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TRANSPORT PHENOMENA AT THE LIQUID-VAPOR INTERFACE
OF MERCURY USING A RADIOACTIVE TRACER

DISSERTATION

Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in the Graduate
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By

Kyril Francis Wylie, B.S., M.A.

* * * * *

The Ohio State University
1970

Approved by

Adviser

Department of Mechanical
Engineering

Adviser

Department of Chemical
Engineering

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VITA

February 24, 1931 . . . Born - Akron, Ohio

1958 B.S., Kent State University, Kent, Ohio

1960 M.A., Kent State University, Kent, Ohio

1960-1965 Research Physicist, Mound Laboratory,
Miamisburg, Ohio

1966 Senior Research Physicist, Mound Laboratory,
Miamisburg, Ohio

1960-1966 Consultant Physicist to the Nuclear Medicine
Laboratory, Grandview Hospital, Dayton, Ohio

1966-1968 Graduate Research Associate, Department of
Chemical Engineering, The Ohio State
University, Columbus, Ohio

1968-1969 Graduate Teaching Associate, Department of
Mechanical Engineering, The Ohio State
University, Columbus, Ohio

1969 National Science Foundation Summer Training
Fellowship, The Ohio State University,
Columbus, Ohio

1969-1970 Graduate Research Associate, Department of
Chemical Engineering, The Ohio State
University, Columbus, Ohio

PUBLICATIONS

"Investigation of the Anomalous Proton Magnetic Resonance Behavior in
Lithium Sulfate Monohydrate," Bulletin of the American Physical
Society, Series II, Volume 5, Number 6, p. 466, 1960, with A. A.
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- "Transport Phenomena at Liquid-Metal-Vapor Interfaces Using Radioactive Tracers." Nuclear Applications, Volume 5, pp. 263-268, 1968, with C. E. Dryden.

FIELDS OF STUDY

Major Field: Nuclear Engineering

Studies in "broad-line" nuclear magnetic resonance. Professors A. A. Silvidi and J. W. McGrath.

Studies in diagnostic medicine using radioactive tracer techniques.

Studies in experimental uses of radioactive tracers.

Studies in low-level counting of radionuclides.

Studies in transport phenomena at liquid-metal-vapor interfaces using radioactive tracers. Professors R. S. Brodkey, C. E. Dryden, D. D. Glower, and R. A. Krakowski.

TABLE OF CONTENTS

	Page
ACKNOWLEDGMENTS	ii
VITA	iii
NOMENCLATURE AND SYMBOLS	viii
LIST OF FIGURES	xv
LIST OF TABLES	xvii
Chapter	
I. INTRODUCTION	1
II. PREVIOUS RELATED STUDIES	3
Heat Transfer Analysis	
Nusselt's Analysis	
Examinations of Nusselt's Analysis	
Experimental Studies	
Mass Transfer Analysis	
Theoretical Basis	
Liquid-Vapor Models	
Experimental Studies	
Summary	
III. DISSERTATION RATIONALE	42

Chapter	Page
IV. ISOTHERMAL EQUILIBRIUM STUDY	45
Isothermal Equilibrium Model	
Experimental Procedure	
Production and Analysis of the Radiomercury	
Data and Results	
Discussion	
V. NONISOTHERMAL STUDY	65
Nonisothermal Condition Model	
Condensation Coefficient Determination	
Evaporation Coefficient Determination	
Heat Flux Determination	
Experimental Procedure	
Production and Analysis of the Radiomercury	
Data and Results	
Discussion	
VI. CONCLUSIONS	95
Isothermal Equilibrium	
Nonisothermal Conditions	
VII. RECOMMENDATIONS	97
APPENDIX	
A. DATA AND SAMPLE CALCULATIONS FOR THE ISOTHERMAL EQUILIBRIUM STUDY	98
Data	
Sample Calculations	
Determination of τ	

Determination of σ

Alternative Analysis

B. DATA AND SAMPLE CALCULATIONS FOR THE NONISOTHERMAL STUDY	102
--	-----

Data

Some Basic Data

Counting Rates

GM Detector-Ratemeter-Recorder Response

GM Detector Counting Data

Sample Calculations

Determination of a_1/a_b

Slope Determination

Determination of $\Gamma \sigma_c$

Determination of σ_c

Determination of σ_e

Determination of the Heat Flux

Determination of h_i

C. RADIOMERCURY IN THE BOILER FOR THE NONISOTHERMAL STUDY	123
--	-----

Radiomercury in the Boiler

Dilution in the Boiler

REFERENCES	126
----------------------	-----

NOMENCLATURE AND SYMBOLS

Symbol	Description	Units
a_b	Specific activity of liquid phase in boiler	cpm/g
a_l	Specific activity of liquid phase	cpm/g
a_v	Specific activity of vapor phase	cpm/g
A	Area	cm ²
$\langle A \rangle$	Average of the total number of nucleons in the radioactive nuclei present	----
A_l	Total activity of liquid phase	cpm
A_v	Total activity of vapor phase	cpm
A_3	Total number of nucleons in the Hg-203 nucleus	----
A_7	Total number of nucleons in the Hg-197 nucleus	----
c	Radiation detector efficiency	cpm/dpm
$\langle c \rangle$	Average radiation detector efficiency	cpm/dpm
c_p	Heat capacity (specific heat) of the condensate film	(g-cal)/(g °C)
$c_{p,v}$	Heat capacity (specific heat) of the vapor	(g-cal)/(g °C)
c_T	Total number of molecules per unit vapor volume	molecules/cm ³
c_v	Number of radionuclides per unit vapor volume	molecules/cm ³
dc_v	Differential change in number of radionuclides per unit vapor volume	molecules/cm ³

Symbol	Description	Units
c_3	Radiation detector efficiency for Hg-203	cpm/dpm
c_7	Radiation detector efficiency for Hg-197	cpm/dpm
C_l	Heat capacity (specific heat) of liquid phase	(g-cal)/(g °C)
d	Thickness of liquid-vapor interphase zone	cm
g	Gravitational acceleration	cm/min ²
G	Transfer flux (mass or molecular)	g/(cm ² min) or molecules/(cm ² min)
G_c	Condensation transfer flux (mass or molecular)	g/(cm ² min) or molecules/(cm ² min)
G_e	Evaporation transfer flux (mass or molecular)	g/(cm ² min) or molecules/(cm ² min)
$G_{e,exp}$	Experimentally determined evaporation transfer flux (mass or molecular)	g/(cm ² min) or molecules/(cm ² min)
$G_{e,max}$	Maximum evaporation transfer flux (mass or molecular) according to Hertz and Langmuir	g/(cm ² min) or molecules/(cm ² min)
G_{net}	Net transfer flux (mass or molecular)	g/(cm ² min) or molecules/(cm ² min)
G_R	Reflection transfer flux (mass or molecular)	g/(cm ² min) or molecules/(cm ² min)
h_c	Inside film heat transfer coefficient (inside of condenser pipe)	(g-cal)/(cm ² min °C)
h_i	Liquid-vapor interfacial heat transfer coefficient	(g-cal)/(cm ² min °C)
h_l	Condensate film heat transfer coefficient	(g-cal)/(cm ² min °C)

Symbol	Description	Units
h_{Nu}	Heat transfer coefficient predicted from theories which take into account only the condensate film's thermal resistance (Nusselt heat transfer coefficient)	(g-cal)/cm ² min °C)
$\overline{h_{Nu}}$	Average Nusselt heat transfer coefficient	(g-cal)/(cm ² min °C)
h_w	Heat transfer coefficient due to the condenser wall	(g-cal)/(cm ² min °C)
i	Coordinate	cm
k	Thermal conductivity of condensate film	(g-cal)/(cm min °C)
L	Condenser length	cm
L	Mercury mass in circulating liquid phase at time $t = t$	g
dL	Differential change in mercury mass in circulating liquid phase	g
L_0	Mercury mass in circulating liquid phase at time $t = 0$	g
ΔL	Change in mercury mass in circulating liquid phase ($= L - L_0$)	g
m	Mass per molecule	g/molecule
m_v	Total mercury mass in vapor volume	g
m_3	Mass of Hg-203	g
m_7	Mass of Hg-197	g
M	Molecular weight	g/(g-mole)
N	Number of radioactive molecules	----
N	Number of molecules	----
N^*	Avagadro's number	molecules/(g-mole)
N_s	Molecular concentration of molecules evaporating from liquid phase	molecules/(cm ³)

Symbol	Description	Units
N_v	Molecular concentration of molecules condensing from vapor phase	molecules/(cm ³)
N_3	Number of Hg-203 molecules	----
N_7	Number of Hg-197 molecules	----
P	Pressure	dyne/cm ²
P_R	Saturation pressure associated with temperature T_R	dyne/cm ²
P_s	Saturation pressure associated with temperature T_s	dyne/cm ²
P_v	Pressure of pure saturated bulk vapor	dyne/cm ²
P_{si}	Pressure associated with temperature T_{si}	dyne/cm ²
P_{vi}	Pressure associated with temperature T_{vi}	dyne/cm ²
q	Heat flux for interfacial energy transfer	(g-cal)/(cm ² min)
q_c	Condensation heat flux	(g-cal)/(cm ² min)
Q	Rate at which energy (heat) is supplied to the boiler	(g-cal)/min
r	"Boil-up" rate from the boiler	g/min
R	Gas constant	(dyne-cm)/(g °K)
R_u	Universal gas constant (= M R)	(dyne-cm)/(g-mole °K)
t	Time	min
Δt	Change in time	min
dt	Differential time change	min
T	Temperature	°C
T_{ave}	Average temperature	°C
ΔT_f	Temperature drop across the condensate film (= $T_s - T_w$)	°C

Symbol	Description	Units
T_R	Temperature of vapor molecules reflected from condensate film	$^{\circ}\text{C}$
T_S	Temperature of condensate surface	$^{\circ}\text{C}$
T_{si}	Interfacial temperature for molecules from condensate surface	$^{\circ}\text{C}$
T_v	Temperature of bulk vapor	$^{\circ}\text{C}$
T_{vi}	Interfacial temperature of vapor molecules	$^{\circ}\text{C}$
T_w	Surface temperature of condenser wall	$^{\circ}\text{C}$
$\bar{u}$	Net stream velocity in x-direction	cm/min
U	Overall heat transfer coefficient	(g-cal)/(cm ² min $^{\circ}\text{C}$)
V	Volume	cm ³
V	Mean molecular velocity	cm/min
V_s	Mean velocity of evaporating molecules	cm/min
V_v	Mean velocity of condensing molecules	cm/min
V_x	Molecular progress velocity in x-direction	cm/min
x	Coordinate	cm
x	Mass fraction	----
x_b	Mass fraction of radionuclides in boiler	----
x_l	Mass fraction of radionuclides in liquid phase	----
dx_l	Differential mass fraction change of radionuclides in liquid phase	----
x_v	Mass fraction of radionuclides in vapor phase	----

