop if the drop does not begin to solidify after release.

THEORY

The rate at which a heated sphere will lose heat energy during free fall in the drop tube filled with a gas is given by Zemansky [1] as:

$$\frac{dQ}{dt} = -\epsilon A\sigma(T^4 - T_o^{\,4}) - hA(T - T_o) \quad . \tag{1}$$

The first term on the right of equation (1) is due to radiation loss and is dependent on the emissivity of the material, ϵ; the surface area of the sphere, A; the

Boltzman's constant, σ; as well as the fourth power of the sphere temperature, T, and the ambient temperature, T_o. The second term on the right of equation (1) is due to a combination of conduction and forced convection and is dependent upon A, T, T_o, and the heat transfer coefficient, h. The amount of heat a drop must lose for complete solidification will be equal to the product of the mass, m, of the drop and the latent heat of fusion, H_f, of the drop material. If during solidification the temperature of the drop is assumed to be constant at the melting point, T_m, of the drop material, then equation (1) can be reduced to

$$t_s = mH_f / [\epsilon A\sigma(T_m^4 - T_o^4) + hA(T - T_o)] \quad . \tag{2}$$

The solidification time, t_s, of equation (2) will be the time required to completely solidify the drop. If the drop tube is evacuated, h goes to zero and the solidification time becomes

$$t_s = mH_f / [\epsilon A\sigma(T_m^4 - T_o^4)] \quad . \tag{3}$$

A comparison of equations (2) and (3) for a given drop shows the effect that the presence of a gas environment will have upon solidification time for a falling drop.

If the drop does not begin to solidify after release, it will supercool. In this event, Q of equation (1) will be equal to the product of the mass, m, of the drop and the specific heat, C, of the molten material. Equation (1) will then reduce to the differential equation

$$\frac{dT}{dt} = -K_o(T^4 - T_o^4) - H(T - T_o) \tag{4}$$

where

$$K_o = \frac{\epsilon A \sigma}{mC} \qquad \text{and} \qquad H = \frac{hA}{mC} \quad .$$

A solution to equation (4) has been provided by Katz and Wills [2] by assuming that the drop material parameters do not change during the time of free-fall, that the drop is spherical, and that the drop is small enough so that there is no thermal lag between the sphere's surface and center. The solution to equation (4) is then

$$\begin{aligned} t = \frac{1}{K_o[4(T_o)^3 + H_o]} \Bigg[& -\ell n(T - T_o) + \left(\frac{[(C_1)^2 - 4C_1 + 22]}{3[(C_1)^2 + 2]} \right) \ell n \left[T + \frac{T_o}{3}(C_1 + 1) \right] \\ & + \frac{(C_1 + 4)(C_1 - 2)}{3[(C_1)^2 + 2]} \ell n \left\{ \left[T - \frac{T_o}{3}\left(\frac{C_1}{2} - 1 \right) \right]^2 + \frac{(T_o)^2 (C_2)^2}{12} \right\} \\ & + \left(\frac{4(C_1 + 1)(C_1 + 4)}{\sqrt{3}\, C_2[(C_1)^2 + 2]} \right) \tan^{-1} \left(\frac{T - \frac{T_o}{3}\left(\frac{C_1}{2} - 1 \right)}{\frac{T_o C_2}{2\sqrt{3}}} \right) \Bigg] + \text{const.} \end{aligned} \tag{5a}$$

where $H_o = H/K_o$ and C_1 and C_2 are given by

$$\begin{aligned} C_1 &= \sqrt[3]{\frac{K_1 + K_2}{2}} + \sqrt[3]{\frac{K_1 - K_2}{2}} \\ C_2 &= \sqrt[3]{\frac{K_1 + K_2}{2}} - \sqrt[3]{\frac{K_1 - K_2}{2}} \end{aligned} \tag{5b}$$

where

$$K_1 = 20 + 27H_o/T_o^3$$

and (5c)

$$K_2 = \sqrt{K_1^2 + 32} \quad .$$

The time t_c of equation (5a) is the time required for a drop to supercool from the initial temperature, T_i, to the final temperature, T_f. The constant is evaluated at $t = 0$ and $T = T_i$. If the drop tube is evacuated, h goes to zero and equation (5) reduces to

$$t_c = \frac{1}{4K_o T_o^3}\left[\ln\left(\frac{T + T_o}{T - T_o}\right) + 2\tan^{-1}\left(\frac{T}{T_o}\right)\right] + \text{const.} \qquad (6)$$

where the constant is again evaluated at $t = 0$ and $T = T_i$. The effect that the presence of helium will have upon the supercooling rates of falling drops can be seen from a comparison of the supercooling times of equations (5) and (6).

ASSUMPTIONS

To calculate the cooling and solidification times for a drop, certain assumptions must be made. As previously stated, the drop is assumed to be spherical, have no thermal lag between surface and center, and have constant material parameters during the time of cooling. The validity of assuming the drop to be isothermal during cooling is addressed in the Appendix.

An empirically determined heat transfer coefficient, h, for a sphere in a gas flow with a Reynolds number between 17 and 17 000 is recommended by McAdams [3] as

$$h = 0.37\,\frac{K_f}{D}\left(\frac{U_\infty D}{V_f}\right)^{0.6} \tag{7}$$

where D is the diameter of the drop, U_∞ is the free stream velocity of the gas flow, and K_f and V_f are the thermal conductivity and kinematic viscosity of the gas at the thin film temperature, T_R. Although the drop is accelerated through the gas by gravity when falling in the drop tube, for ease of calculation, the velocity of the drop relative to the gas will be assumed to be a constant 12 m/s, which is the average velocity of a drop falling the length of the drop tube (30.5 m). Although, according to Loeb [4], the thermal conductivity and viscosity of the gas are pressure independent for the pressures to be considered in this report (i.e., 1 to 760 Torr), these parameters are evaluated at T_R since they are temperature dependent. Since equation (7) is dependent on the kinetic viscosity which is the quotient of the viscosity and density of the gas, the heat transfer coefficient h will be pressure dependent. Holman [5] gives the thin film temperature as the average of the surface temperature, T_s, of the sphere and the free stream temperature, T_∞, of the gas

$$T_R = \frac{T_s + T_\infty}{2} \quad . \tag{8}$$

Since the change with temperature of K_f and V_f is slow, T_s can be assumed to be the average of T_i and T_f, and equation (8) becomes

$$T_R = \frac{T_i + T_f + 2T_o}{2} \tag{9}$$

by also assuming $T_\infty = T_o$.

Although the previously described assumptions introduce some error into equations (2), (3), (5), and (6), these equations should still provide a good estimate of the supercooling and solidification times for a drop in both a helium and vacuum environment.

RESULTS

Niobium, copper, and lead have been selected as typical materials because of the wide range of material parameters, especially the range of melting points, characteristic of these materials. The range of melting points of these materials is very important considering the fact that the drop is at or slightly above the melting point when the drop is released. The characteristic parameters for the selected materials are presented in Table 1.

The solidification times, as given by equation (3) for radiation cooling only, for niobium, copper, and lead drops of 3, 5, 7, and 10 mm diameter are presented in Table 2. The solidification times of Table 2 reflect the fourth power of T_m dependency of t_s of equation (3) as evidenced by the marked increase of the required times for complete solidification for the drops of the materials with the lower melting points. Also from Table 2 it should be noted that radiation cooling is not sufficient to solidify drops of copper or lead within the 2.5 s time limit of the drop tube.

The solidification times for niobium, copper, and lead drops falling through a helium gas, as given by equation (2), are presented in Figures 1, 2, and 3 as a function of helium gas pressure. The helium pressure dependency of the solidification time enters equation (2) by way of the kinematic viscosity of the gas from the heat transfer coefficient. As can be seen in Figure 1, the effect of helium cooling on the solidification times for niobium drops would be sufficient to allow solidification of 7 and 10 mm diameter niobium drops in the drop tube. The cooling effect of helium is even more pronounced in the curves of Figures 2 and 3, which show that copper drops of up to 5 mm in diameter and lead drops of diameters up to 7 mm could be completely solidified if the drop tube were filled with helium gas at near standard atmospheric pressure. It should be noted from Table 2 that none of the copper and lead drops discussed would solidify in the drop tube if it were evacuated. A comparison of Figures 1, 2, and 3 reveals that the cooling of the lower melting point copper and lead drops is less dependent on radiation than on heat transfer to the helium, as evidenced by the rapid decrease in drop solidification times as helium gas pressure increases. To illustrate the fact further, the percentage of heat loss at T_m due to the radiation term of equation (1) is presented in Figure 4 for the three materials as a function of helium pressure. Figure 4 clearly shows that radiation cooling is much less a factor for lead and copper drops than for the higher melting point of niobium drops and, therefore, agrees with Figures 1, 2, and 3. Since radiation loss adds very little to the cooling of copper and lead drops near the standard atmospheric pressure of helium, the solidification times for these drops should depend less on the melting temperature than on other material parameters. Therefore, the fact that larger lead drops can be solidified in the drop tube than copper drops (as can be seen by comparing Figures 2 and 3) can be attributed to the relatively low heat of fusion of lead.

It should be noted that the heat transfer coefficient used in the calculations presented in Figures 1 through 4 is not accurate for Reynolds numbers below 17. The regions of the curves in Figures 1 through 4 for which the pressure of the helium gas is such that the Reynolds number of the gas flow past the falling drop is near or below 17 are shown as dotted. These regions are presented since they should still provide a good estimate of the effect the helium will have upon drop solidification times for this range of pressures.

Also of interest is the rate at which a drop will supercool if it does not begin to solidify immediately after being released. The times required for 3, 5, 7, and 10 mm diameter drops of niobium to supercool a given amount are given in Figures 5 through 8. These are times for niobium drops falling through various pressures of helium gas, as given by equation (5), and times for drops falling through a vacuum, as given by equation (6). Corresponding supercooling curves for copper are given in Figures 9 through 12, and for lead in Figures 13 through 16. Although the drops are not expected to supercool more than a few hundred degrees, the curves of Figures 5 through 12 have been extended to 1000 deg of supercooling to further illustrate the effect of the presence of helium gas on the supercooling rates of niobium and copper drops.

