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FILM BOILING OF MERCURY DROPLETS

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Abstract

Vaporization times of mercury droplets in Leidenfrost film boiling on a flat horizontal plate are measured in an air atmosphere. Extreme care was used to prevent large amplitude droplet vibrations and surface wetting; therefore, these data can be compared to film boiling theory. For these data, diffusion from the upper surface of the drop is a dominant mode of mass transfer from the drop. A closed-form analytical film boiling theory is developed to account for the diffusive evaporation. Reasonable agreement between data and theory is seen.

Introduction

Up to the present time, in the study of film boiling of liquid droplets, most experiments have considered fluids such as water, hydrocarbons, or cryogens. References 1 to 6 provide a comprehensive summary of past work plus a discussion of the phenomenon of droplet film boiling, often called Leidenfrost boiling. Recent interest in liquid-metal fast-breeder reactors and concern for their safe operation has lead to the need for film boiling data for liquid metals. In this paper, the authors' report vaporization times of mercury droplets in film boiling on a flat horizontal plate in an air atmosphere and correlate the data. The heat transfer coefficient, plate temperatures, etc, are also reported.

Poppendiek, et al. have reported some film boiling data of mercury droplets.[7,8] They reported that large amplitude droplet vibrations and interaction of the droplet with the surface occurred during the vaporization process. The authors report the vaporization time of only a single size droplet and unfortunately give only an average droplet radius rather than an exact volume measurement.

To eliminate the problems described by Poppendiek, et al. only very small mercury droplet volumes (0.3? cm in diam. or less) were used herein to obtain stable film boiling with a minimum of droplet vibration and surface wetting.[7]

In an air atmosphere in the volume range considered, diffusion from the upper surface of the drop is a dominant mode of mass transfer as shown by an evaluation of the dimensionless groups developed herein and shown by some experimental data from Ref. 8. The previously-developed closed form theories for the heat transfer coefficient and vaporization times either neglected or considered

diffusion from the upper surface of the drop as a small part of the mass transfer process. As a result, these theories, such as developed in Refs. 1 or 10, can not be used to correlate the data. The numerical scheme presented in Ref. 2 could be applied. However, since a closed-form analytical theory is convenient, a closed-form solution was sought and is presented herein for the heat transfer coefficient and vaporization time of droplets in diffusion-dominated film boiling.

The analysis is limited to two-component systems involving a pure single-component liquid with small solubilities for the gas and a single-component gas of high purity (with the exception of air for which measured values of the diffusion coefficient exist).

Experimental Apparatus and Procedure

The experimental equipment consisted basically of a heated flat surface insulated on its sides. The initial droplet vaporization experiments on the hot surface were conducted in a cylindrical tank 60 cm in diameter and 60 cm long with a helium atmosphere. This apparatus was the same one used in Ref. 8 to measure the effects of the environmental diffusivity on the vaporization times of the drops. The first data point taken for a drop in a dry environment was always repeated at the end of a data set to check for repeatability.

Although the remote handling was inherently a safer way to conduct the vaporization experiments with mercury which is toxic, remote handling made close observation and accurate volume control difficult. Instead of using the closed chamber, the flat heated plate was operated in an open air atmosphere inside a hood. In this way, the mercury droplet could be placed by hand on the surface in a more gentle manner and thereby eliminate much of the droplet vibration problems discussed in Ref. 7 and seen in our earlier experiments.

The oxidation of the mercury drop did not appear to be much of a problem based on the volume of the residue left at the end of the evaporation. In addition, a check was made against data in the helium atmosphere (approximately the same theoretical diffusivity) and found to be in agreement. Also, as will be shown later, in either air or an inert atmosphere, the dominant mechanism of mass transfer is diffusion. Air is the only gas for which a measured value of the diffusion constant for mercury is given.[12]

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Mercury vapor contamination of the room air was of concern even with the hot plate inside the ventilated laboratory hood. The initial check using an air flow meter indicated that instabilities of air under the ventilation hood in the forms of swirls and eddys could cause leakage of mercury vapor into the room. A detailed ventilation study was performed. It was found that the experiment could be conducted safely with the hood window opened to 25 cm and an air flow of 1 m/sec. All the prescribed government safety recommendations[11] were followed including the taking of blood and hair samples.