Symbol	Description	Units
x_3	Mass fraction of Hg-203	----
x_7	Mass fraction of Hg-197	----
Greek Letters		
β_v	$= \sqrt{M/(2RT)}$	min/cm
β_3	Fraction of Hg-203 atoms that disintegrate per unit time	min ⁻¹
β_7	Fraction of Hg-197 atoms that disintegrate per unit time	min ⁻¹
$\langle \beta \rangle$	Average disintegration rate	min ⁻¹
Γ	Correction factor, defined by equation (12)	----
δ	Condensate film thickness	cm
Δ	Signifies difference between properties	----
λ^*	Latent heat of vaporization (Nusselt's equation)	(g-cal)/g
λ	Latent heat of vaporization	(g-cal)/g
μ	Viscosity of the condensate	g/(cm min)
ξ	Temperature jump coefficient	
ρ	Density	g/cm ³
ρ_L	Density of the condensate	g/cm ³
σ	"Evaporation" coefficient according to Knudsen	----
σ	"Transfer" coefficient	----
σ_c	Condensation coefficient	----
σ_e	Evaporation coefficient	----
τ	Defined by equation (49)	min

Symbol	Description	Units
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ϕ	Non-dimensional number. A measure of bulk movement of vapor molecules towards the liquid-vapor interface ($= \beta_v \bar{u}$)	----
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$\text{erf}(\phi)$	$= \frac{2}{\sqrt{\pi}} \int_0^{\phi} [\exp(-\phi^2)] d\phi$	----
--------------------	--	------

Subscripts

ave	Average
c	Condensation or condensing
e	Evaporation or evaporating
f	Film
i	Liquid-vapor interface
l	Liquid or condensate
net	Net
Nu	Nusselt
o	Initial
R	Reflection or reflecting
s	Liquid or condensate surface
T	Total
u	Universal
v	Vapor
w	Condenser wall
x	Coordinate
3	Hg-203
7	Hg-197

LIST OF FIGURES

Figure	Page
1. Flow diagram of the heat transfer problem in film type condensation	4
2. Nusselt's model of film condensation	6
3. The " $T_i = T_v$ " condensation model	17
4. The Bornhorst-Hatsopoulos liquid-vapor interface model	23
5. The Fedorovich-Rohsenow liquid-vapor interface model	25
6. The vertical " $T_i = T_v$ " condensation model	28
7. A hypothetical isothermal experimental system	46
8. The isothermal experimental system	54
9. The resolution of the spectrometer system as a function of photon energy. (The two points marked Ba-133 represent two major peaks in the disintegration scheme of the same nuclide, namely, Ba-133.)	58
10. An extended counting study of an aliquot of the irradiated mercury	59
11. A model of the steady state nonisothermal experimental system	66
12. The nonisothermal experimental system	71
13. Thermocouple locations for the nonisothermal experimental system	74
14. Movable chromel-alumel thermocouple readings for nonisothermal experimental study number 1	79
15. Movable chromel-alumel thermocouple readings for nonisothermal experimental study number 2	80

Figure	Page
16. Movable chromel-alumel thermocouple readings for nonisothermal experimental study number 3	81
17. Movable chromel-alumel thermocouple readings at the liquid-vapor interface for nonisothermal experimental study number 1	82
18. Movable chromel-alumel thermocouple readings at the liquid-vapor interface for nonisothermal experimental study number 2	83
19. Movable chromel-alumel thermocouple readings at the liquid-vapor interface for nonisothermal experimental study number 3	84
20. The flow from a flat plate and the crude similarity to the nonisothermal experimental configuration	88
21. Condensation coefficients and drift velocity correction factors from data based upon the evaporation coefficient being equal to unity	92
22. An alternative approach to the display of the six isothermal equilibrium studies	101
23. GM-Ratemeter-Recorder response for nonisothermal study number 1 (-8 to 25 minutes)	107
24. GM-Ratemeter-Recorder response for nonisothermal study number 1 (25 to 56 minutes)	108
25. GM-Ratemeter-Recorder response for nonisothermal study number 2 (-5 to 22 minutes)	109
26. GM-Ratemeter-Recorder response for nonisothermal study number 2 (22 to 51 minutes)	110
27. GM-Ratemeter-Recorder response for nonisothermal study number 3 (-8 to 25 minutes)	111
28. GM-Ratemeter-Recorder response for nonisothermal study number 3 (25 to 57 minutes)	112
29. $(a_1/a_b)_{GM \text{ net}}$ as a function of time for nonisothermal study number 1	118

LIST OF TABLES

Table	Page
1. A Summary of the Experimentally Determined Transfer Coefficients for Water	40
2. A Summary of the Experimentally Determined Transfer Coefficients for Liquid Metals	41
3. Calculated and Measured Data and Results Required for the Determination of the Transfer Coefficient for Film Condensation of Mercury	62
4. Calibration of the Pressure Transducer to Mercury Vapor Temperature and Saturation Pressure	76
5. Calculated and Measured Data and Results Required for the Determination of the Condensation and Evaporation Coefficients, Heat Flux, and Interfacial Heat Transfer Coefficient for Film Condensation of Mercury	78
6. Data for the Isothermal Equilibrium Studies	98
7. Basic Data for the Nonisothermal Studies	102
8. Counting Rates for the Nonisothermal Studies	104
9. GM Detector Counting Data for the Nonisothermal Studies . .	113

CHAPTER I

INTRODUCTION

The advent of power stations employing nuclear fuel necessitated an evaluation of heat transfer agents, such as liquid metals, ordinary and heavy water. From studies of the characteristics of water, it is implied that inherent deficiencies exist for water as a heat transfer media. Included in this list are: decomposition under the action of irradiation, rather large thermal neutron capture cross section, corrosional aggressiveness for most metals, and reaction on contact with uranium. Furthermore, high pressures are required to achieve high water temperatures. On the other hand, owing to their thermal-physical properties, one is tempted to employ liquid metals as a heat-transfer agent. In general, good radiation stability and neutron-capture cross sections smaller than water are exhibited by liquid metals. The potential of higher heat transfer rates (for a given temperature difference) is offered by the high thermal conductivity of liquid metals. As a result of their lower vapor pressures (at a given temperature), compact-light weight high temperature heat transfer systems can be used.

One of many heat transfer characteristics being examined by researchers is the vapor-liquid interface transfer coefficient associated with the film condensation of liquid metals. The experimental

procedure presented herein provides an approach to the determination of the condensation-evaporation coefficients which has not been attempted with liquid metals, possesses relative freedom from experimental difficulties, and allows examination under nonisothermal conditions or isothermal equilibrium of the vapor-liquid phases. Effectiveness of this radioactive tracer technique is demonstrated by employing normal (non-radioactive) mercury and mercury tagged with Hg-197 and/or Hg-203.

CHAPTER II

PREVIOUS RELATED STUDIES

The need to accurately predict the heat-transfer of a given system coupled with discrepancies between classical interpretation of film type condensation and experimentally determined results provided the stimuli which have induced re-examination of the problem by researchers. In order to facilitate an orderly discussion of these, a rather methodical presentation is employed as shown in Figure 1.

The format used herein consists of considering the classical energy transfer model for film type condensation (topic 1, Figure 1), modifications to this model (topic 3, Figure 1), and the experimental results (topic 4, Figure 1). In general, the experimental results for liquid metals do not agree with the classical model or modifications to it. Since the transfer of energy (in the form of heat) across the condensate film is directly related to molecular action and reaction at the liquid-vapor interface, one is obviously led to a more basic examination of the evaporation and condensation phenomenon at the liquid-vapor interface (topic 2, Figure 1). Here, researchers have proposed the use of several models to overcome the apparent discrepancies between predicted and experimentally determined results. These various models and their interpretations (topic 5, Figure 1), as well as the associated experimental results (topic 4, Figure 1) are presented.

Since the research presented herein is concerned with the study of mercury, those papers containing information deemed pertinent to the study of liquid metals are selectively cited. On the other hand, experimental determinations of the condensation and/or evaporation coefficient of water are cited as an example of work dealing with other fluids, and the implications that this has for the liquid metal problem.

Heat Transfer Analysis

Nusselt's Analysis, (topic 1, Figure 1). Generally speaking, when a saturated vapor is contained in a vessel whose wall temperature is lower than that of the vapor, the vapor is condensed on the wall. In such a process, the liberated energy (in the form of heat) must be removed through the wall. Using a liquid-film model (Figure 2), Nusselt (1) analyzed the heat transfer associated with this process (topic 1, Figure 1).

For the condensation of a pure saturated vapor on a cold flat plate, Nusselt assumed that:

1. The condensed liquid formed a continuous film on the wall.
2. Laminar motion existed throughout the thickness of the continuous film of condensate.
3. The condensed liquid builds up a film which flows by gravity (alone) downward and is continuously renewed by newly condensing pure vapor. Hence, the possible effect of drag on the movement of the condensate film by the stationary vapor is neglected.

Figure 2. Nusselt's model of film condensation

4. The film thickness at the upper edge of the wall is zero, and the thickness is increased in a downward direction.
5. The total thermal resistance to the condensation process is in the film of condensate, through which the latent heat of condensation is conducted, neglecting the cooling of the condensate. In other words, the liquid temperature at the liquid-vapor interface is the saturation temperature associated with the pressure of the bulk vapor.
6. The vertical wall is maintained at some constant temperature, and the temperature difference between vapor and condenser wall is constant at all points across the condensate. Furthermore, the temperature distribution in the film is essentially linear.
7. All properties of the fluid are constant.
8. Momentum changes through the condensate can be neglected. Thus, an essentially static balance of forces is implied by this particular assumption.

Employing these assumptions, the local heat-transfer coefficient at a distance x from the upper edge of the plate is given by

$$h_{Nu} = \frac{k}{\delta} = \left[\frac{g \rho_L^2 k^3 \lambda^*}{4 \mu x \Delta T_f} \right]^{1/4} \quad (1)$$

The average value along the length L of the plate is found by integration to be

$$\overline{h}_{Nu} = \frac{4}{3} \left[\frac{g \rho_L^2 k^3 \lambda^*}{4 \mu L \Delta T_f} \right]^{1/4} \quad (2)$$

where

$$\Delta T_f = T_s - T_w \quad (3)$$

Examinations of Nusselt's Analysis, (topic 3, Figure 1). Discrepancies between experimental results and theoretical predictions have led many researchers to examine and elaborate on Nusselt's original work. Bromley (2) extended Nusselt's work to include the effect of heat obtained through a decrease in condensate temperature without a change in phase, composition of the mass, or pressure (topic 3.3.3, Figure 1). The resulting equation, valid for values of less than 3 for the ratio $(\Delta T_f c_p)/\lambda$, implied a slight increase in the heat transfer coefficient (for fluids exhibiting an essentially constant heat capacity value). Consideration of non-linearities in the film temperature distribution (topic 3.1.8, Figure 1) on the condensation heat transfer coefficient by Rohsenow (3) led to the latent heat of vaporization (λ^*) in Nusselt's relationship being replaced by $\lambda + 0.68 c_p \Delta T_f$ for the case where the parameter $(\Delta T_f c_p)/\lambda$ exhibited large numerical values. In most practical applications $0 < [(\Delta T_f c_p)/\lambda] < 0.2$, and Rohsenow's correction term is of negligible importance.