From Figures 5 through 9 it can be seen that the niobium drops will supercool very quickly. Before a 3 mm niobium drop has fallen 1 m (0.45 s) in vacuum, it could have supercooled as much as 250 deg. Within the 2.5 s limit of the drop tube, a 3 mm diameter drop of niobium can possibly supercool down to the homogeneous nucleation temperature which is given by Miller [8] as approximately 700 K. Helium gas cooling would increase the supercooling rate by as much as 80 percent near the atmospheric pressure of helium. This increased cooling rate could also be expected for the 5, 7, and 10 mm diameter niobium drops. For the copper drops discussed in Figures 10 through 13, the supercooling rates are much slower. A 3 mm diameter copper drop could possibly supercool approximately 50 deg in the evacuated drop tube. However, with helium cooling, the same size drop could supercool to the homogeneous nucleation temperature of 240 K after only 0.54 s of free fall. Figures 13 through 16 show an even greater difference between the vacuum and helium curves for lead drops. If the drop tube were evacuated, a 3 mm diameter lead drop could not be expected to supercool more than a few degrees. Likewise, with helium present in the drop tube at standard atmospheric pressure, the same diameter lead drop could possibly supercool to the homogeneous nucleation temperature of 80 K after only 0.34 s of free fall. By comparing the supercooling curves of niobium, copper, and lead, it can be seen that cooling is more dependent upon heat transfer to the helium gas than on radiation loss for the lower melting temperature materials. This result could have been predicted from Figure 4.

CONCLUSION

Since drops of sample materials are released in the drop tube at or slightly above their melting temperature, the effect of helium gas cooling will be greater for drops of materials with low melting points. By the use of helium of less than 1 atm of pressure, solidified drops of significant size (i.e., ≥ 3 mm) can be obtained for materials which could not be solidified in useful sizes in the 2.5 s time limit for the evacuated tube. Also, if they do not begin to solidify immediately upon release, the rate at which the molten drops supercool will be considerably affected. The presence of helium can increase the supercooling rate enough to supercool drops to the homogeneous nucleation point. Thus, the present drop tube facility can be used to study nucleation and the solidified structure of drops of metals and alloys supercooled the maximum extent possible. In conclusion, it can be stated from the foregoing estimates that the presence of helium gas in the drop tube would have a significant effect on both time required to solidify drops of sample materials and on the supercooling rates of the drops. Thus, the use of helium gas could greatly extend the versatility and usefulness of the MSFC space processing drop tube to carry out containerless low-gravity supercooling and solidification experiments.

TABLE 1. CHARACTERISTIC PHYSICAL PROPERTIES OF SAMPLE MATERIALS [6,7]

	Niobium	Copper	Lead
Density (g/cm^3)	8.60	8.96	11.34
Melting Point (K)	2741	1357	600
Heat Capacity (J/g-K)	0.268	0.385	0.126
Heat of Fusion (J/g)	284.6	211.8	26.4
Emissivity (Near T_m)	0.25	0.16	0.075

TABLE 2. SOLIDIFICATION TIMES IN SECONDS FOR DROPS FALLING IN A VACUUM

Diameter (mm)	Niobium	Copper	Lead
3	1.5	30.9	288.3
5	2.5	51.5	480.4
7	3.6	72.1	672.6
10	5.1	103.0	960.9

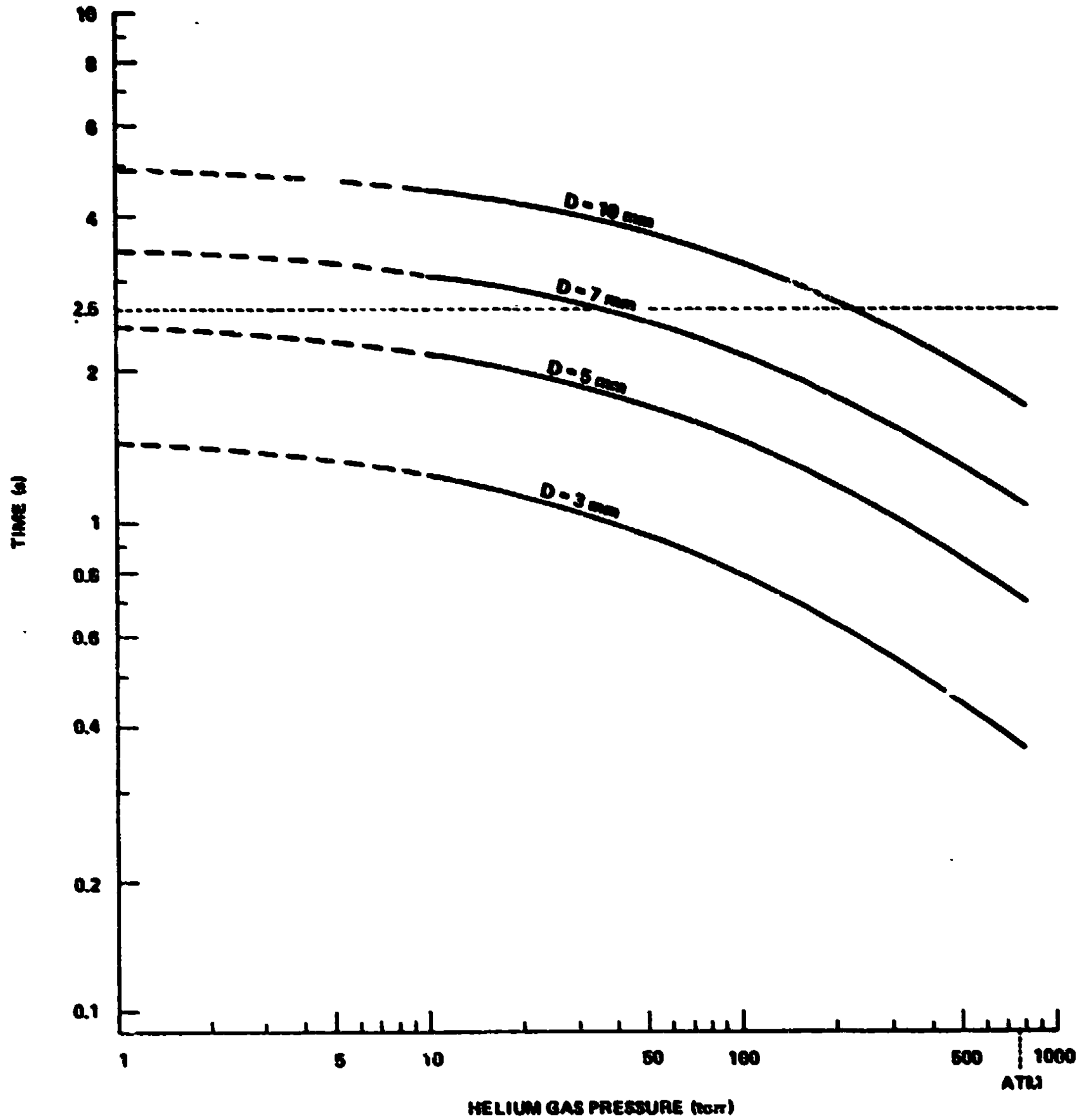

Figure 1. Time required to completely solidify niobium drops of 3, 5, 7, and 10 mm diameter as a function of helium gas pressure.

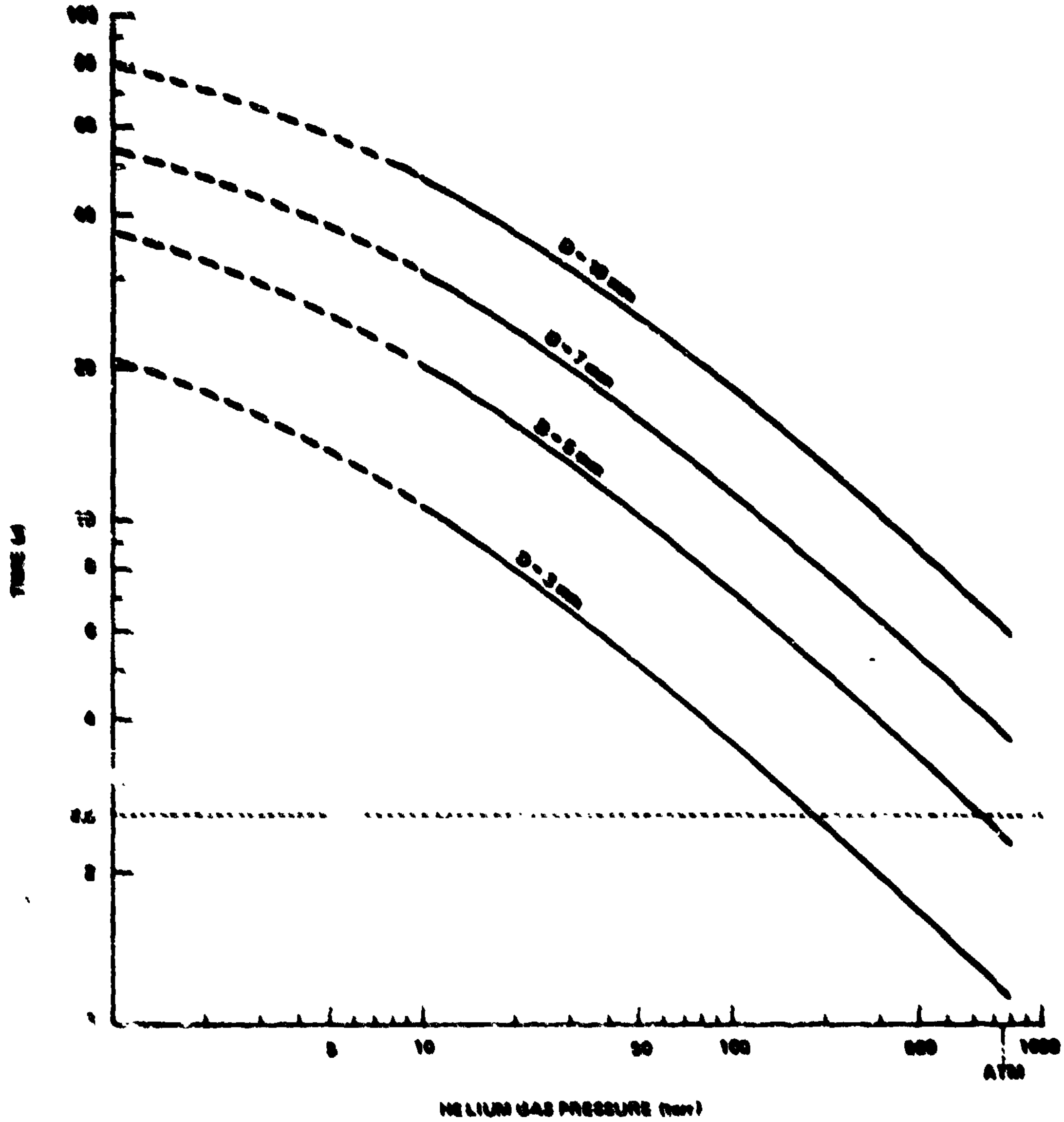

Figure 2. Time required to completely solidify copper drops of 3, 5, 7, and 10 mm diameter as a function of helium gas pressure.