The mercury was delivered to the heated surface using a thin pipette attached to a hypodermic syringe. The mercury was preheated for some of the experiments by an electrical resistance type heater wound around a metal storage cup. For these hot liquid measurements, the delivery pipette was stored with its tip in the hot mercury to reduce cooling during delivery.

An electrical timer which measured time to one tenth of a second was used to measure the vaporization time. The vaporization times ran between 15 and 60 seconds. The plate temperature was measured using imbedded chromel-alumel thermocouples and extrapolating to find the surface temperature. The volume of each mercury drop expelled onto the hot plate was found by measuring the length of the liquid column it had occupied in a pipette. The pipettes were calibrated using laboratory scales to develop calibration curves in terms of the length of mercury in the pipette (measured to the top of the miniscus). Prior to delivery, the pipette was placed next to a scale graduated in millimeters with a readability of 0.5 mm or an accuracy of ±0.00025 ml in volume.

Data were first taken with cold mercury, and then the mercury was heated to 265° C in a stainless steel cup. It was noticed that when volumes above 0.014 ml (0.32 cm diam.) were used, the droplet would jump and vibrate violently when first delivered onto the heated plate and this erruption of the droplet would usually occur again during evaporation. Some of these droplets never reached a stable film boiling condition but continued to bounce across the plate and vibrate throughout the entire evaporation process. Consequently, drops with diameters less than 0.32 cm (0.014 cc) were used in the experiment.

Experimental Results

The measured vaporization time data are displayed in Fig. 1 for mercury initially at room temperature and preheated to 265° C. As seen in Fig. 1, preheating the mercury had a very small effect on the vaporization time. Since the ratio of the heat of vaporization to $C_p\Delta T$ required to raise the drop to its boiling point is small, these results were expected.

The overall average heat transfer coefficients to the mercury droplets can be estimated from these curves; however, it is first necessary to estimate the area through which the heat passes. This will

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be considered in a later section.

Previous Correlation of Vaporization Times

Previous equations for predicting the vaporization times of drops over predicted the time required for the mercury drops to vaporize by as much as 300 percent. This was surprising since these theories had correlated water, hydrocarbons and cryogens over a very large range of droplet volumes. Reference 10 had even correlated water vaporizing into a helium atmosphere where diffusion had a major contribution.

Diffusive evaporation from the upper surface of the droplet explains why earlier correlations did not work on the mercury data. As shown in Appendix A of this paper, if the following dimensionless group is greater than 2, diffusion from the upper surface of the drop will be a dominant mechanism.

$$N_{DC} = \frac{\lambda D_{AB} M P_s}{R T_s r_o h_o \, \Delta T} \frac{A_d}{A_B} > 2 \qquad (1)$$

These and other symbols are defined in the Nomenclature list at the end of this paper. Since N_{DC} is greater than 2 for mercury drops, the previously-developed closed-form theories require some modification.

Poppendiek[8] vaporized mercury drops in film boiling into a saturated atmosphere of mercury vapor. His results are shown in Fig. 2. When diffusion is minimized by reducing the partial pressure driving force, the vaporization times increase by a factor of 7. Clearly, both the theory and experiment indicate that diffusion from the upper surface of the drop can be dominant mechanism in mercury film boiling.

Unfortunately, Poppendiek did not document the volume of mercury used so that the saturated atmospheric data could not be compared to theory.

In addition, as will be discussed later, surface wetting beneath the drop (nucleate boiling) and convection augmented diffusion from the upper surface of the drop could also contribute to the difference between previous theories and the data.

Analysis of Vaporization Times

The experimentally measured vaporization time of a discrete liquid drop undergoing film boiling can be determined by a direct integration of an energy balance on the drop:

$$-\lambda \rho_L \frac{dV}{dt} = h_T(V) A_B(V) \Delta T \qquad (2)$$

where the total heat transfer coefficient, h_T, and the drop's bottom area A_B are dependent on the volume V of liquid that exists at any time, t.