Sparrow and Gregg (4) considered the effects of convection and acceleration in their boundary layer treatment of laminar film condensation on a vertical surface (topics 3.1.4 and 3.1.3, Figure 1). They transformed the conservation of mass, momentum, and energy equations to ordinary differential equations which were then solved numerically. Their results (for fluids exhibiting low values of the

ratio $(\mu c_p)/k$, such as liquid metals) indicated that the numerical value of the heat transfer coefficient can be fifty percent of Nusselt's theoretical prediction. The disagreement is found to increase as the parameter $\lambda/(\Delta T_f c_p)$ is decreased. On the other hand, their results tended to be in good agreement (for fluids where the ratio $(\mu c_p)/k$ is greater than or equal to unity) with theoretical predictions. Their results for materials such as liquid metals are in contrast with the work of Bromley (2) and Rohsenow (3). Implied in these findings by Sparrow and Gregg is the fact that subcooling of, or convection in, a liquid metal condensate produces an increase in the condensate film thickness and a decrease in the heat transfer coefficient. An integral method employed by Mabuchi (5) provided results similar to those obtained by Sparrow and Gregg. On the other hand, Sukhatme and Rohsenow (6) employed a gamma attenuation technique to actually measure the condensate film thickness (topic 3.1.4, Figure 1) for pure, saturated mercury vapor condensing on a vertical tube. From their results, it is indicated that the thickness of the vertically falling liquid metal condensate is described by Nusselt's theory.

The effects on film type condensation of the simultaneous occurrence of convection and acceleration in the condensate, and acceleration or drag in the vapor boundary layer due to interfacial shear have been studied by several researchers (topic 3.1.3, Figure 1). As an example, Koh, Sparrow and Hartnett (7) solved the conservation equations for film condensation in the presence of these effects, (assuming a constant condenser wall temperature and a saturated vapor).

From this study it is found that the effect for liquid metals is essentially the same as that for zero interfacial shear. Koh (8) also approached this same problem employing an integral method. In this analysis he used polynomial expressions for the temperature and velocity profiles to approximate the heat transfer results of film type condensation experiencing a drag-induced vapor boundary layer. This approach provided essentially the same results as obtained by Koh, Sparrow and Hartnett (7). Chen (9) examined the same problem and obtained similar results. In general, the results of these various examinations substantiated Nusselt's theory for values of $(\Delta T_f c_p)/\lambda$ encountered in most experimental work.

Experimental Studies, (topic 4, Figure 1). Heat transfer studies with liquids exhibiting $(\mu c_p)/k$ greater than 0.5 tended to substantiate the modified Nusselt theoretical prediction (10). On the other hand, experimentally determined heat transfer coefficients obtained with liquid metals undergoing film type condensation are significantly lower than predicted by Nusselt's classical theory or by modifications to that theory. For example, nonisothermal experimental studies have been conducted by Misra and Bonilla (11), Cohn (12), Sukhatme and Rohsenow (6), and Roth (13), employing mercury and sodium, mercury and cadmium, mercury, and sodium and rubidium, respectively. Their experimental results ranged from approximately one to fifteen percent of those predicted by theory. In addition to these quiescent vapor studies, nonisothermal investigations of the condensation of flowing sodium inside a vertical tube made by Broglio, Ravelli and Buongiorno (14),

and of potassium inside a vertical tube carried out by Sawochka (15), have also provided results lower than theoretical estimates. Thus, it appears that an incomplete explanation of the heat transfer process for the particular case of liquid metals is provided by the modified Nusselt model. Obviously, one is faced with the problem of providing an explanation of the apparent discrepancies.

Possible explanations of the discrepancies mentioned above include:

1. A film thickness other than that predicted by Nusselt.
2. The existence of systematic deviations in the analysis of experimental data.
3. A diffusion resistance in the vapor near the condensate brought on by noncondensing gases being present in the vapor.
4. A resistance in the condensate near the vapor-liquid interface owing to the presence of impurities on the film surface.
5. A thermal contact resistance resulting from contaminant deposits on the condenser wall.
6. A resistance to phase transition at the vapor-liquid interface.

As previously stated, Sukhatme and Rohsenow (6) found that the condensate film thickness is essentially equivalent to Nusselt's prediction. Thus, the first possibility for reduced heat transfer rates is eliminated on the basis of this experimental work.

Bromley, Brodkey and Fishman (16) studied the effect of temperature variation around a horizontal condenser tube, while Henderson and Marchello (17) examined the role of surface tension and tube diameter

in laminar condensation on horizontal tubes. Petukhov and Yushin (18) found it necessary to consider the heat transmission due to heat conduction along the axis of the condenser tube. These three papers illustrated the existence of minor but systematic deviations during the performance of actual experimental studies.

In general, the heat transfer rate is reduced when noncondensable gases are present in the vapor phase. Sukhatme and Rohsenow (6) demonstrated that the introduction of a noncondensable gas into the mercury vapor provided an increase in pressure and a decrease in the heat transfer rate. It is assumed that this problem is overcome when a pure material is used in an essentially leak-free system which is frequently purged.

The presence of impurities on the condensate surface can be neglected if pure materials are employed in conjunction with relatively short vapor-liquid exposure times. Under experimental conditions other than these, it must be considered as a possible cause of low heat transfer rates.

A resistance at the liquid-solid interface can be attributed to chemical reactions, adsorbed gases and/or the formation of oxide layers between the condensate and wall. This explanation of reduced heat transfer rates is eliminated by the proper selection of materials and the use of pure fluids.

The temperature difference between vapor and condensate needed for bringing about a phase transition results in the appearance of a resistance to phase transition. Reduced heat transfer rates resulting from the presence of a vapor-liquid interfacial thermal resistance are

deemed to be extremely probable with any experimental technique. Wilhelm (19), Barry (20), and Mills (21) studied this problem. Schrage (22) examined the nature of this resistance in some detail. Assuming that heat transfer does not occur between the vapor and liquid phases by conduction, Schrage found that the thermal resistance (or heat flux) is dependent on a so-called "condensation" coefficient. This experimentally determined coefficient is found to decrease with increasing pressure (6,23). Hence, directly related to the heat transfer coefficient determinations mentioned above is the numerical value assigned to the condensation (and/or evaporation) coefficient. Therefore, a discussion of the latter coefficient is deemed obligatory.

Mass Transfer Analysis

Theoretical Basis, (topic 2, Figure 1). Hertz (24), Knudsen (25), and Langmuir (26) are credited with establishing the experimental and theoretical basis for the study of the rate of evaporation from a material. The maximum rate of evaporation (molecules per unit area per unit time) from a surface in contact with its saturated vapor, when both surface and vapor are at the same temperature, is given by the relationship

$$G_{e,max} = \frac{N_s V_s}{4} \quad (4)$$

If the temperature corresponds to vapor pressures that are sufficiently small, the rate of evaporation from the surface into a pure vacuum is expected to depend on the surface temperature in the same manner as it does in the case of evaporation into the saturated vapor. That is, the

rate of evaporation into a vacuum is represented by equation (4). Hertz (24) and Langmuir (26) employed this equation in their respective studies.

On the other hand, Knudsen considered a more general situation in which a so-called transfer coefficient σ is introduced. By way of explanation of this coefficient, a liquid or solid condensate is considered to be in equilibrium with its saturated vapor. Then the rate at which vapor molecules strike the condensate surface is found to be given by equation (4). Assume that a fraction σ of these molecules is condensed on the surface, while a fraction $1-\sigma$ is reflected. In order to maintain a steady state condition, it is necessary that G_e molecules are evaporated from a unit area in unit time, where

$$G_e = \sigma \left[\frac{N_s V_s}{4} \right] = \sigma G_{e,\max} \quad (5)$$

Since σ is defined as a fractional value, it follows that $G_e = G_{e,\max}$ when $\sigma = 1$. Under this condition G_e is referred to as the "theoretical maximum" rate of evaporation. In practice, an apparent rate of evaporation $G_{e,\exp}$ is determined while $G_{e,\max}$ is calculated. The evaporation coefficient is determined from the ratio

$$\sigma = \frac{G_{e,\exp}}{G_{e,\max}} \quad (6)$$

When a saturated vapor and its condensate exist in equilibrium the rates of evaporation and condensation are equivalent and the evaporation and condensation coefficients are numerically equal. In this

steady state situation, the density of the vapor is assumed to be unchanged during the transfer process. On the other hand, it is possible when both phases are stationary and long condensate-vapor contact times are employed, for a vapor cloud to form above the condensate. This cloud results when some of the evaporated molecules are not immediately removed. Thus, the net rate of evaporation is given by

$$G_{\text{net}} = \sigma \left[\frac{V(N_v - N_s)}{4} \right] \quad (7)$$

where

$$V_s = V_v = V \quad (8)$$

Included in equation (7) is the general case in which the material does not evaporate into a vacuum, but shows an actual vapor pressure $N_v < N_s$ at the condensate-vapor interface. Furthermore, it is assumed that the vapor molecules are identical in type to those on the condensate surface (a pure substance), and that no process occurs which changes the molecular weight or energy content of the molecules (such as association, dissociation, polymerization, etc.). It is also assumed that the density of the vapor cloud is low enough that the emitted molecules do not encounter a significant diffusional resistance.

Liquid-Vapor Models, (topic 5, Figure 1.). Recently, several models have been employed and/or proposed by researchers in an attempt to adequately describe the liquid-vapor interfacial transport phenomena. Thus, it is necessary to present these models and the respective underlying interpretation of each model. For all models, the condensation

process is described in terms of a pure saturated vapor condensing on its own liquid phase.

The film type condensation model to be considered is the so-called " $T_i = T_v$ " model shown in Figure 3. Employing the kinetic theory approach, the interphase mass transfer process for the " $T_i = T_v$ " condensation model is considered to be the difference between the rate of departure of molecules from the surface of the condensate film into the vapor space and the rate of arrival of molecules from the vapor space towards the interface. The condensation process is realized when the latter rate exceeds the former. Implied in this statement is the existence of an interfacial temperature drop. (Nusselt assumed that this temperature drop is negligible compared to that of the temperature drop across the condensate film.)

Employing a Maxwell-Boltzmann distribution, it has been shown (27, 28) that, when a uniform gas (with no mass motion) is bounded by a fixed surface, the mass of the molecules that pass in either direction (to the right or left) through a unit area of an imaginary plane in unit time is given by (when a real gas obeys the perfect gas model)

$$\frac{m N}{A t} = \frac{P}{\sqrt{2} \pi R T} \quad (9)$$

Schrage (22) assumed that equation (9) can be used to describe the motion of molecules at the liquid-vapor interface. In the case of condensation, there is assumed to exist a progress velocity V_x toward

Figure 3. The " $T_i = T_v$ " condensation model

the surface which is negative for condensation

$$\frac{m N}{A t} = \rho V_x \quad (10)$$

and equation (9) becomes for these molecules

$$\frac{m N}{A t} = \Gamma \frac{P_v}{\sqrt{2 \pi R T_v}} \quad (11)$$

which is negative for condensation and where

$$\Gamma = \left[\exp (-\phi^2) \right] + \phi \pi^{1/2} \left[1 + \operatorname{erf} \phi \right] \quad (12)$$

and

$$\phi = - \frac{V_x}{\sqrt{2 R T_v}} \quad (13)$$

or

$$\phi = - \frac{\left[\frac{1}{\rho_v} \right] \left[\frac{m N}{A t} \right]_{\text{net}}}{\sqrt{2 R T_v}} \quad (14)$$

As previously stated, it is assumed that some of the vapor molecules which strike the surface are reflected back into the vapor space. Thus, the fraction of those molecules striking the surface which do condense is called the "condensation coefficient" (σ_c). Similarly, the "evaporation coefficient" (σ_e) is defined as the ratio of the flux of molecules actually evaporated from the condensate surface to the maximum value given by equation (9).