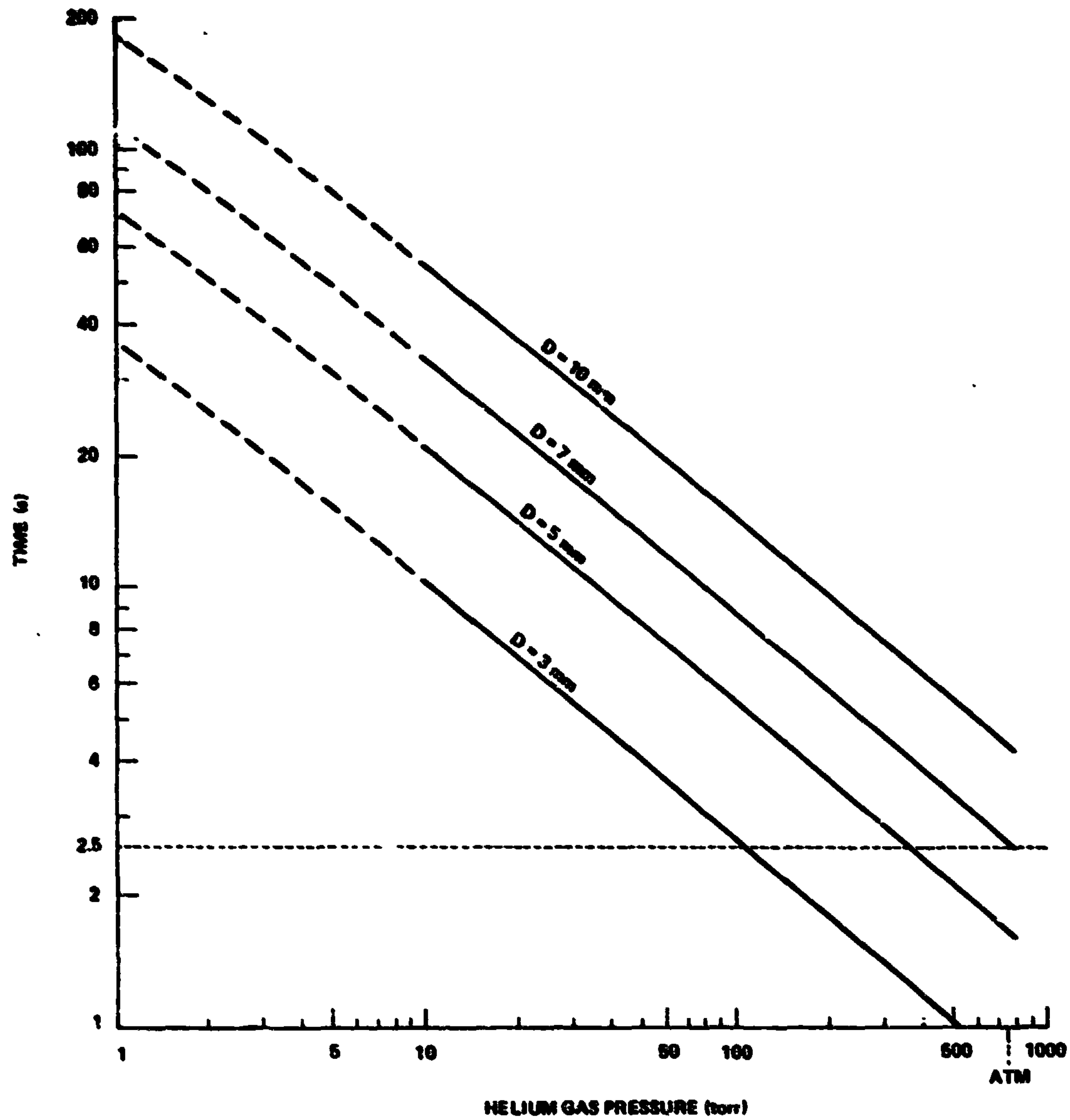

Figure 3. Time required to completely solidify lead drops of 3, 5, 7, and 10 mm diameter as a function of helium gas pressure.

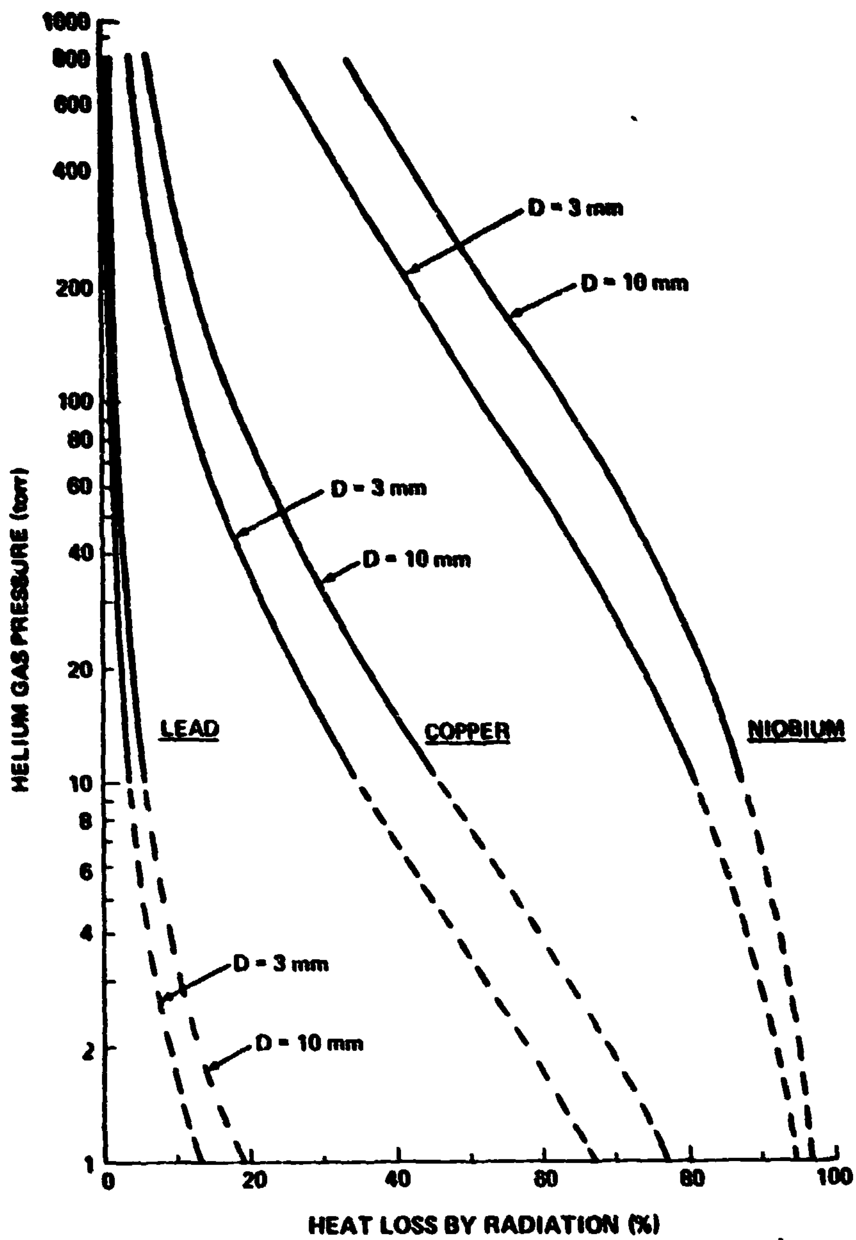

Figure 4. Relative heat lose by radiation for drops falling in helium gas for niobium, copper, and lead spheres of 3 and 10 mm diameter.