In order to integrate eq. (2), h_T and A_B must be related to the droplet volume, properties of the liquid and vapor, plate temperature, and the environmental conditions surrounding the drop. The

expression for the heat transfer coefficient is developed by analyzing the model shown in Fig. 3. Heat transfer to the drop occurs by conduction across the small vapor gap and by radiation from plate to the drop. Mass is lost by diffusive evaporation from the upper surface of the sphere and by film boiling off the bottom of the drop. The complete analysis for h_T is presented in Appendix A. The relationship of A_B to droplet volume is given in Appendix B.

First, eq. (2) will be non-dimensionalized using the same definitions as in Ref. 10. Let

$$t^* = \frac{t}{\lambda \rho_L \left[\frac{\mu L^5}{k^3 \lambda^* g (\rho_L - \rho_V) \rho_V \Delta T^3} \right]^{1/4}} \quad (3)$$

$$v^* = \frac{V}{L^3} \quad (4)$$

$$A_B^* = \frac{A_B}{L^2} \quad (5)$$

$$L = \left[\frac{\sigma g_c}{(\rho_L - \rho_V) g} \right]^{1/2} \quad (6)$$

$$h^* = \frac{h}{\left[\frac{k^3 \lambda^* (\rho_L - \rho_V) \rho_V g}{\Delta T \mu L} \right]^{1/4}} \quad (7)$$

Eq. (2) can now be written as

$$-dV^* = h_T^* A_B^* \, dt^* \quad (8)$$

Integrating eq. (8) gives:

$$t^* = \int_{V^*}^{0} \frac{-dV^*}{h_T^* A_B^*} \quad (9)$$

Substituting the expression for h_T^*, eq. (A15) and A_B^*, (πr_c^2, eq. (B2)), into eq. (9) and performing the integration gives

$$t^* = \frac{6.02}{N^*} v^{*0.664} \quad (10)$$

where

$$N^* = \frac{\left(\frac{30.7 \, \lambda D_{AB} M P_s}{\Delta T \, R T_s} \right)}{\left[\frac{k^3 \lambda^* (\rho_L - \rho_V) \rho_V g}{\Delta T \, \mu L} \right]^{1/4}} \quad (11)$$

Equation (10) will be compared to the experimental data in a later section.

Heat Transfer Coefficient

The various heat transfer mechanisms combine to give a total heat transfer coefficient that can be found by rearranging eq. (2) into the form

$$h_T = \frac{-\lambda \rho_L \frac{dV}{dt}}{A_B \, \Delta T} \quad (12)$$

The dV/dt term was found at selected volumes by measuring the slope of the vaporization curves in Fig. 1. The A_B was evaluated using eq. (B2) in Appendix B.

The heat transfer coefficient data are displayed in Fig. 4. As seen in Fig. 4, the heat transfer coefficients are very large compared to the normal values of 0.01 cal/cm sec c (or 50 BTU/hr ft^2 F) expected from film boiling theory. The analytical predictions for the heat transfer coefficient fall below the experimental values. However, the analytical trend seems to follow the experimental trend. Consequently, if desired, an empirical correction factor could be applied to the analytical expression to give agreement over a wide volume range.

Because the emissivity of mercury is small, the heat is transferred to the drop primarily by conduction across the vapor gap. Therefore, the vapor gap beneath the drop can be estimated by the equation

$$\delta = \frac{k}{h_T} \quad (13)$$

The gap thickness is estimated to be on the order of one ten-thousand of a centimeter. The hot plate surface was polished initially to a 5 micro-cm finish.

There are at least two possible reasons why the analytical heat transfer coefficient underestimates the measured heat transfer coefficient. First, convection currents above the drop could enhance the diffusive mechanism from the upper surface of the drop. Second, because of the very small vapor gap thickness and the large surface tension of mercury, some direct contact between mercury and the surface could be occurring. It should be expected that analytical models, which assume an insulating vapor film and quiescent drops, will underestimate the measured heat transfer coefficient. Bradfield[15] indicated that liquid-solid contact can occur even with large vapor gap thickness (0.001 cm) and low surface tension fluids. Harvey[4](p. 42) indicates that a large percent change in the droplet diameter can occur due to direct contact. In addition, Marangoni forces could also play a role in enhancing the heat transfer coefficient.