Schrage (22) visualized the pure saturated vapor (defined by T_v, P_v) moving toward the condensate surface as given by

$$\left[\frac{m N}{A t} \right]_c = - \frac{\sigma_c \Gamma P_v}{\sqrt{2 \pi R T_v}} \quad (15)$$

while the molecular counter flow (defined by T_s, P_s) from the condensate surface is given by

$$\left[\frac{m N}{A t} \right]_e = \frac{\sigma_e P_s}{\sqrt{2 \pi R T_s}} \quad (16)$$

Thus, during the condensation process, the net mass flux toward the liquid-vapor interface is given by the difference

$$\left[\frac{m N}{A t} \right]_{net} = - \frac{\sigma_c \Gamma P_v}{\sqrt{2 \pi R T_v}} + \frac{\sigma_e P_s}{\sqrt{2 \pi R T_s}} \quad (17)$$

or

$$G_{net} = \sigma_c \Gamma G_c + \sigma_e G_e \quad (18)$$

where for condensation

$$|\sigma_c \Gamma G_c| > |\sigma_e G_e| \quad (19)$$

and G_c is negative (defined), Γ is a positive correction factor, G_e is positive (defined). Thus, G_{net} is negative for the condensation process to take place. From equations (15) and (18)

$$G_c \equiv \frac{-P_v}{\sqrt{2 \pi R T_v}} \quad (20)$$

$$G_e \equiv \frac{P_s}{\sqrt{2 \pi R T_s}} \quad (21)$$

Under the assumption that the perfect gas model is obeyed by the real vapor, substitution of equation (14) into (12) yields (for $|\phi| < 10^{-3}$)

$$\Gamma \approx 1 - \left\{ \frac{\left[\frac{m N}{A t} \right]_{\text{net}}}{P_v \sqrt{\frac{2}{\pi R T_v}}} \right\} \quad (22)$$

or

$$\Gamma \approx 1 - \left\{ \frac{G_{\text{net}}}{P_v \sqrt{\frac{2}{\pi R T_v}}} \right\} \quad (23)$$

Hence, the condensation mass balance given by equation (17) or (18) becomes

$$G_{\text{net}} = - \frac{\sigma_c P_v}{\sqrt{2 \pi R T_v}} + \frac{\sigma_c G_{\text{net}}}{2} + \frac{\sigma_e P_s}{\sqrt{2 \pi R T_s}} \quad (24)$$

under the assumption that

$$\sigma_c = \sigma_e = \sigma \quad (25)$$

equation (24) can be simplified to read

$$G_{\text{net}} = - \left\{ \left[\frac{2 \sigma}{2 - \sigma} \right] \left[\frac{1}{\sqrt{2 \pi R}} \right] \left[\frac{P_v}{\sqrt{T_v}} - \frac{P_s}{\sqrt{T_s}} \right] \right\} \quad (26)$$

The net mass transfer flux relationship presented in equation (26) is "derived" in other phenomenological interpretations yet to be considered in this section. Both Wilhelm (19) and the author question the "equality of transfer coefficients" assumption which is required to obtain equation (26), on the basis that a physical consideration is lacking to support this assumption in the case where G_{net} does not vanish. The validity of this assumption is to be considered by the author in his experimental work.

Under the assumption of a pseudo isothermal phase change (that is, $T_v = T_s$), equation (26) can be written

$$G_{\text{net}} = - \left\{ \left[\frac{2 \sigma}{2 - \sigma} \right] \left[\frac{1}{\sqrt{2 \pi R T_v}} \right] \left[P_v - P_s \right] \right\} \quad (27)$$

In another phenomenological interpretation (topic 5.2, Figure 1) Kucherov and Rikenglaz (29, 30) examined the directed phase transition model (Figure 3) which takes into account the convective motion of the vapor which accompanies condensation, as well as the temperature (pressure) discontinuity which prevails at the liquid-vapor interface. Their study is restricted to the case where the rate of vaporization is considerably less than that into a vacuum at the same temperature. (That is to say, the velocity of convective motion of the vapor is much less than the molecular velocity.) Consideration of these

effects, while assuming a Maxwell-Boltzmann velocity distribution for molecules evaporating from the condensate, led to an equation for the resultant mass flow of molecules during condensation on a liquid surface identical to that presented in equation (24) where $\sigma_c = \sigma_e = \sigma$. Their expansion of this net mass flux relationship resulted in the approximation

$$G_{net} \approx - \left[\frac{2\sigma}{2 - \sigma} \right] \left[\frac{1}{\sqrt{2\pi R T_v}} \right] \left\{ \left[P_v - P_s \right] - \frac{P_s}{2} \left[\frac{T_v - T_s}{T_s} \right] \right\} \quad (28)$$

An irreversible thermodynamics approach performed by Bornhorst and Hatsopoulos (31) constitutes the third film condensation interpretation (topic 5.2, Figure 1) to be considered. Employing the model shown in Figure 4 for the case of small transfer rates, they considered the mass, energy and momentum flux at the interface to describe the net condensation mass transfer flux by

$$G_{net} \approx - \left[\frac{2\sigma}{2 - \sigma} \right] \left[\frac{1}{\sqrt{2\pi R T_{vi}}} \right] \left\{ \left[P_{vi} - P_{si} \right] - \frac{P_{si}}{2} \left[\frac{T_{vi} - T_{si}}{T_{si}} \right] \right\} \quad (29)$$

and the interfacial thermal gradient by

$$\left[\frac{dT}{dx} \right]_{x=0} = - \left[\frac{c_p}{k} \right]_v \left[T_v - T_{vi} \right] G_{net} \quad (30)$$

for a real gas which can be described by the perfect gas model. The momentum effect is neglected in their linear analysis, on the basis that it appeared as a second order term in the mass flux relationship which provided a negligible contribution. It is important to notice in equation (29) that Bornhorst and Hatsopoulos assumed that the temperature of the vapor and liquid molecules at the interface need not necessarily be the same as those in the "bulk" phase.

Fedorovich and Rohsenow (32) employed the condensation model shown in Figure 5 to analyze the interfacial transfer process (topic 5.2, Figure 1). An energy balance on the control volume bounded by planes at $x=i$ and one in the bulk vapor, yielded the interfacial thermal gradient

$$\left[\frac{dT}{dx} \right]_{x=i} = \left[\frac{c_p}{k} \right] [T_v - T_{vi}] G_{net} \quad (31)$$

where the net mass transfer is given by

$$G_{net} = \left[\frac{2\sigma}{2-\sigma} \right] \left[\frac{1}{\sqrt{2\pi R T_{vi}}} \right] [P_{vi} - P_s] \quad (32)$$

The condensation heat flux (q_c) is given by

$$q_c = - G_{net} \left\{ \lambda + 0.68 \left[C_1 (T_s - T_w) \right] \right\} \quad (33)$$

The temperature jump coefficient (ξ) and interphase zone thickness (d) shown in Figure 5 are contained in the expression

Figure 5. The Fedorovich-Rohsenow liquid-vapor interface model

$$\left[\frac{T_v - T_{vi}}{T_{vi} - T_s} \right] = \left[\frac{k}{c_p} \right]_v \left[\frac{1}{(\xi + d) G_{net}} \right] \quad (34)$$

In their analysis, Fedorovich and Rohsenow assumed that conduction need only be considered in the flow to the condensate surface (thereby neglecting the convection effect). Here again, the model is limited by the assumption of small transfer rates.

All of the models mentioned above are classified as being phenomenological interpretations (topic 5.2, Figure 1) because experimental results are required to complete the analysis for each model. From an examination of these various interfacial transport models, it is seen that they differ with respect to the thickness of the interphase zone and/or the temperatures required to describe the transfer process. In the kinetic theory model (such as shown in Figure 3), it is assumed that the "bulk" vapor conditions prevail up to the interface, i.e., the interphase zone is finite but extremely thin. This, in turn, is based on the assumption that interactions between condensing, evaporating, and reflecting molecules do not alter the velocity distribution and density of the vapor adjacent to the condensate surface. On the other hand, the Bornhorst-Hatsopoulos and Fedorovich-Rohsenow models relax the " $T_v = T_i$ " assumption, as well as suggesting that the interphase zone thickness is greater than that proposed in the " $T_v = T_i$ " model. An implication as to the validity of each model is obtained later when their use is considered in conjunction with experimental studies.

To this author's knowledge, Wilhelm (19) is the only researcher to attempt a theoretical interpretation (topic 5.1, Figure 1) of the interfacial mass transfer process. In this kinetic theory approach, using the model shown in Figure 6, the condensation and evaporation coefficient, condensing heat flux, and temperature of the condensate surface are expected to be uniquely defined by the fluid under investigation, temperature of the bulk vapor, and the condenser wall temperature. Governing the phase change process for a liquid metal are the conservation of mass, momentum, and energy, as well as the normalization of the composite velocity distribution function. A fifth equation is provided by the incorporation of the perfect gas model restriction. As stated by Wilhelm, the solution is a function of the assumed form of the velocity distribution function, and the perfect gas model is applicable only when the distribution function is Maxwellian. Now, for various combinations of the condensing heat flux (q/A) and vapor temperature (T_v), the above mentioned system of five equations can be solved simultaneously for T_s , σ_c and σ_e . A further restriction on the solution is that it must meet the requirement that the value of the condensation and evaporation coefficient be within the range zero to unity. If the above mentioned equations and model of the kinetic theory of condensation adequately describe the condensation heat transfer, they are expected to provide physically consistent solutions.

In his first examination of the model, Wilhelm assumed that the velocity distribution function associated with pure mercury is

Figure 6. The vertical " $T_i = T_v$ " condensation model

Maxwellian. These results indicated that a consistent solution existed only during thermodynamic equilibrium ($G_{\text{net}} = 0$) and for the case where the temperature difference ($T_v - T_s$) is less than a few degrees. The computer determined results also provided multiple solutions. Wilhelm concluded that these results implied that the Maxwellian velocity distribution function employed failed to describe the process when coupled with the requirement that the density of the vapor up to the interface is considered to be constant.

Next, Wilhelm relaxed the requirement of "constant density" by considering the conservation of mass, momentum, and energy equations for a perfect gas. Obviously, relaxing the requirement of constant density failed to reduce the number of unknown quantities while decreasing the number of governing equations. Under these conditions it is necessary to provide additional assumptions, such as requiring that the evaporation coefficient be set equal to unity; the evaporation and condensation coefficients be equal; and condensation favors molecules of a particular energy range. For all cases considered, Wilhelm's analyses resulted in multiple solutions and failed to provide a systematic trend.