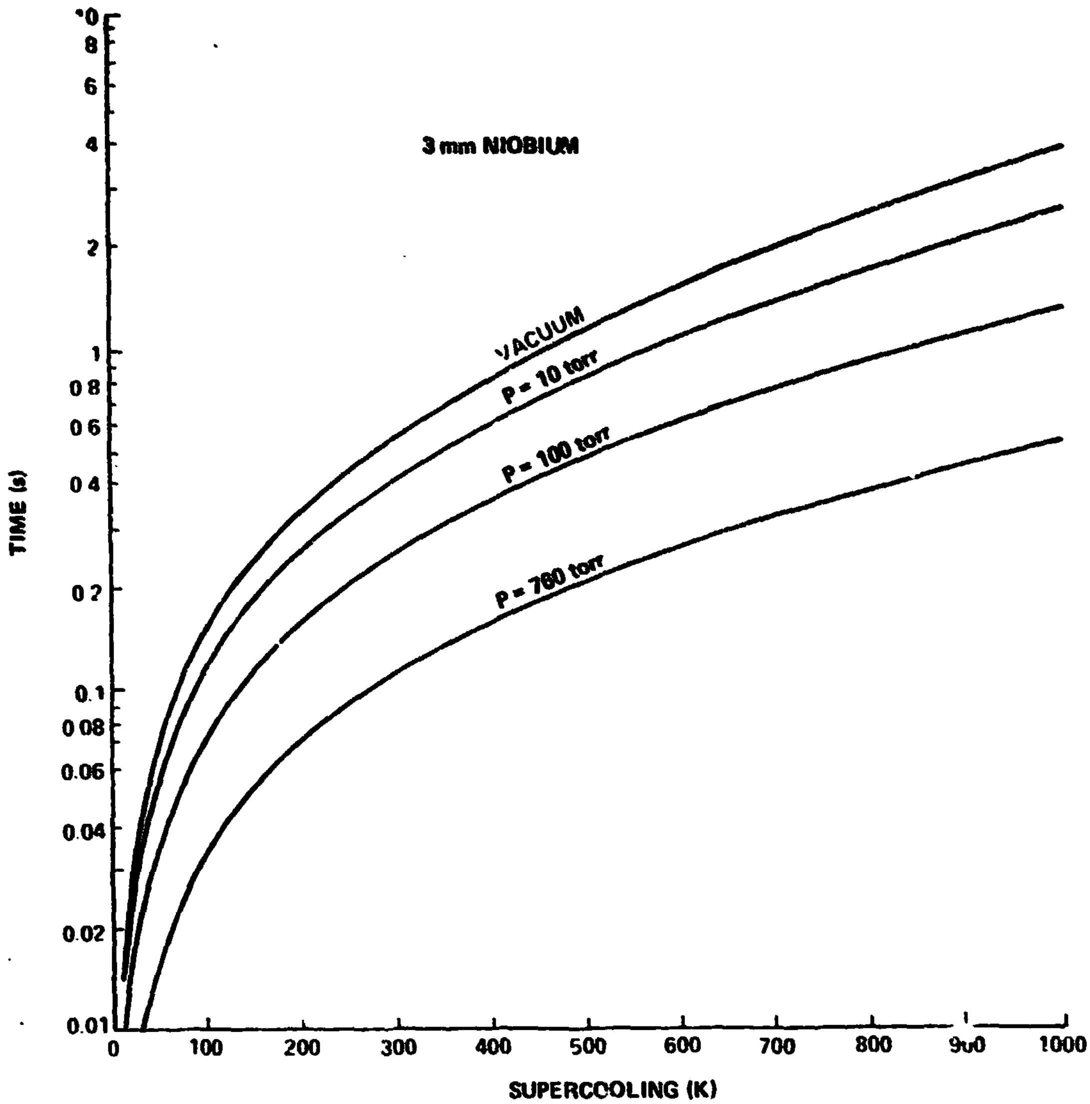

Figure 5. The amount of supercooling for a 3 mm diameter niobium sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

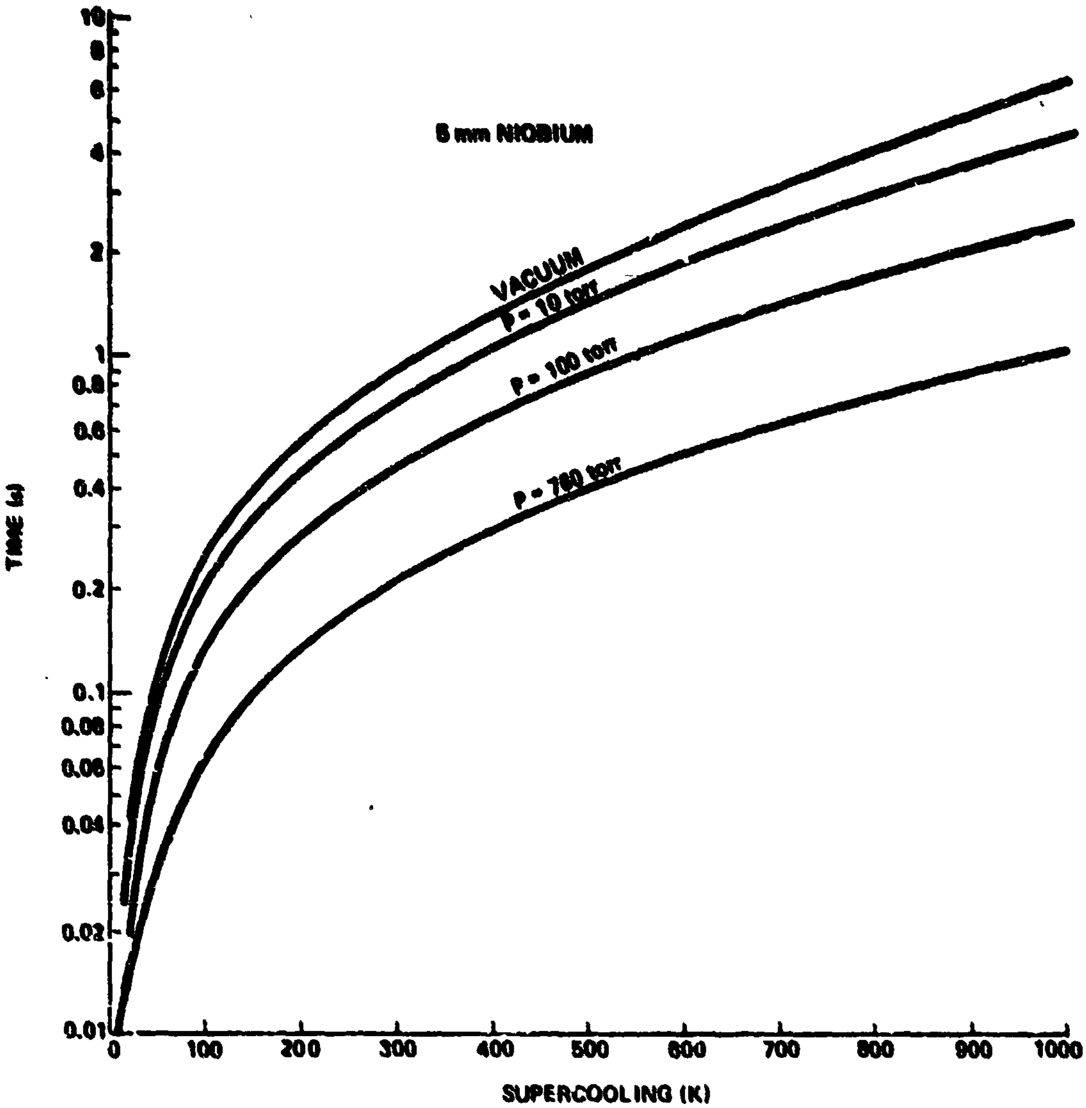

Figure 6. The amount of supercooling for a 5 mm diameter niobium sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

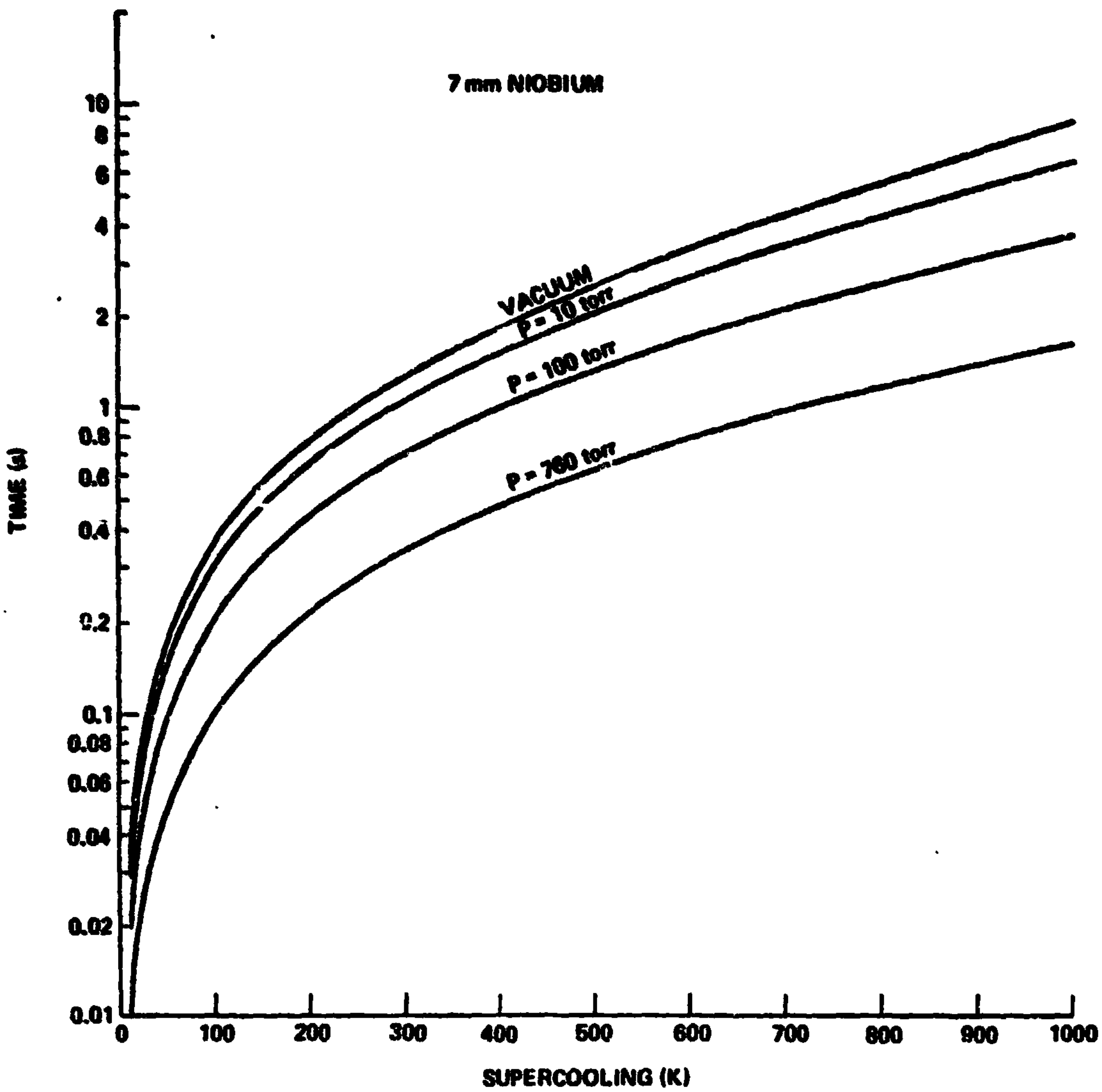

Figure 7. The amount of supercooling for a 7 mm diameter niobium sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