Correlation of the Vaporization Time

The theory, eq. (10), is compared to experiment in Fig. 5. Reasonable agreement is seen especially at the smaller droplet volumes. The deviation between experiment and theory could be due to the extra heat that is transferred to the drop by direct contact or removed by convection augmented diffusion from the upper drop surface as was just discussed.

It is expected that solid-liquid contact will decrease at the smaller droplet volumes. In this range, the surface tension forces will compact the mercury into a tight sphere and thereby reduce the possibility of liquid surface contact. In this low volume range, the vaporization times are seen to be in improved agreement with the measured data.

Conclusions

Diffusive evaporation from the upper surface of the drop is a major mechanism of mass transfer for liquid mercury drops in Leidenfrost film boiling in an air atmosphere. Theoretical expressions are derived for the heat transfer coefficient and the vaporization times, which are in fair agreement with experimental data.

Nomenclature

A_B bottom area of drop

A_d area of drop in which diffusion occurs

A^* dimensionless area, defined by eq. (5)

c_p specific heat at constant pressure of vapor

D_{AB} diffusion coefficient

g coefficient of gravity

g_c gravitational constant

h heat transfer coefficient

h_o film boiling heat transfer coefficient when radiation and diffusion are assumed zero

h_{rad} radiative heat transfer coefficient

h_{conv} convection heat transfer coefficient

h_T total heat transfer coefficient to drop

k thermal conductivity of vapor

L characteristic length, eq. (6)

M molecular weight

N_{DC} dimensionless diffusion parameter defined by eq. (A4)

N^* dimensionless diffusion parameter defined by eq. (11)

P_s saturation pressure

R gas constant

r radial coordinate

r^* dimensionless radius, r/L

r_o drop radius

r_c contact radius

T temperature

T_p plate temperature

T_s saturation temperature

ΔT temperature difference, $(T_p - T_s)$

t time

t^* dimensionless time, eq. (3)

V droplet volume

V^* dimensionless volume, eq. (4)

$w(\delta)$ axial velocity at bottom of drop

Z axial coordinate

δ vapor gap thickness

δ_o vapor gap thickness when diffusion and radiation are not present

ϵ_L emissivity of liquid mercury ($\epsilon_L = 0.12$)

λ latent heat of vaporization

λ^* modified latent heat of vaporization defined in Ref. 10 as

$$\frac{c_p \Delta T}{\lambda} \leq 2 \qquad \lambda^* = \lambda \left(1 + \frac{7}{20} \frac{c_p \Delta T}{\lambda}\right)^{-3}$$

$$\frac{c_p \Delta T}{\lambda} > 2 \qquad \lambda^* = \lambda \left[\frac{0.874 \ln\left(1 + \frac{c_p \Delta T}{2\lambda}\right)}{\frac{c_p \Delta T}{2\lambda}}\right]^3$$

μ viscosity of vapor

ρ_L density of liquid

ρ_v vapor density

σ surface tension

Appendix A

Heat Transfer Coefficient

The overall energy balance on the droplet shown in Fig. 3 can be written as

$$h_T A_B \, \Delta T = \frac{k A_B \, \Delta T}{\delta} + h_{rad} A_r \, \Delta T - h_{CONV} A_c (T_s - T_a)$$

$$= -\rho_v \lambda w(\delta) A_B + \frac{\lambda D_{AB} M P_s A_d}{R T_s r_o} \qquad (A1)$$

On the right-hand side of eq. (A1), the first term represents heat removed from the drop by evaporation from the lower surface, while the second term represents the latent heat requirements for the evaporation by diffusion from the upper surface. For the form of diffusive energy loss, the partial pressure of the liquid in the vapor environment is assumed small and an equivalent spherical shape has been assumed. Heat transfer to the drop occurs by conduction across the small vapor gap δ, by radiation from the plate to the drop and by natural convection from the hot vapor and atmosphere surrounding the drop.