Wilhelm also undertook a parametric study (topic 5.2, Figure 1) by employing the conservation of mass, momentum, and energy equations, while examining the effects of systematically varying the temperature of the liquid surface and evaporation coefficient for various combinations of the vapor temperature and condensation heat flux. As with other phenomenological interpretations, unique solutions of the

condensing heat transfer problem are not realized with this parametric study. On the other hand, trends in the value of the condensation coefficient are observed. Associated with the film condensation process governed by the relationship $(\sigma_c \Gamma G_c) \gg (\sigma_e G_e)$, for a given vapor temperature and pressure (T_v, P_v), is the existence of an obvious minimum value of the condensation coefficient (σ_c) required to sustain a constant heat flux. This minimum occurred either when the evaporation coefficient is approximately zero, or when the temperature of the vapor (T_v) is much larger than that of the condensate surface (T_s). It is also observed that the minimum condensation coefficient values (for a given heat flux) decreased as the vapor pressure (P_v) increased. This result is expected from the kinetic theory because of the corresponding increase in the total number of molecules striking the interface. Other trends in the condensation coefficient observed by Wilhelm include:

1. When the heat flux, temperature of the vapor, and surface temperature of the condensate film are held constant, an increase in the value of the evaporation coefficient resulted in a corresponding increase in the condensation coefficient in order to maintain a constant heat flux.
2. When all other factors are held constant, a decrease in the condensation coefficient is the resultant of a decrease in the heat flux.
3. For a fixed heat flux and bulk vapor temperature, and a given evaporation coefficient, the condensation coefficient must increase as the temperature of the condensate film surface approaches the

bulk vapor temperature in order to compensate for the increased evaporation as well as to maintain a constant heat flux. (It is also observed that a change in the surface temperature of the condensate film is realized when the condensation coefficient changes.)

4. From a comparison of several values of the heat flux at the same bulk vapor temperature and pressure (when $G_{\text{net}} \ll \Gamma G_c$), it is seen that values of the condensation coefficient are essentially independent of the heat flux, but are a function of the evaporation coefficient and the surface temperature of the condensate film.

Experimental Studies, (topic 4, Figure 1). Experimental studies employing the previously mentioned models of the liquid-vapor interface (topic 5, Figure 1) provided a variety of values for the transfer (evaporation and/or condensation) coefficient. It is seen that these values tend to be a function of the particular model employed. To illustrate this trend it is necessary that only a few of the recent experimental studies be considered.

Sukhatme and Rohsenow (6) examined the film condensation of mercury on a vertical nickel tube. They undertook special precautions to eliminate noncondensable gases as well as surface contamination from the experimental setup. Employing the " $T_i = T_v$ " condensation model shown in Figure 3 and equation (26), they obtained values of the "condensation" coefficient ranging from 0.365 to 0.605. Expecting this coefficient to exhibit a value of unity for a pure material, they concluded that the experimental values indicated the existence of some

form of contamination on the condensate surface.

Kroger and Rohsenow (23) studied the film condensation of saturated potassium vapor on a vertical surface of nickel as well as stainless steel. Here again, a clean and leak tight (10^{-3} inch of mercury or less for four days) system is employed to obtain numerical values of the "condensation" coefficient ranging from 0.061 to 0.670 for bulk vapor pressures (P_v) of 0.6 to 0.01 atmosphere (absolute). Employing the same condensation model ($T_i = T_v$) and approach as used by Sukhatme and Rohsenow (6), their results indicated that the "condensation" coefficient decreases approximately linearly with increasing pressures (P_s) associated with the surface temperature of the condensate film (T_s).

Subbotin, Ivanovskii, Sorokin and Chulkov (33) described the net mass transfer associated with the film condensation of a pure stationary potassium vapor on a cylindrical steel condenser by equation (28). Their study indicated that the "condensation" coefficient for a pure liquid metal in a clean and leak tight experimental setup is essentially unity in the saturation pressure range of $0.001 \lesssim P_s \lesssim 0.006$ atmosphere (absolute). Remembering that equation (28) is the same as equation (24) when $\sigma_c = \sigma_e = \sigma$, their results tended to imply that the "transfer" coefficients (σ_e and σ_c) are numerically equal. Employing a movable thermocouple probe, they measured the temperature distribution across the vapor and condensate film (up to the condenser wall). From these data, temperature discontinuities at the liquid-vapor interface tended to exhibit a " $T_i = T_v$ " rather than a " $T_i \neq T_v$ " model in

the range $336 \lesssim T_v \lesssim 402$ degrees centigrade for discontinuities of $2 \lesssim (\Delta T) \lesssim 6$ degrees centigrade. On the other hand, one study conducted at a bulk vapor temperature of 640 degrees centigrade and a temperature discontinuity of less than 1.5 degrees centigrade tended to substantiate the existence of the " $T_i \neq T_v$ " film condensation model.

Using basically the same approach and technique as described above [Subbotin et al. (33)], Ivanovskii, Milovanov and Subbotin (34) found the condensation coefficient for mercury to range from 0.71 to unity for liquid-vapor exposure times ranging from 5 to 2 minutes. Since diffusion resistances related to vapor contamination are excluded by the direct measurement of the temperature distribution across the vapor and condensate, they reasoned that the deviation in values is caused by basic nonuniformity of the purity of the mercury condensate surface from one experiment to the next. This is confirmed by a plot of the experimentally determined condensation coefficient as a function of the liquid-vapor interaction time which showed that the values of essentially unity corresponded to the shortest exposure times where the mercury condensate exhibited a high degree of purity.

The film condensation studies of a pure saturated potassium vapor ($0.0139 \leq P_v \leq 0.5662$ atmospheres) on a horizontal stainless steel surface performed by Meyrial, Morin and Rohsenow (35) assumed the validity of the Fedorovich-Rohsenow vapor subcooling ($T_i \neq T_v$) model shown in Figure 5. Employing equation (32) and an interphase zone thickness (d) of five mean free path lengths, they found the "condensation" coefficient to be essentially unity over the saturation pressure

range $0.0120 \leq P_s \leq 0.4970$ atmospheres. Furthermore, their application of the " $T_i \neq T_v$ " model to studies previously performed by Sukhatme and Rohsenow (6), Misra and Bonilla (11), Barry (20), and Kroger and Rohsenow (23), and consideration of the values reported by Subbotin, Ivanovskii, Sorokin and Chulkov (33) tended to demonstrate that the "condensation" coefficient is unity over the range of saturation pressures $0.003 \lesssim P_s \lesssim 1.0$ atmosphere. This result for a pure saturated material is anticipated from an examination of Paul's (36) tabulated values for other fluids. Herein, water is used to illustrate the experimental film condensation studies conducted with other materials.

According to Mills (21), Paul (36), and Mills and Seban (37), studies undertaken to determine the condensation and/or evaporation coefficient associated with water have provided numerical values ranging from 0.006 to unity. In general, the experimental techniques employed have involved evaporation from a suspended drop, evaporation (condensation) from (on to) a horizontal surface, and evaporation (condensation) from (on to) a fluid jet. Concerning the first two techniques mentioned above, the major experimental problem (arising from the large temperature gradient normal to the surface) is the determination of the evaporating surface temperature. Values of the "transfer" coefficient obtained from experiments employing a water jet are questioned on the basis that the surface temperature of the jet is estimated rather than being measured. Besides these two, other explanations of the range of values include contamination of the surface,

the existence of a noncondensable gas in the vapor phase, the values are correct as reported and are a function of temperature, etc.

In 1954, Hickman (38) determined the evaporation coefficient of water by examining the rate at which molecules leave the surface of a water jet passing through an evacuated region. In this experimental setup the mass of the "released" molecules collected in a cold trap for a given time period provides the evaporation rate. In order to carry out the rate determination, the researcher must know such facts as the area of the evaporating surface, liquid density, velocity of the jet, temperature of evaporating surface, duration of evaporation (exposure time), and the amount of liquid evaporated. Owing mainly to an inability to accurately determine the temperature of the evaporating surface, Hickman concluded that a clean, new surface of water exhibits an evaporation coefficient greater than 0.25.

Jamieson (39) employed an experimental setup similar to that used by Hickman (38) to determine the condensation coefficient. In this particular study, a clean jet of non-radioactive water is passed through tritiated steam, thereby allowing the researcher to differentiate condensed "steam" molecules from those originally in the stream. Employing this radioactive tracer technique, Jamieson concluded that the condensation coefficient of water is 0.35. In an analysis of the experimental procedure and resulting coefficient, Jamieson considered the possibility of condensed tagged molecules being evaporated back into the steam, dilution of the tritiated vapor with non-radioactive molecules from the jet, selective condensation due to the mass of

tritium being threefold greater than that of protium, entrainment of tritiated vapor in the liquid, employing the average jet velocity to describe the surface velocity, possible contamination of the liquid surface, and changes in the surface tension. After a rather careful examination of this list of possible explanations, Jamieson concluded that (even though he failed to observe a systematic change in the value of the condensation coefficient with changes in jet velocity) it may be possible for the condensation coefficient to vary with the velocity or rate of shear at the liquid surface. This author is of the opinion that the above mentioned list should also have included the fact that the liquid surface temperature is estimated rather than measured. The saturation pressure associated with this temperature is directly related to the calculated condensation coefficient.

Nabavian and Bromley (40) studied the rapid condensation of pure steam on the external grooved surface of a horizontal pure copper tube. Cooling of the condenser is accomplished by high velocity cold water flowing through it. In this experiment, the condensation coefficient is determined from the relationship

$$h_i = \left(\frac{2 \sigma}{2 - \sigma} \right) \left(\frac{\lambda^2}{T_s T_v} \right) \left(\frac{1}{\sqrt{2 \pi R T_{ave}}} \right) \quad (35)$$

where the interfacial heat transfer coefficient between the vapor and liquid condensate (h_i) is calculated from the heat transfer coefficient sum

$$\frac{1}{h_i} = \frac{1}{U} - \frac{1}{h_c} - \frac{1}{h_w} - \frac{1}{h_1} \quad (36)$$

where U , h_c , h_w , and h_1 represent the measured overall heat transfer coefficient, calculated inside film coefficient (inside of the condenser pipe), calculated heat transfer coefficient due to the pipe wall, and calculated condensate film coefficient, respectively. Owing to the fact that a range of values is determined for both h_w and h_1 , Nabavian and Bromley concluded that the condensation coefficient is described as existing within the range 0.35 to unity.

The basic experimental setup and analysis employed by Mills and Seban (37) to determine the condensation coefficient is similar to that of Nabavian and Bromley (40). Here again, the researchers found it impossible to determine an exact numerical value for the coefficient due to the inherent limitation in the technique, and hence deduced that it has a value between 0.45 and unity. On the other hand, Mills (21) and/or Mills and Seban (37) examined previous studies of the condensation coefficient of water. Reanalysis of the experiments performed by Hickman (38) and Jamieson (39) involving an analytical calculation of the jet surface temperature provided a different temperature which resulted in a condensation coefficient essentially equal to unity. Thus, it is implied from these studies that the condensation coefficient for pure, saturated water is unity.