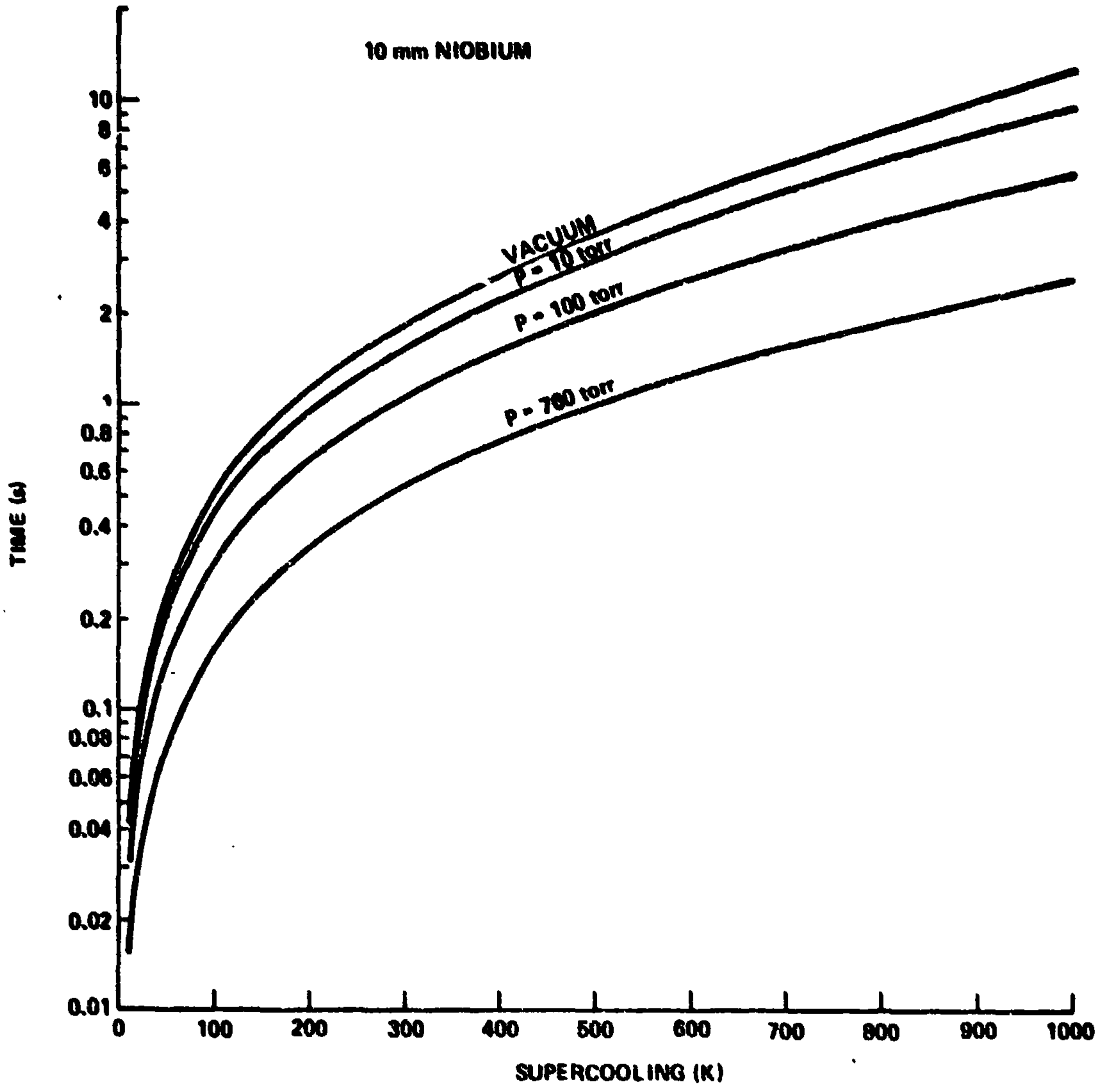

Figure 8. The amount of supercooling for a 10 mm diameter niobium sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

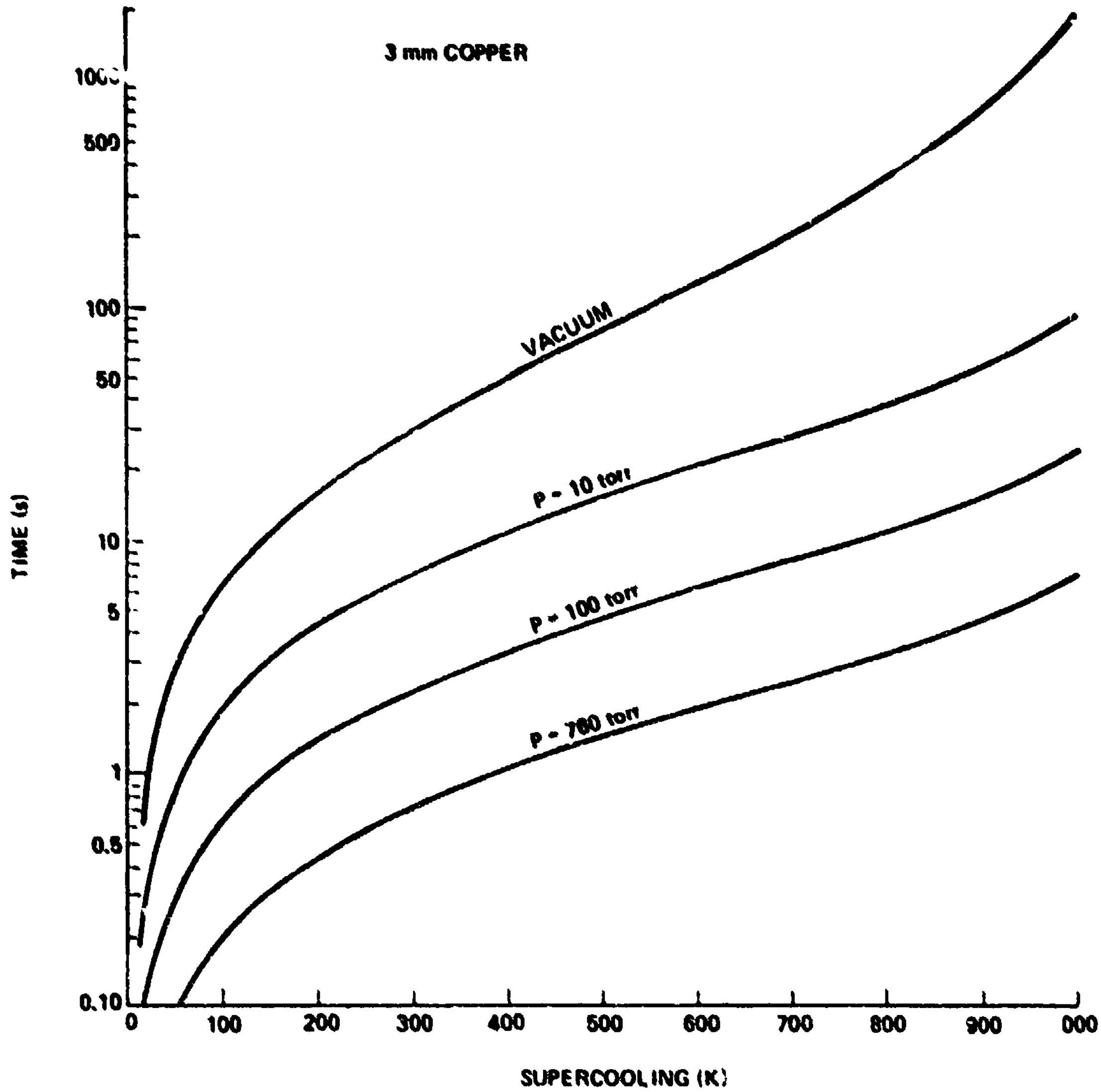

Figure 9. The amount of supercooling for a 3 mm diameter copper sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