The momentum equation along with the appropriate boundary conditions was solved in Ref. 12 to give

$$w(\delta) = \frac{2\pi}{3} \frac{(\rho_L - \rho_v) g \delta^3}{\mu L} \left(\frac{V^*}{A_B^{*2}}\right) \qquad (A2)$$

where V^* and A_B^* are dimensionless volume and area of the bottom of the drop respectively. They are defined by eq. (4) and (5) in the body of this report.

Equation (A2) is now combined with the energy balance, eq. (A1), to determine the gap thickness of the vapor δ,

$$\delta^4 = \delta_o^4 \left[1 - \left(N_{DC} - \frac{h_{NET}}{h_o}\right) \frac{\delta}{\delta_o}\right] \qquad (A3)$$

where

$$N_{DC} = \frac{A_d}{A_B} \frac{\lambda D_{AB} M P_s}{R T_s r_o h_o \, \Delta T} \qquad (A4)$$

$$h_{net} = h_{rad} \frac{A_r}{A_B} - h_{conv} \frac{A_c (T_s - T_a)}{A_B \, \Delta T} \qquad (A5)$$

and as shown in Ref. 12 for pure film boiling, no radiation or diffusion

$$\delta_o = \left[\frac{3k \, \Delta T \mu L}{2\pi \lambda (\rho_L - \rho_v) \rho_v g}\right]^{1/4} \left(\frac{A_B^{*2}}{V^*}\right)^{1/4} \qquad (A6)$$

and for the heat transfer coefficient

$$h_o = \frac{k}{\delta_o} = 1.21 \left(\frac{k^3 \lambda^* (\rho_L - \rho_v) \rho_v g}{\Delta T \mu L}\right)^{1/4} \left(\frac{V^*}{A_B^{*2}}\right)^{1/4} \qquad (A7)$$

where the latent heat of vaporization has been replaced by the modified latent heat of vaporization

which accounts for convection effects in the vapor gap (see list of symbols).

The dimensionless group N_{DC} is a measure of the importance of diffusion from the upper surface of the drop to conduction beneath the drop.

Equation (A3) can be written as

$$N_{DC} - \frac{h_{NET}}{h_o} = \frac{1 - \left(\frac{\delta}{\delta_o}\right)^4}{\left(\frac{\delta}{\delta_o}\right)} \qquad (A8)$$

The exact solution of eq. (A8) is presented in Fig. 6. For small values of N_{DC}

$$N_{DC} - \frac{h_{NET}}{h_o} \leq 2 \qquad (A9)$$

The solution for δ/δ_o can be written as

$$\frac{\delta}{\delta_o} \approx 1 - \frac{1}{4} \left(N_{DC} - \frac{h_{NET}}{h_o}\right) \qquad (A10)$$

For large values of N_{DC}

$$N_{DC} - \frac{h_{NET}}{h_o} > 2 \qquad (A11)$$

$$\frac{\delta}{\delta_o} \approx \frac{1}{N_{DC} - \frac{h_{NET}}{h_o}} \qquad (A12)$$

The solution for the heat transfer coefficient and vaporization times presented in Ref. 10 was based on eq. (A10), that is, diffusion is relatively small compared to heat conduction across the vapor gap. For mercury drops in either air or an inert atmosphere, eq. (A10) is not valid because N_{DC} is large (in the range of 5 to 10 for the mercury data). The balance of this appendix is concerned with the derivation of the heat transfer coefficient using eq. (A12) where diffusion dominates.

The total heat transfer coefficient can now be found by substituting from eq. (A12) into the left-hand side of eq. (A1).

$$h_T = \frac{\lambda D_{AB} M P_s A_d}{R T_s \, \Delta T r_o A_B} \qquad (A13)$$

This form of the heat transfer coefficient could be obtained directly from eq. (A1) by simply neglecting the effect of vapor flow ($-\rho_v \lambda w A_B$) beneath the drop. The analysis has indicated that this is a permissible assumption if $N_{DC} - h_{NET}/h_o$ is greater than 2.