A condensation coefficient of 0.83 ± 0.15 is reported by Koros, Deckers, Andres and Boudart (41) from their analysis of a jet of water molecules impinging on a target of ice suspended by a balance in a vacuum chamber. In this so-called direct method, the condensation coefficient is determined from the increased mass of the target. A major

experimental difficulty encountered with this technique is an inability to measure the beam intensity. Realizing this, the researchers developed a technique whereby this quantity is eliminated. To accomplish this, a cylindrical reflector is added around the target in an attempt to focus once-reflected molecules back onto the ice. Since the cylinder failed to provide perfect reflection, they found themselves forced to estimate the fraction of molecules lost from the experimental setup.

Summary

Nusselt's classical analysis, the modified Nusselt theory, and the more basic mass transfer approach have been used to determine the film type condensation heat transfer characteristics of a given system. Experimental studies have demonstrated that even though small but systematic deviations are known to exist, the empirical equations resulting from a Nusselt type analysis provide sufficiently accurate information for coolants employed in common engineering heat transfer applications. With the advent of the nuclear power plant and its design requirements that the coolant exhibit relatively low vapor pressures at high temperatures, not be affected by nuclear radiation, etc., engineers have examined liquid metals as a heat transfer agent. These experimental studies showed that Nusselt's classical analysis failed to provide sufficiently accurate heat transfer information, and that the theoretical-experimental discrepancies are not remedied

by merely accounting for Nusselt's basic assumptions. In an attempt to provide a model which predicted the heat transfer characteristics of liquid metals, researchers examined the basic mass transfer approach. Employment of this particular approach required that the condensation and evaporation coefficients (which are related to the heat transfer coefficient) be known. Experimentally determined values of the condensation and/or evaporation coefficients reported in previously mentioned papers are tabulated in Tables 1 and 2.

TABLE 1

A SUMMARY OF THE EXPERIMENTALLY DETERMINED
TRANSFER COEFFICIENTS FOR WATER

Author(s)	Ref.	Transfer Coefficient	Experimental Technique
Hickman	38	$> 0.25^a$	Evaporation from a liquid jet into a vacuum
Jamieson	39	0.35^b	Condensation of steam onto a liquid jet
Nabavian and Bromley	40	Between 0.35 and unity	Condensation on a horizontal cylinder
Mills and Seban	37	Between 0.45 and unity	Condensation on a horizontal cylinder
Koros et al.	41	0.83	Condensation on ice from a liquid jet

^aReanalysis by Mills and Seban (37) indicated that the transfer coefficient is approximately unity.

^bReanalysis by Mills (21) indicated that the transfer coefficient is approximately unity.

TABLE 2

A SUMMARY OF THE EXPERIMENTALLY DETERMINED
TRANSFER COEFFICIENTS FOR LIQUID METALS

Author(s)	Ref.	Metal	Transfer Coefficient	Experimental Technique
Sukhatme and Rohsenow	6	mercury	0.365 to 0.605 ^a	Condensation on a vertical cylinder
Kroger and Rohsenow	23	potassium	0.061 to 0.670 ^a	Condensation on a vertical flat surface
Subbotin et al.	33	potassium	~ 1	Condensation on a horizontal cylinder
Ivanovskii et al.	34	mercury	0.71 to 1	Condensation on a horizontal flat surface
Meyrial et al.	35	potassium	~ 1	Condensation on a horizontal flat surface
Misra and Bonilla	11	mercury	0.02 to 0.61 ^{b,a}	Condensation on a vertical cylinder
Barry	20	sodium	0.04 to 0.15 ^a	Condensation on a vertical cylinder

^aReanalysis by Meyrial et al. (35) using the Fredorovich-Rohsenow Model indicated that the transfer coefficient ranged from 0.42 to 1 (Sukhatme and Rohsenow), is essentially unity (Kroger and Rohsenow), ranged from 0.13 to 1 (Misra and Bonilla), and ranged from 0.24 to 0.41 (Barry).

^bCalculated by Wilhelm (19) from the experimental data and results of Misra and Bonilla using the " $T_i = T_v$ " model.

CHAPTER III

DISSERTATION RATIONALE

Generally speaking, the purpose of this research is to provide a unique experimental procedure whereby both the condensation and evaporation coefficients can be determined, as well as one which possesses relative freedom from experimental difficulties, and allows examination under nonisothermal conditions or isothermal equilibrium of the vapor-liquid phases.

The tabulated results contained in Tables 1 and 2 tend to imply that the so-called transfer coefficient for any pure, saturated vapor is essentially unity. In each of these nonisothermal experimental studies the listed transfer coefficient is obtained by assuming that the condensation and evaporation coefficients are numerically equal. This "aide-toi, le ciel t'aidera" assumption is questioned for the non-isothermal case, and its validity is to be examined by the author employing the radioactive tracer technique to determine both of these coefficients.

Employment of the Fedorovich-Rohsenow liquid-vapor interface model has implied that the transfer coefficient for liquid metals is essentially unity when the interphase zone is approximately ten mean free path lengths (related to vapor molecules) thick (Table 2). Owing to a movable thermocouple probe being incorporated into the author's

experimental setup it is possible to measure the temperature distribution across the bulk vapor, vapor-liquid interface and into the bulk liquid. These temperature measurements permit an examination of the possible existence of an interphase zone as well as an interface temperature (T_i) different than that of the bulk vapor (T_v). Furthermore, the possibility of diffusion resistances related to vapor contamination are excluded by these direct measurements of the temperature distribution across the vapor and liquid.

Nonisothermal liquid metal experimental studies have employed a stationary vapor undergoing film type condensation on some type of surface (Table 2). This type of experimental setup has suffered from the possible existence of a thermal contact resistance resulting from contaminant deposits on the condenser wall. On the other hand, the nonisothermal experimental setup to be employed by the author does not employ a condenser. The setup consists of a pure, saturated stationary vapor condensing on a cooler circulating liquid. Thus, the possible existence of a thermal contact resistance is eliminated by removing the condenser from the experimental setup.

It is possible to examine the transport phenomena at a liquid-vapor interface either under the nonisothermal condition or isothermal equilibrium. Herein, the two cases are considered employing a radio-nuclide tracer technique. Prior to performing the nonisothermal experiment, the feasibility of the radioactive tracer technique is demonstrated by conducting an isothermal equilibrium study. The non-isothermal and isothermal studies are similar in that differentiation

of the vapor and liquid molecules is accomplished by initially employing normal (nonradioactive) mercury in one phase and a radioactive tagged (Hg-197 and/or Hg-203) mercury in the other phase. Owing to the fact that the same experimental setup is not employed for both studies, the two cases are treated independently (in the following chapters). Afforded by this particular approach is an unambiguous presentation of the two cases.

CHAPTER IV

ISOTHERMAL EQUILIBRIUM STUDY

The isothermal equilibrium study presented below is essentially that reported by Wylie and Dryden (42) employing a radioactive tracer to examine the transport phenomena at the liquid-vapor interface of mercury.

Isothermal Equilibrium Model

Consider a hypothetical experimental system (Figure 7) immersed in a constant temperature bath. Initially, subsystem 1 is filled with normal (nonradioactive) mercury, subsystem 2 is evacuated, and subsystem 3 is filled with radioactive (tagged with Hg-197 and/or Hg-203) mercury. Molecular exchange between any two or all three of the subsystems is prevented by the existence of diaphragms 1-2 and 2-3. The exchange process between subsystems 1 and 2 is commenced when diaphragm 1-2 is removed. After an equilibrium condition is attained (as determined by the vapor pressure of mercury) the liquid and vapor phases are examined over an arbitrary time interval Δt . During this time interval, the increase in the number of molecules in the vapor phase due to evaporation is

$$\sigma_e G_e A \Delta t.$$

Figure 7, A hypothetical isothermal experimental system

Since the vapor volume is maintained at a constant temperature, the number of molecules in the vapor is a constant. Thus, those molecules evaporated from the liquid phase have replaced "vapor" molecules which have undergone condensation. The decrease in the number of "vapor" molecules due to condensation is

$$\sigma_c G_c A \Delta t.$$

At equilibrium, the number of evaporated molecules is equivalent to the number of condensed molecules. Thus,

$$\sigma_e G_e A \Delta t + \sigma_c G_c A \Delta t = 0 \quad (37)$$

or

$$\sigma_e G_e + \sigma_c G_c = 0 \quad (38)$$

Equation (38) is transformed from a molecular to a mass relationship by multiplication of the mass per molecule. This isothermal equilibrium mass transfer relationship is a special case of equation (18), employed by Schrage (22) to describe the interfacial mass transfer process under nonisothermal conditions, where neither a net mass flux nor progress velocity exist. From equations (12) and (13), it is seen that a correction term (Γ) of unity is implied by the lack of a progress velocity. In equation (38), the condensation and evaporation mass flux terms are defined by equations (20) and (21), respectively. Since the temperatures as well as the pressures are equal (under isothermal

equilibrium)

$$G_e = G_c = G. \quad (39)$$

Thus, implied from equations (38) and (39)

$$\sigma_e = \sigma_c = \sigma \quad (40)$$

The equilibrium mass exchange process between subsystems 1 and 2 is halted when diaphragm 1-2 is returned to its original location.

Now, a saturated vapor is contained in subsystem 2. As previously stated, the number of molecules per unit vapor volume (c_T) is determined completely by the vapor pressure of mercury, and is numerically constant. The ratio of tagged molecules to the total number of liquid molecules in subsystem 3 is represented by x_1 .

Exchange between tagged liquid and normal vapor mercury molecules is realized by removal of diaphragm 2-3. The instant at which this diaphragm is removed is designated as zero time. Now that the exchange process has started, it is necessary to consider the increase in tagged molecules in the vapor. The concentration of these molecules (molecules per unit vapor volume) in the vapor phase at any arbitrary instant (greater than zero time) is represented by c_v . The molecular increase and decrease in the numerical value of c_v during the time interval dt is given by

$$\sigma_e G_e A x_1 dt \quad (41)$$

and

$$\sigma_c G_c A \frac{c_v}{c_T} dt \quad (42)$$

respectively. The ratio c_v/c_T is simply the fraction of radiomercury to the total number of molecules in the vapor at time t . A molecular balance on the radiomercury in the vapor phase is

$$(V) \quad dc_v = (\sigma_e G_e A x_1) dt - (\sigma_c G_c A \frac{c_v}{c_T}) dt \quad (43)$$

where (for isothermal equilibrium) the molecular transfer flux as well as the transfer coefficient values are equivalent. Thus, upon rearranging equation (43)

$$\left[\frac{\sigma G A}{V c_T} \right] dt = \left[\frac{1}{x_1 c_T - c_v} \right] dc_v \quad (44)$$