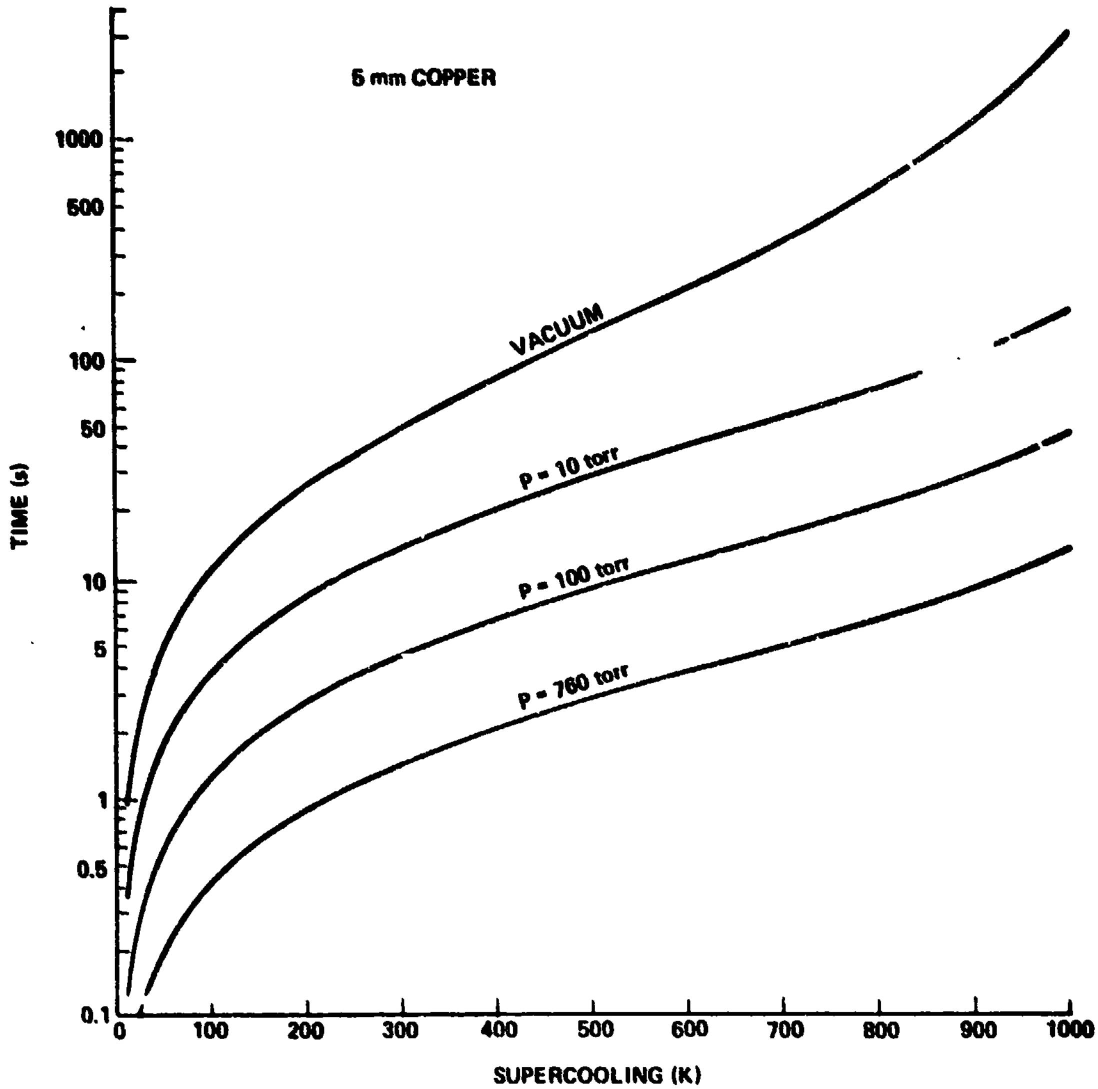

Figure 10. The amount of supercooling for a 5 mm diameter copper sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

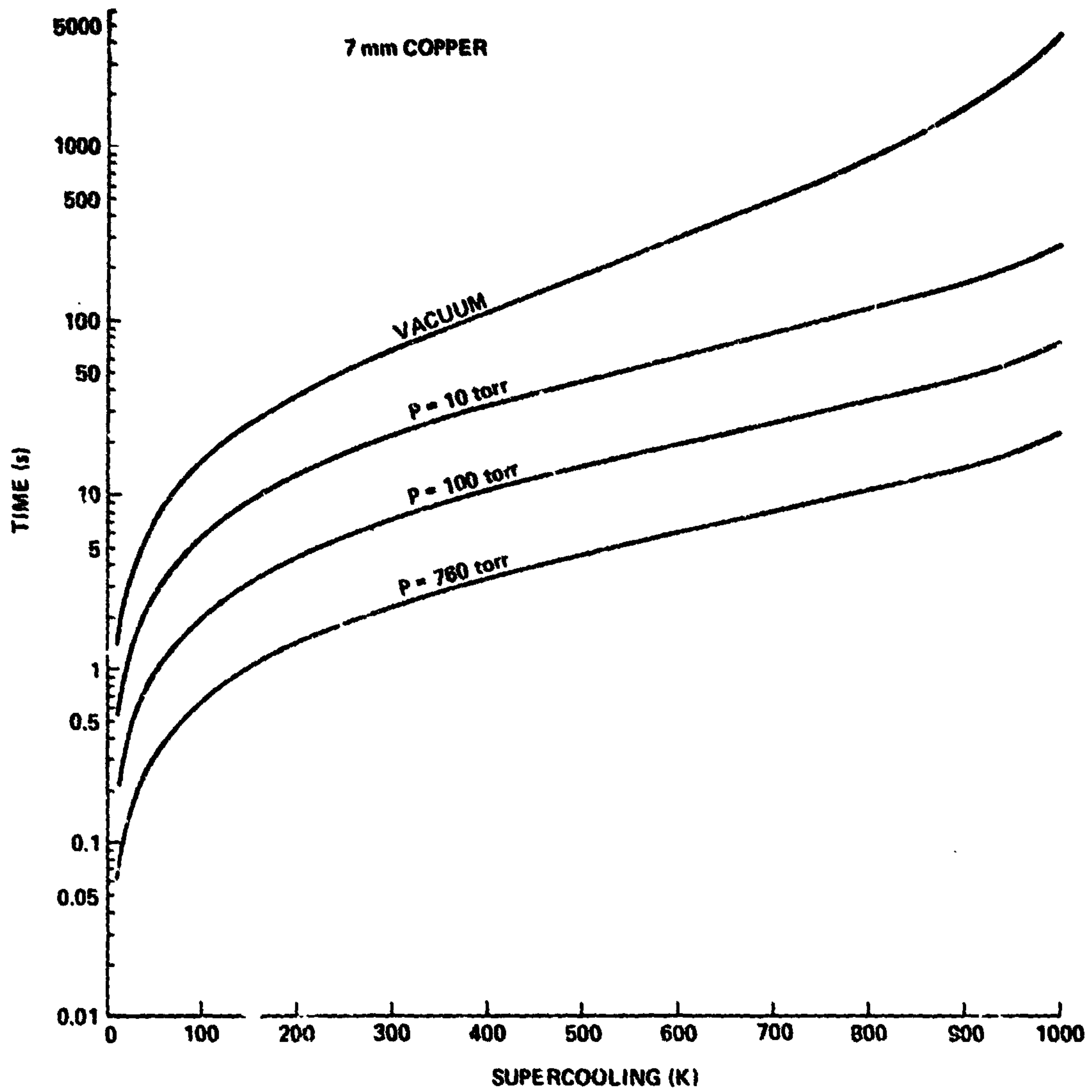

Figure 11. The amount of supercooling for a 7 mm diameter copper sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

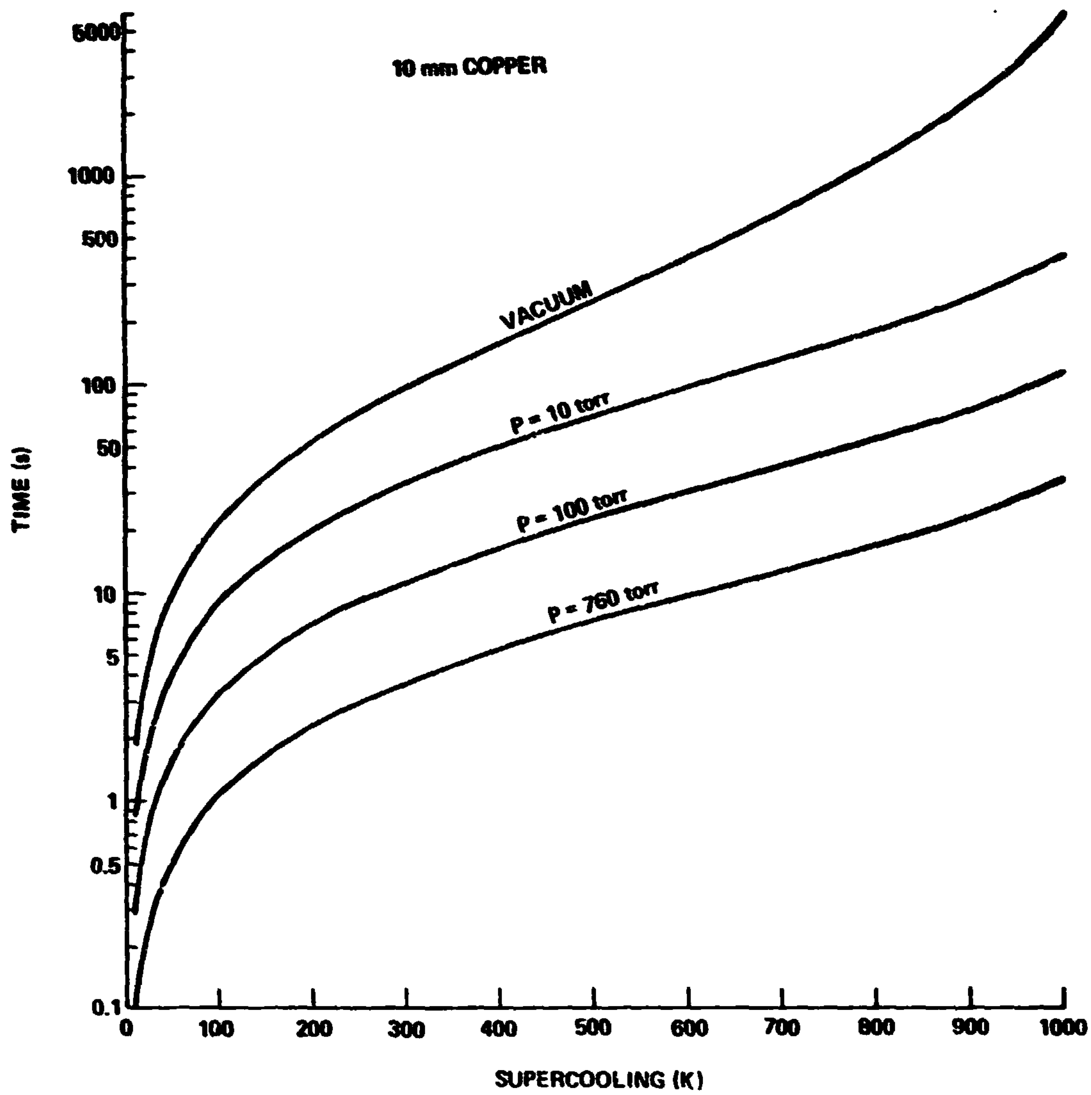

Figure 12. The amount of supercooling for a 10 mm diameter copper sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