Non-dimensionalizing eq. (A13) in the manner suggested by eq. (7) in the body of the report gives

$$h_T^* = \frac{\lambda D_{AB} M P_s}{R T_s \Delta T r_o} \frac{A_d}{A_B} \left(\frac{k^3 \lambda^* (\rho_L - \rho_v) \rho_v g}{\Delta T \mu L} \right)^{-1/4} \qquad (A14)$$

The expression for h_T depends on volume through the terms r_o, A_d, and A_B. The volume dependence of r_o, A_d, and A_B are given respectively by eqs. (B4), and (B6). Substituting these values into eq. (A14) gives

$$h_T^* = \frac{N^*}{V^{*0.964}} \qquad (A15)$$

where

$$N^* = \frac{30.7 \lambda D_{AB} M P_s}{\Delta T \, R T_s} \left(\frac{k^3 \lambda^* (\rho_L - \rho_v) \rho_v g}{\Delta T \mu L} \right)^{-1/4} \qquad (A16)$$

Appendix B

Area of Drop

The theory of Ref. 10 based the area of heat transfer on the maximum radius of the drop. However, for dimensionless drop volumes less than 5, that is

$$V^* = \frac{V}{\left[\dfrac{\sigma g_c}{(\rho_L - \rho_v) g} \right]^{3/2}} < 5 \qquad (B1)$$

the contact area is considerably smaller than the area subtended by r_{max}. This is shown pictorially in Fig. 7. The curves were generated from the data of Ref. 14. The curves can be fitted by the following equations:

$$r_c^* = 0.282 \, V^{*0.65} \qquad V^* < 5 \qquad (B2)$$

$$r_c^* = \frac{1}{3} V^{0.525} \qquad V^* \geq 5 \qquad (B3)$$

$$r_o^* = \left(\frac{3V^*}{4\pi} \right)^{1/3} \qquad V^* < 5 \qquad (B4)$$

$$r_o^* = 0.4 \, V^{*0.465} \qquad V^* \geq 5 \qquad (B5)$$

The ratio of diffusion to contact area is given by

$$\frac{A_d}{A_B} = \frac{4\pi r_o^{*2}}{\pi r_c^{*2}} = \frac{4\pi \left(\dfrac{3V^*}{4\pi} \right)^{2/3}}{0.25 \, V^{*1.3}} \qquad V^* \leq 5 \qquad (B6)$$

$$\frac{A_d}{A_B} = \frac{\pi r_o^{*2}}{\pi r_c^{*2}} = 2.25 / V^{*0.12} \qquad V^* > 5 \qquad (B7)$$

As an analytical simplification, the equation for the drop's surface area in eq. (B6) is assumed to be represented by a complete sphere. That is, the area due to the flattening of the sphere at the plate is neglected. For V^* greater than 5, the drop is assumed to be represented by a flat cylinder, such that only the upper area contributes to diffusive evaporation.

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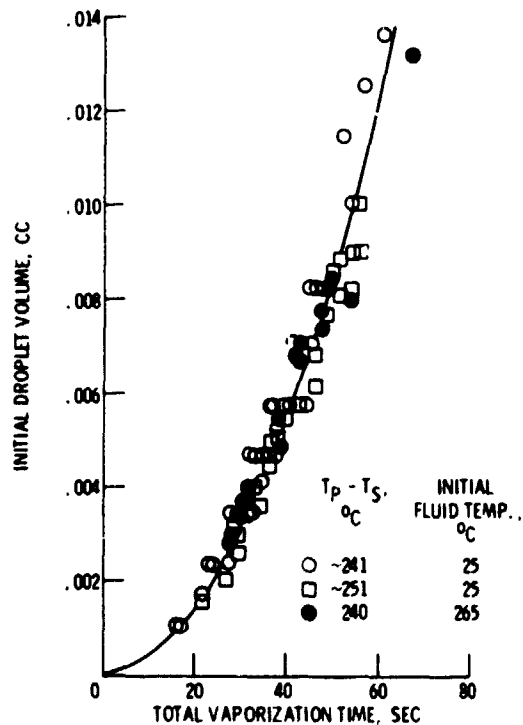15. Bradfield, W. S., Ind. Eng. Chem. Fundamentals, 5, 200-204, 1966.
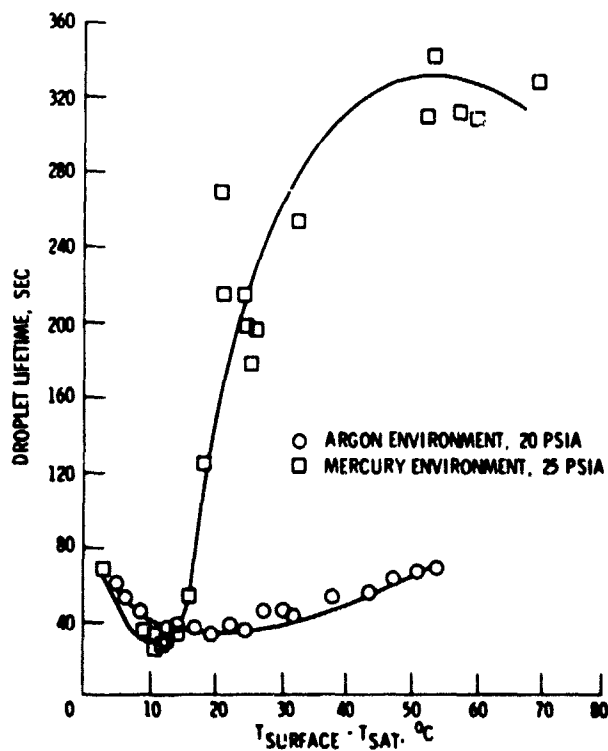
Figure 1. - Total vaporization time of mercury.