Under the assumption that x_1 is (for all practical purposes) numerically constant over the time duration of the study, and employing the boundary condition that at $t = 0$, $c_v = 0$ and $t = t$, $c_v = c_v$, equation (44) can be integrated to

$$\left[\frac{\sigma G A}{V c_T} \right] t = - \left\{ \ln \left[1 - \frac{c_v / c_T}{x_1} \right] \right\} \quad (45)$$

or

$$\left[\frac{c_v / c_T}{x_1} \right] = \left\{ 1 - \left[\exp \left(- \frac{\sigma G A t}{V c_T} \right) \right] \right\} \quad (46)$$

or

$$\left[\frac{x_v}{x_1} \right] = \left\{ 1 - \left[\exp \left(- \frac{t}{\tau} \right) \right] \right\} \quad (47)$$

where

$$x_v \equiv \frac{c_v}{c_T} \quad (48)$$

and

$$\frac{1}{\tau} \equiv \frac{\sigma G A}{V c_T} \quad (49)$$

This particular equilibrium exchange process between subsystems 2 and 3 is terminated when diaphragm 2-3 is returned to its original location. Following this, the subsystems are brought to room temperature and the activity per unit mass of the mercury collected from each is measured. These measured quantities, a_v and a_1 are related to x_v and x_1 , respectively. For example, consider the mass (m_v) collected from subsystem 2. The activity of this mass of mercury is represented by A_v (counts per unit time). The total activity of the Hg-197 and Hg-203 radionuclides (A_v) in the vapor phase is

$$A_v = c_7 \beta_7 N_7 + c_3 \beta_3 N_3 \quad (50)$$

where c , β , and N represent the detector efficiency, fraction of radioactive atoms that disintegrate per unit time, and number of radioactive atoms present, respectively. The subscripts 7 and 3 refer to the two radioactive species Hg-197 and Hg-203. Employing the fact that

$$\left. \begin{aligned} N_7 &= \frac{m_7 N^*}{A_7} \\ N_3 &= \frac{m_3 N^*}{A_3} \end{aligned} \right\} \quad (51)$$

and

$$\left. \begin{aligned} m_7 &= m_v x_7 \\ m_3 &= m_v x_3 \end{aligned} \right\} \quad (52)$$

where m , N^* , A , and x represent the mass, Avogadro's number, mass number, and mass fraction, respectively. Equation (50) can be given as

$$\frac{A_v}{m_v} = c_7 \beta_7 \left[\frac{x_7 N^*}{A_7} \right] + c_3 \beta_3 \left[\frac{x_3 N^*}{A_3} \right] \quad (53)$$

where

$$a_v \equiv \frac{A_v}{m_v} \quad (54)$$

If the first term on the right hand side of equation (53) is several fold larger than the second, then

$$a_v = c_7 \beta_7 \frac{x_v N^*}{A_7} \quad (55)$$

On the other hand, employing average values when both species exist in approximately equal amounts yields

$$a_v = \frac{\langle c \rangle \langle \beta \rangle x_v N^*}{\langle A \rangle} \quad (56)$$

Employment of similar arguments to subsystem 3 leads to

$$a_1 = \frac{c_7 \beta_7 x_1 N^*}{A_7} \quad (57)$$

or

$$a_1 = \frac{\langle c \rangle \langle \beta \rangle x_1 N^*}{\langle A \rangle} \quad (58)$$

substitutions of equations (55) and (57), or equations (56) and (58) into equation (47) leads directly to

$$\left[\frac{a_v}{a_1} \right] = \left\{ 1 - \left[\exp \left(- \frac{t}{\tau} \right) \right] \right\} \quad (59)$$

Thus, measurements of the specific activities in the vapor and liquid phases as well as the liquid-vapor exposure time (t) yields a numerical value for τ . In accord with Schrage (22), the transfer flux relationship

$$G = \frac{P}{\sqrt{2 \pi R T}} \quad (9)$$

is assumed to be valid for molecules approaching as well as those leaving the liquid surface. Now, employing equation (49) the transfer coefficient (σ) can be determined. Both the vapor volume V and the exposed surface area A are calculated. The former is determined from P-V-T measurements which employ the expansion of argon gas from a known

standard volume (503.27 ± 0.05 ml) into the vapor volume, while the latter is obtained during the exchange process from magnified visual observation with a cathetometer telescope of the mercury meniscus and a comparison of its shape to that of the inside diameter of the true bore capillary tube. The mass concentration of mercury in the vapor volume c_T is calculated as a function of the vapor temperature (which is in equilibrium with the constant temperature bath).

Experimental Procedure

The equipment employed to determine the transfer coefficient at isothermal equilibrium is shown in Figure 8. Four equally spaced baffles in the 5.021 ± 0.002 liters vapor volume and a stirrer (in excess of 105 rpm) combined to provide a completely mixed vapor. The system was capable of holding a pressure of < 20 μ m for at least one hour. Pressure measurements were made with a swivel type McLeod gage; the accuracy was 3% of the reading or 1 mm of the scale division (whichever was greater). Reproducibility was two to five times better than the accuracy. The heater for the constant temperature bath employed a power relay and a bimetal thermoregulator. This thermoregulator was capable of operating over a temperature range of -73 to $+177^\circ\text{C}$ with a control accuracy of $\pm 0.02^\circ$. Counting of the radioactive material was accomplished with a shielded 2×2 NaI(Tl) well type crystal detector-scaler system. The radioactive mercury was introduced into a plastic coil (internal volume of 1.6×10^{-4} ml/cm) located in

- 1 VALVE
- 2 COLD TRAP
- 3 IRRADIATED MERCURY RESERVOIR
- 4 NORMAL MERCURY RESERVOIR
- 5 CONSTANT TEMPERATURE BATH
- 6 MAGNETIC OPERATED VALVE
- 7 5-LITERS MERCURY VAPOR VOLUME
- 8 STIRRER
- 9 SWIVEL McLEOD GAGE
- 10 VARIABLE SPEED MOTOR
- 11 TO FORE PUMP

Figure 8. The isothermal experimental system

the well of the crystal. All inside surfaces of the system were cleaned and degased until outgasing was not observed when the critical portions were closed off from the vacuum pump. The normal mercury in the vapor phase was double distilled, and the tagged mercury was decontaminated by standard distillation techniques upon its return from the nuclear reactor.

The procedure involves bringing the vapor volume to the desired temperature, completely evacuating the system, starting the stirrer, and closing the tagged mercury capillary tube with the cover valve. The normal mercury is introduced, and equilibrium in the vapor state is attained. Following this, the liquid mercury is removed, and the tagged mercury capillary is filled to the top. The exchange process is started by raising the tagged mercury cover valve and exposing it to the mercury vapor for a predetermined time interval. At the end of this period, the cover valve is rapidly lowered to seal the capillary and the tagged mercury is transferred to the cold trap. The pressure in the vapor space is increased to a little less than atmospheric pressure by adding argon gas. (This added pressure aids in forcing the mercury from the vapor volume into the cold trap.) Following this, the vapor volume is opened to the vacuum pump through the other cold trap. Mercury collected from the vapor volume in this trap is weighed and counted to determine its net activity. A weighed sample of the tagged mercury is also counted. Employing the experimental data just mentioned, equation (49) is solved to determine the transfer coefficient.

In the use of mercury vapor, one must guard against poisoning due to the sudden escape of the vapor and chronic poisoning due to the continuous slow leakage of the vapor. The experimental setup (Figure 8) is located in a hood. Mercury traps in the lines leading to the vacuum pump employ a refrigerant of liquid nitrogen. At this temperature, the vapor pressure of mercury is 1.7×10^{-27} mm. The volume of each trap is adequate to hold all the mercury contained in the system. Total failure of the hood system (i.e., a reversal of the air flow) would result in a concentration of $\sim 6 \times 10^{-3}$ milligrams of mercury per cubic meter of laboratory volume. The accepted non-poisonous concentration limit is 1 mg/m^3 . Under these conditions, the maximum concentration of radioactivity would be $< 2 \times 10^{-9} \mu \text{ Ci/cm}^3$.

Production and Analysis of the Radiomercury

A weighed sample of twice-distilled mercury was loaded into a double-jacketed neutron irradiation chamber. The chamber was designed to maximize the heat transfer from the mercury to the external surface of the outside container and minimize the possibility of mercury leakage into the nuclear reactor during neutron irradiation. The inner container had a volume $> 30 \text{ cm}^3$, which was three times that of the mercury that it held. The chamber was heated to $\sim 100^\circ\text{C}$ for a period of 66 hours. Immediately following this, it was leak checked, and no leaks were detected. The chamber was irradiated for 44.2 MWh in the Bulk Shield Facility (BSF) located at Wright-Patterson Air Force

Base. During irradiation, the chamber was positioned in the BSF such that it should have received 4.5×10^{12} n/(cm² sec) (thermal), 1.0×10^{11} n/(cm² sec) (> 2.9 MeV), and 4.0×10^7 R/h gamma radiation. Following standard decontamination techniques, the irradiated mercury was considered to be free from surface contaminations by materials that do not dissolve in mercury, dissolved metals, and those contaminants which are oxidizable.

Examination of an aliquot of the irradiated mercury with a shielded 3 x 3 NaI(Tl) flat-face crystal and a 400-channel analyzer spectrometer system indicated the presence of Hg-197m, Hg-197 and Hg-203. The resolution (FWHM) of this spectrometer system as a function of photon energy was determined and is shown in Figure 9. Besides the three radionuclides mentioned above, the neutron bombardment of normal mercury was expected to produce Hg-199 and Hg-205 also but owing to their extremely short half-lives, they were not detected in this study. An extended counting study of the aliquot mentioned above was started four days after the neutron irradiation was terminated. This waiting period was selected to allow the Hg-197m to pass through four half-lives before the condensation coefficient studies were performed. Thus, the contribution to the total counting rate from this particular radionuclide was considered to be negligible compared with that from Hg-197 and Hg-203. As shown in Figure 10, the tagged mercury employed during the determination of the condensation coefficient contained the radionuclides Hg-197 and Hg-203.

Figure 9. The resolution of the spectrometer system as a function of photon energy. (The two points marked Ba-133 represent two major peaks in the disintegration scheme of the same nuclide, namely, Ba-133.)

Figure 10. An extended counting study of an aliquot of the irradiated mercury

Data and Results

Two studies relating to the performance of the experimental setup were deemed necessary. To ascertain the reproducibility of the counting technique, a single sample of the tagged mercury was introduced five times into the plastic coil, counted, and removed. The resulting standard deviation for the counts per minute was 10661 ± 49 or $\pm 0.45\%$. Self-shielding was considered to be a potential source of counting error. Thus, the plastic coil was fabricated from a tube that possessed both a minimal wall thickness and an internal diameter in an attempt to obtain a constant counting rate per unit mass of mercury regardless of the sample size. Four different sample sizes of tagged mercury were used to evaluate this possibility. For tagged mercury samples ranging from 0.0266 to 0.9217 g, the counts per minute per gram were $(1.154 \pm 0.0015) \times 10^5$ or $\pm 13\%$.

The solution of equation (44) is based on the assumption that the mass fraction in the tagged liquid x_1 can be considered constant over the short time duration of the study. Two related phenomena to be considered in making such an assumption are 1) that the evaporation and condensation processes at the interface are isotopically random and 2) that isotopic separation does not exist in the tagged mercury capillary tube. Since this study is conducted at isothermal equilibrium, the former seems to be a valid assumption. On the other hand, a simple experiment can be performed to ascertain the validity of the latter.