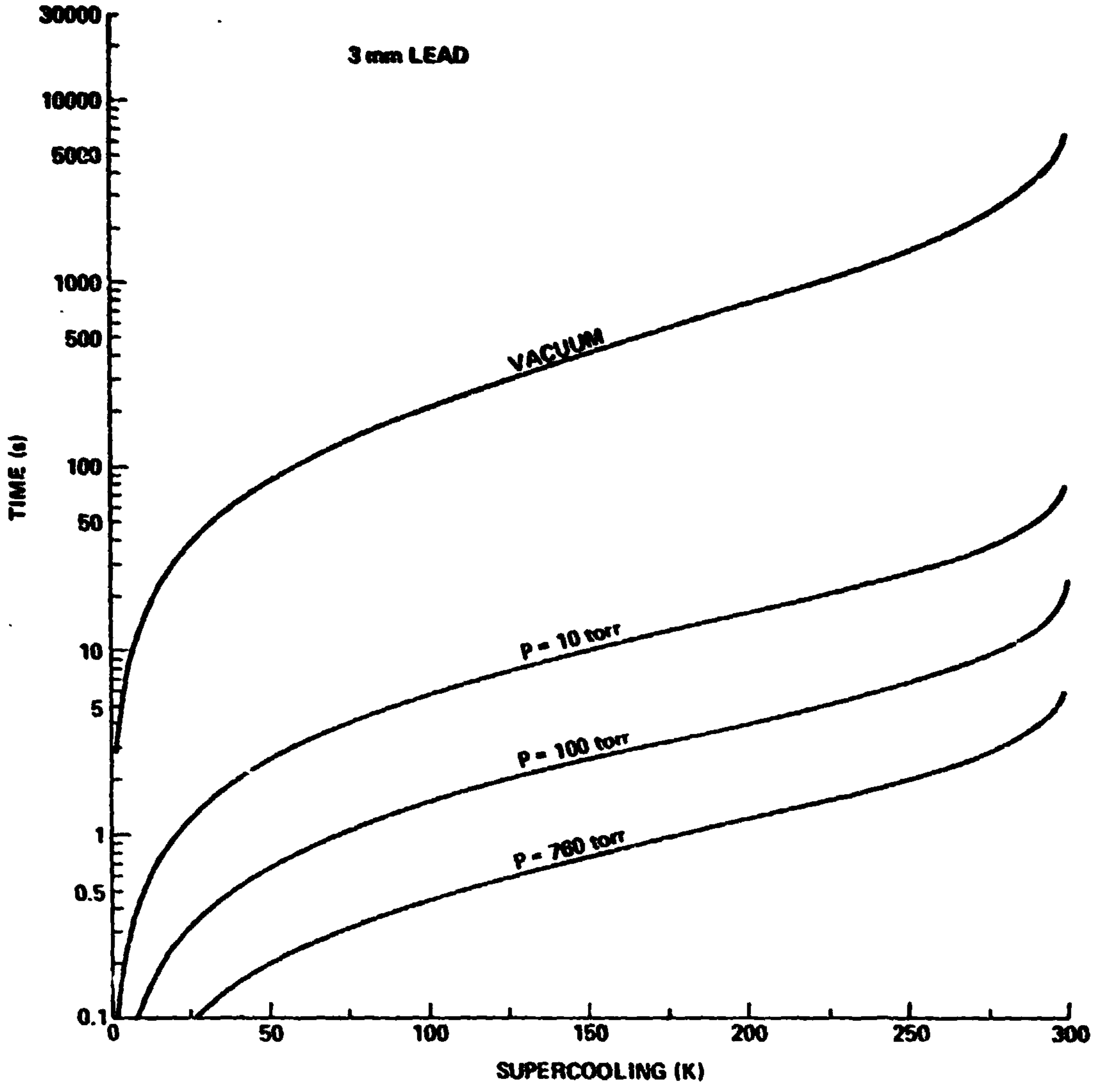

Figure 13. The amount of supercooling for a 3 mm diameter lead sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

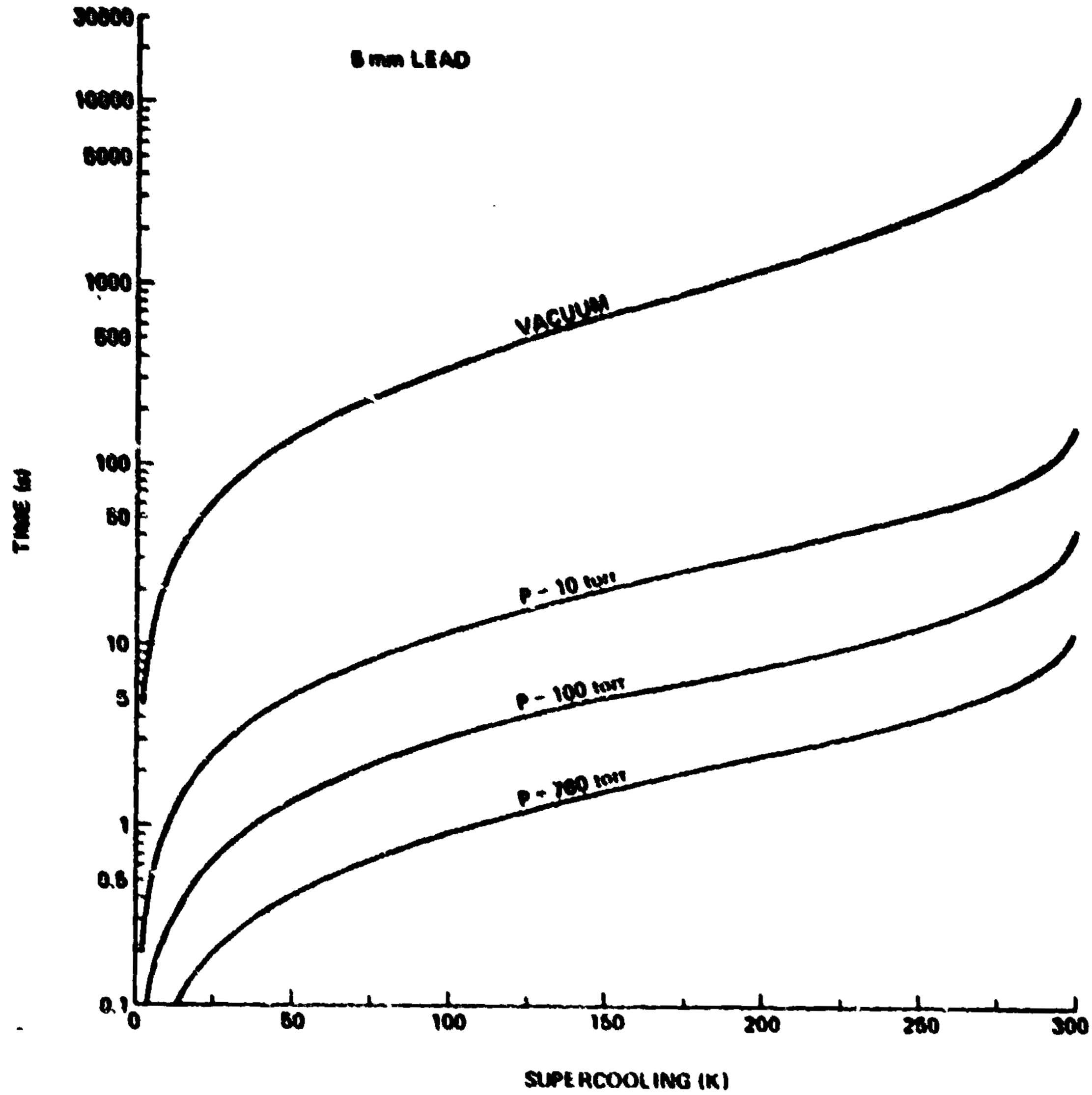

Figure 14. The amount of supercooling for a 5 mm diameter lead sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