Figure 2 - The lifetimes of mercury droplets vaporizing on a tantalum surface in an argon and mercury environment (reference 8).
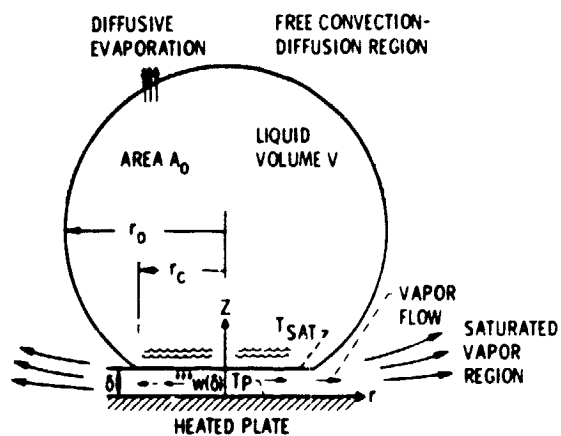
DIFFUSIVE
EVAPORATION

FREE CONVECTION-
DIFFUSION REGION

LIQUID
VOLUME V

AREA A_0

r_0

r_c

Z

T_{SAT}

VAPOR
FLOW

SATURATED
VAPOR
REGION

δ

w(δ)

TP

r

HEATED PLATE

Figure 3. - Schematic model for evaporation of flat bottom sphere.
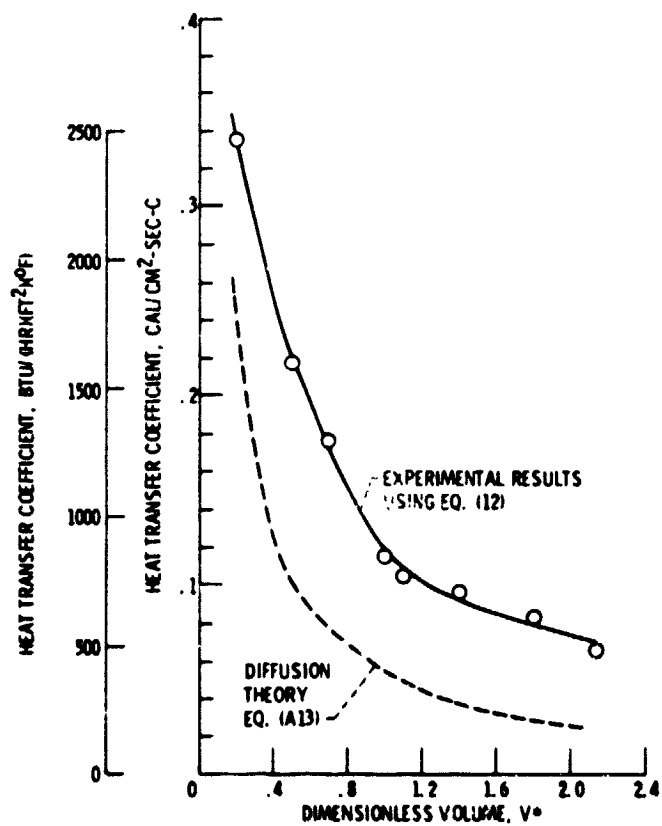
Figure 4. - Dependence of heat transfer coefficient on dimensionless
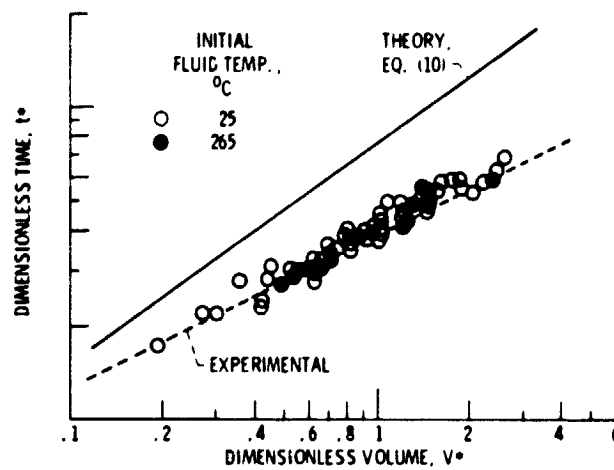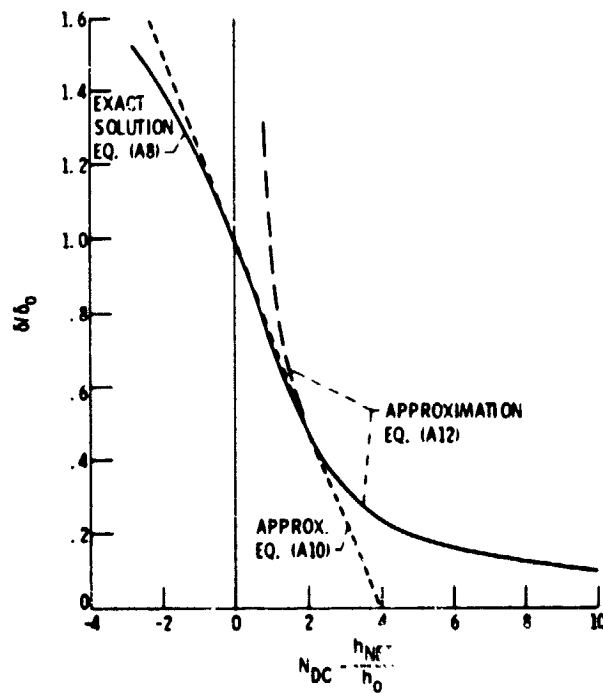volume for mercury droplets.

Figure 5. - Total vaporization time of mercury droplets.

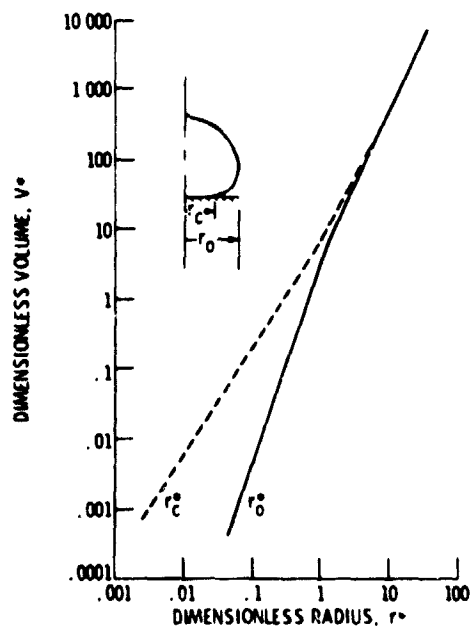Figure 6. - Exact and approximate relationships of δ/δ_0 to the quantity $(N_{DC} - h_{NET}/h_0)$.

Figure 7. - Contact and maximum radii for non-wetting drop.