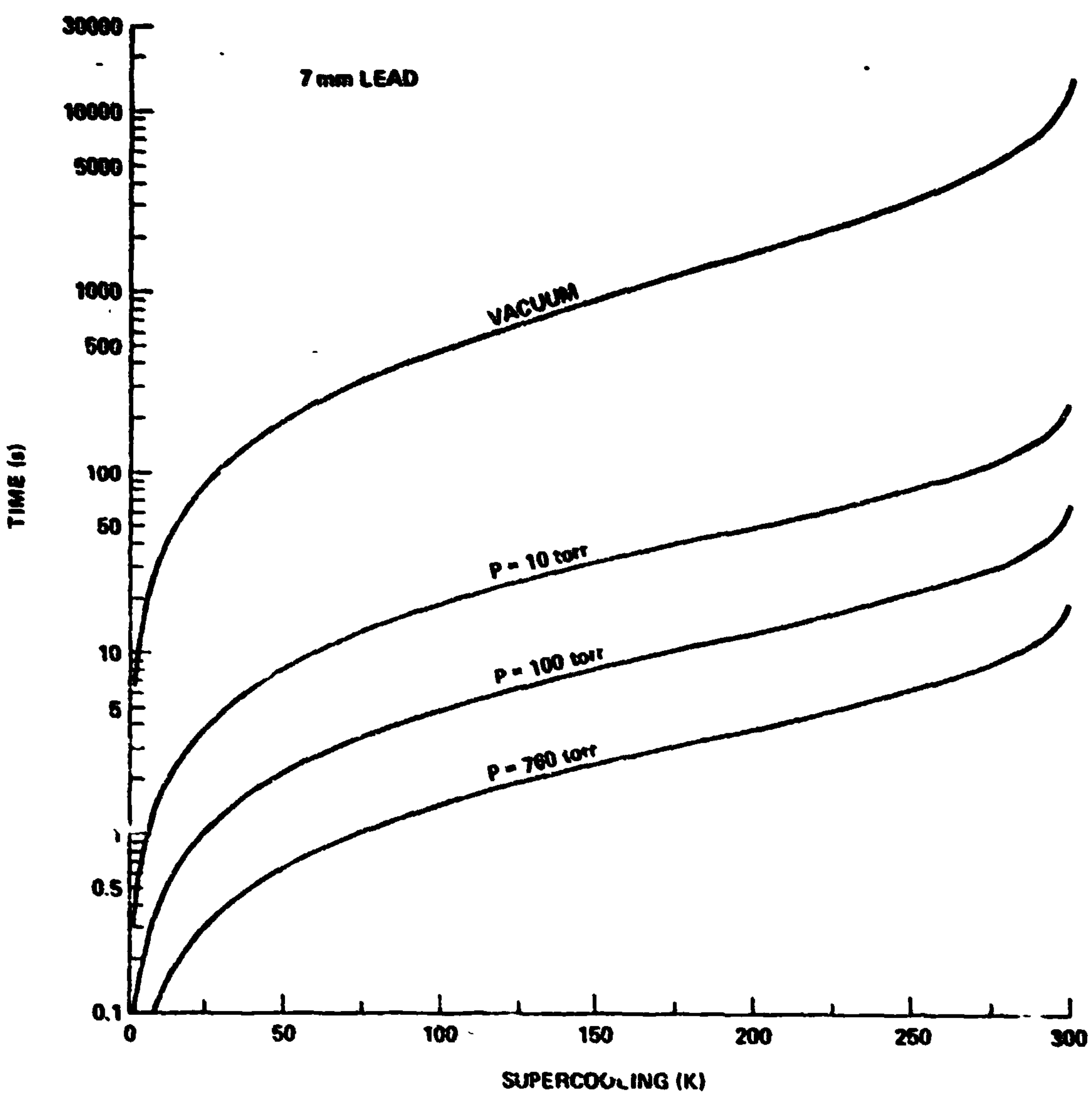

Figure 15. The amount of supercooling for a 7 mm diameter lead sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

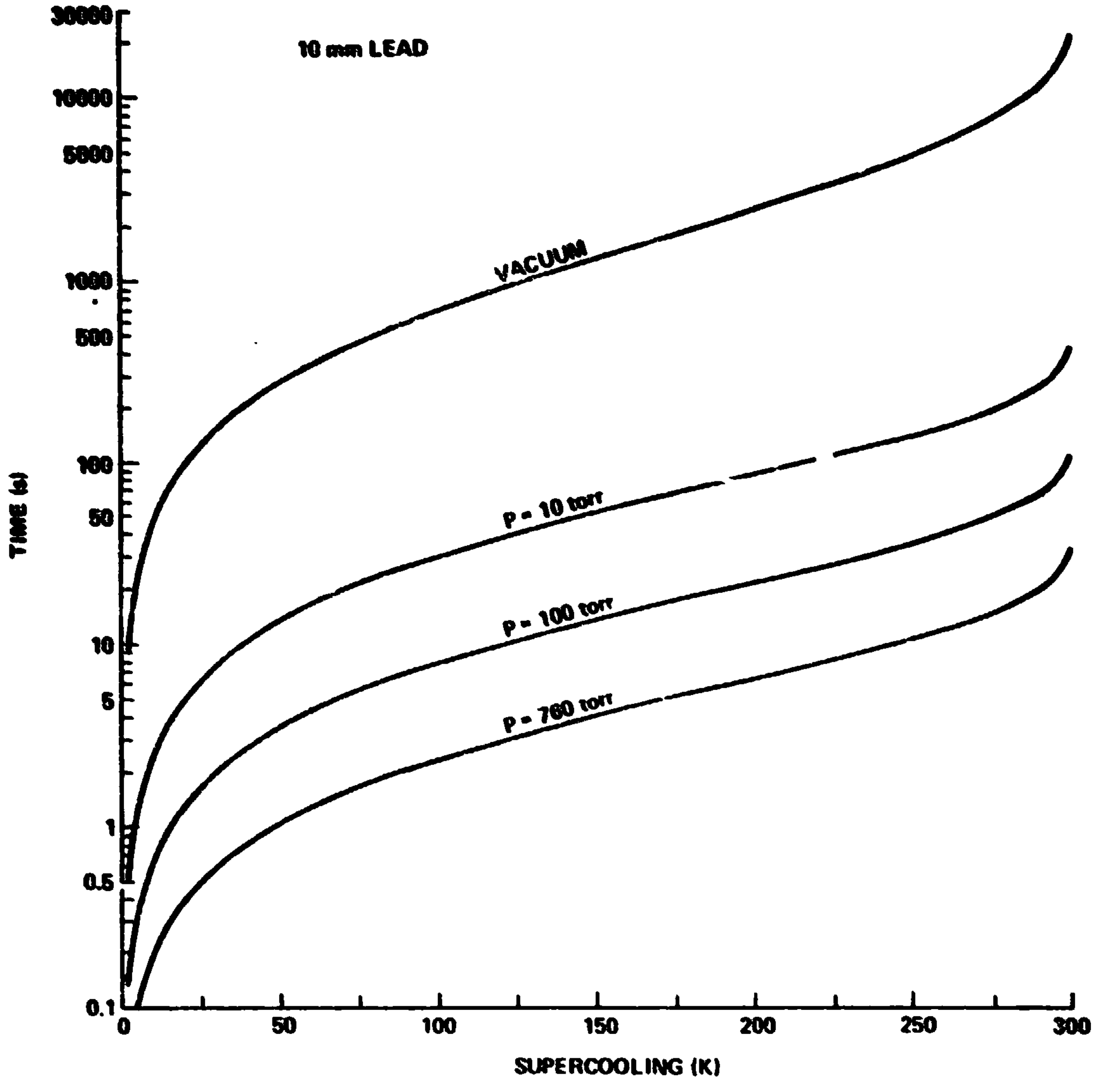

Figure 16. The amount of supercooling for a 10 mm diameter lead sphere plotted as a function of time before nucleation. (The calculations are for a molten drop falling in a vacuum or helium gas at different pressures.)

APPENDIX

VALIDITY OF ISOTHERMAL COOLING

The question of the validity of the isothermal approximation, for the purpose of this report, can be answered by considering the temperature distribution within the cooling sphere. The radial heat flow within the sphere must satisfy Poisson's equation [9].

$$\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r} = \alpha \frac{\partial T}{\partial r} \tag{A-1}$$

where $\alpha = K/\rho C$ is the thermal diffusivity of the material and K denotes the material's thermal conductivity and ρ the material's density. The boundary conditions would be:

$$T = T_i \qquad \text{at } t = 0 \tag{A-2}$$

and at the surface

$$\frac{\partial T}{\partial r} = -\frac{h(T - T_o) + \epsilon\sigma(T^4 - T_o^4)}{K} \qquad \text{for } t \geq 0. \tag{A-3}$$

Since equation (A-3) is a time-dependent, nonlinear boundary condition, equation (A-1) has no closed form solution. However, equation (A-3) can be linearized by using a Taylor's series expansion which yields

$$\frac{\partial T}{\partial r} = -\frac{h(T_1 - T_o) + \epsilon\sigma(T_1^4 - T_o^4)}{K} - \frac{h + 4\epsilon\sigma T_i^3}{K}(T - T_1) \tag{A-4}$$

where T_1 is a temperature selected near the mid-range between T_i and T_f. T_1 can be chosen so as to minimize the difference between equations (A-3) and (A-4) over the desired temperature range. Using boundary conditions (A-2) and (A-4), Carslaw and Jaeger [9] offer a series solution to equation (A-1) as:

$$\frac{\nu}{V} = \frac{2J}{r} \sum_{N=1}^{\infty} e^{-\alpha\beta_N^2 t} \left[\frac{a^2\beta_N^2 + (aJ-1)^2}{\beta_N^2[a^2\beta_N^2 + aJ(aJ-1)]} \right] \sin(a\beta_N) \sin(r\beta_N) \tag{A-5}$$

where a is the radius of the sphere and

$$\nu = T - T_1 + T_2$$

$$V = T_i - T_1 + T_2$$

with

$$T_2 = \frac{h(T_1 - T_o) + \epsilon\sigma(T_1^4 - T_o^4)}{KJ}$$

and

$$J = \frac{h + 4\epsilon\sigma T_1^3}{K}$$

and where β_N is given as the roots of

$$a\beta_N \cot(a\beta_N) + aJ - 1 = 0 \ .$$

The temperature difference between the drop's surface and center, defined as ΔT, can now be found by using equation (A-5) and the values of K [10] and α as given in Table A-1. The results show that 3 mm diameter copper and lead drops will have a ΔT of less than 4 K even with the drop tube filled to one atmospheric pressure of helium. These ΔT's for copper and lead correspond to less than 1 percent of the melting temperature. For a 3 mm niobium drop falling through one atmospheric pressure of helium, the expected ΔT would be less than 45 K, or approximately 1.6 percent of the melting temperature. A niobium drop of the same size cooled by radiation would have a ΔT of only 14 K. For convenience, the ratios of the maximum ΔT to the melting temperature, T_m, for 3 mm diameter drops of the three materials, cooled with helium gas, are also listed in Table A-1.

Since the temperature difference from center to surface for drops of niobium, copper, and lead is such a small percent of the melting temperature, the assumption that the drops will cool isothermally will not appreciably affect the results as presented in this report.

TABLE A-1. TEMPERATURE DIFFERENCES

	Niobium	Copper	Lead
K(J/S-ca-K)	0.52	3.42	0.34
α(cm^2/S)	0.11	0.99	0.24
$\frac{\Delta T\ max}{TM}$	0.016	0.002	0.007

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APPROVAL

RADIATIVE AND GAS COOLING OF FALLING MOLTEN DROPS

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The information in this report has been reviewed for technical content. Review of any information concerning Department of Defense or nuclear energy activities or programs has been made by the MSFC Security Classification Officer. This report, in its entirety, has been determined to be unclassified.

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