Consider the evaporation of the tagged mercury into the volume after it has been evacuated. The isotopes present in the capillary tube range from Hg-196 to Hg-205, and the relative amounts of Hg-197 and Hg-203 in any sample can be ascertained by gamma radiation analysis. Thus, the existence of a measurable isotopic separation during evaporation can be expected to result in a change in the relative Hg-197 and Hg-203 abundance. Such evaporation studies were conducted at temperatures of 90.6, 110.3, and 123.7°C. Before each study, the net counting rate per gram of tagged mercury was determined, the areas under the 0.078- and 0.269-MeV photopeaks were calculated, and the peak heights of these two photopeaks were noted. Following each study, the determinations and calculations were repeated for the mercury in both the vapor and the liquid phases. Use of these photopeaks as being representative of the two radionuclides is verified by the fact that both lie on the resolution curve (Figure 9). The resulting photopeak area and height ratios were constant within an experimental error of 2.0 and 2.1%, respectively. The counting rate per gram of mercury was a constant within 0.11%. From an examination of the results it was found that isotopic separation did not occur. Therefore, the mass fraction in the liquid x_1 can be considered constant, and the solution of equation (44) is valid.

The calculated and experimentally determined data for the evaluation of condensation-evaporation coefficients are tabulated in Table 3. Six studies were conducted at three different vapor temperatures.

TABLE 3

CALCULATED AND MEASURED DATA AND RESULTS REQUIRED FOR THE DETERMINATION
OF THE TRANSFER COEFFICIENT FOR FILM CONDENSATION OF MERCURY

Vapor Temp. (deg C)	Mass Transfer Flux G^a g/(cm ² sec)	Mass Concentration in Vapor c_T (g/cm ³)	Tagged Mass Fraction in Capillary x_1 (cpm/g)	Exposure Time t (sec)	Tagged Mass Fraction in Vapor x_V (cpm/g)	Transfer Coefficient ^b σ
92.1	7.77×10^{-3}	1.56×10^{-6}	5.52×10^5	9.0±0.5 14.0±0.5	3.92×10^5 4.91×10^5	0.98 1.10
111.4	2.07×10^{-2}	4.07×10^{-6}	2.24×10^5	12.0±0.5 15.0±0.5	1.82×10^5 2.00×10^5	0.98 1.05
123.6	3.71×10^{-2}	7.12×10^{-6}	8.62×10^4	11.0±0.5 10.0±0.5	6.81×10^4 2.70×10^4	0.97 0.25

^aSee Wilhelm (19).

^bDetermined exposure surface area A of $(1.41 \pm 0.03) \times 10^{-1}$ cm².

Discussion

Calculations of the transfer coefficient are made using the specific activities in the vapor and liquid phases. This approach is based on the assumption that the liquid-vapor transfer process for this experimental setup is described by equations (9), (49) and (59). The transfer coefficient at liquid-vapor interfaces of mercury for isothermal equilibrium in a clean system is essentially unity (Table 3). The sixth study provided a coefficient with a value approximately one-quarter that of the other five. This value is assumed to be the result of a dirty system. The stirrer employed was Teflon-coated glass. After 10 sec of this study had elapsed, bits of Teflon were observed in the vapor volume, and the system began to leak at the stirrer feed-through. Visual examination of the mercury revealed the existence of surface contamination. (This contamination was not observed during the first five studies.)

As previously stated, the calculations employed herein were based on the assumption that the vapor be well stirred (which implied the absence of concentration gradients at or near the liquid-vapor interface). The use of the stirrer, baffles in the vapor volume, and short exposure times between the liquid and vapor were utilized in an attempt to realize this assumption. The main reason for believing that these gradients were not present in the studies described was the fact that a condensation-evaporation coefficient of unity was obtained. On the other hand, these concentration gradients in the liquid and vapor might be expected to play a more important role at higher temperatures

where the mean-free-path of the vapor molecules would be reduced. The effect of the gradients (should they exist) would be to decrease the measured condensation coefficient.

Dropwise condensation was not observed during any of the six studies. The value of the coefficient mentioned above is in good agreement with that reported by Subbotin et al. (33), Ivanovskii et al. (34) and Meyrial et al. (35). It is also in agreement with theoretical and experimental results obtained with materials other than liquid metals. The isothermal equilibrium measurements reported herein have demonstrated the feasibility of the radioactive tagged liquid-metal technique for measuring the evaporation-condensation coefficient. The same technique can be applied to nonisothermal studies.

CHAPTER V

NONISOTHERMAL STUDY

In this study the radioactive tracer technique is employed to examine the mass transfer process under nonisothermal conditions of the liquid-vapor phases. The primary objectives of the study presented below are: to determine whether the condensation and evaporation coefficients are numerically equal; the possible existence of the interphase zone proposed in the Fedorovich-Rohsenow mass transfer model (32); and the heat flux associated with the condensation process.

Nonisothermal Condition Model

A model of the steady state nonisothermal experimental setup is shown in Figure 11. Initially, a saturated radioactive mercury vapor (tagged with Hg-197 and Hg-203) is separated from a flowing nonradioactive liquid mercury. To provide a finite condensation rate when the two phases are allowed to come into contact, the circulating liquid phase is kept at a lower temperature than the vapor phase. Thus, the net rate of condensation is directly related to the measured increase of radioactivity in the flowing liquid phase. Furthermore, a steady state transfer process is realized by maintaining a constant pressure in the vapor volume and a steady liquid flow rate.

Figure 11. A model of the steady state nonisothermal experimental system

Condensation Coefficient Determination. To maintain a constant pressure within the vapor chamber, the "boil-up" rate (r) is given by

$$r = \frac{Q}{\lambda} \quad (60)$$

where Q and λ represent the rate at which energy (heat) is supplied to the boiler and the latent heat of condensation, respectively. A mass balance on the radioactive mercury in the liquid phase is

$$(\Gamma \sigma_c G_c A x_v) + (\sigma_e G_e A x_1) = \frac{d}{dt} (x_1 L) \quad (61)$$

or

$$\begin{aligned} (\Gamma \sigma_c G_c A x_v) + (\sigma_e G_e A x_1) &= \\ &= x_1 \left(\frac{dL}{dt} \right) + L \left(\frac{dx_1}{dt} \right) \end{aligned} \quad (62)$$

where L represents the mass of the mercury in the circulating liquid phase. The condensation and evaporation mass transfer flux are defined by equations (20) and (21). In this steady state model, it is assumed that the "boil-up" rate (r) from the boiler is a constant, equal to the "net condensation" transfer rate of mercury from the vapor chamber. That is

$$r = \frac{Q}{\lambda} = \frac{dL}{dt} \quad (63)$$

Substitution of equation (63) into (62) yields

$$\begin{aligned}
 (\Gamma \sigma_c G_c A x_v) + (\sigma_e G_e A x_1) &= \\
 &= (x_1 r) + L \left(\frac{dx_1}{dt} \right)
 \end{aligned} \tag{64}$$

The liquid in the circulating system at any time $t \geq 0$ is

$$L = L_0 + r t \tag{65}$$

where L_0 represents the initial amount of liquid in the circulating system and t is the elapsed time $t \geq 0$. Under these experimental conditions, and remembering that nonradioactive mercury is initially loaded into the circulating liquid system ($x_1 = 0$ at $t \leq 0$), equation (64) becomes at time $t = 0$

$$\Gamma \sigma_c = \frac{L_0 \left[\frac{d}{dt} \left(\frac{x_1}{x_v} \right) \right]_{t=0}}{G_c A} \tag{66}$$

where the mass flux G_c is defined by equation (20). Employment of arguments similar to those used to obtain equations (56) and (58) for the isothermal equilibrium study (Chapter IV), leads directly to a substitution of the specific activities a_1 and a_v for the mass fractions x_1 and x_v in equation (66). It is necessary to relate x_v to x_b through a boiler balance (see Appendix C). If x_b is constant, the result is $(x_v/x_b) = 1 + (r/G^*A^*)$. Thus, it is necessary to experimentally determine or measure the radiomercury in the circulating liquid (a_1) as a function of time, the specific activity of the mercury in the boiler

(a_b), the initial mass in the liquid circulating system (L_0), the liquid-vapor interface area (A), and the bulk vapor temperature and pressure (T_v , P_v). Extrapolation of the measured derivatives to time $t = 0$, and employment of the above mentioned quantities leads to a solution of equation (66) for the product $\Gamma \sigma_c$. In order to provide a steady state experiment which can be conducted by a single individual, it is assumed that only slightly non-equilibrium conditions are employed. This experimental restriction allowed the use of equation (23) to provide an approximate numerical value for Schrage's (22) "correction" term Γ , and a calculation of condensation coefficient.

Evaporation Coefficient Determination. The numerical value of the evaporation coefficient is obtained through the use of equation (18)

$$\sigma_e = \frac{G_{\text{net}} - \Gamma \sigma_c G_c}{G_e} \quad (67)$$

where

$$r = G_{\text{net}} A \quad (68)$$

and the condensation and evaporation mass flux (G_c and G_e) are defined by equations (20) and (21), respectively. The product $\Gamma \sigma_c$ is obtained from the condensation coefficient determination.

Heat Flux Determination. The heat flux (q) for interfacial energy transfer can be computed from

$$q = [\lambda + c_{p,v} (T_v - T_s)] G_{net} \quad (69)$$

employing the net mass flux obtained from equation (68), the measured interface temperature jump ($T_v - T_s$), and the known values of latent heat of condensation (λ), and specific heat ($c_{p,v}$). The interfacial heat transfer coefficient between the vapor and liquid phases can be obtained from (40)

$$h_i = \frac{G_{net} \lambda}{T_v - T_s} \quad (70)$$

Experimental Procedure

The equipment employed to determine the condensation and evaporation coefficients, heat flux, and interfacial heat transfer coefficient under nonisothermal conditions is shown in Figure 12. This particular experimental setup is a modified version of that employed by Subbotin et al. (33) to investigate the film type condensation of pure potassium vapor. Measurements made with a Pirani type gage demonstrated that the system was capable of holding a pressure of $<20\mu\text{m}$ for at least 12 hours. The heater for the constant temperature bath employed a power relay and a bimetal thermoregulator. This thermoregulator was capable of operating over a temperature range of $+25$ to $+360^\circ\text{C}$ with a control accuracy of $\pm 0.02^\circ$. Continuous counting of the radioactive material in the circulating liquid was accomplished with a thin-metal wall, gas filled, beta-gamma radiation detector-ratemeter-strip chart

- 1 TO ARGON GAS
- 2 TO FORE PUMP
- 3 PIRANI GAGE
- 4 CONSTANT TEMPERATURE BATH
- 5 MOVABLE THERMOCOUPLES
- 6 ELECTROMAGNET OPERATED COVER VALVE
- 7 BOILER HOLDING RADIOMERCURY
- 8 EXCHANGE CHAMBER
- 9 RADIATION DETECTOR
- 10 HOLDING TANK
- 11 CIRCULATING PUMP
- 12 PRESSURE TRANSDUCER
- 13 CROSS-OVER VALVE
- 14 SAMPLE PORT

Figure 12. The nonisothermal experimental system

recorder system. Liquid samples periodically removed from the circulating mercury were counted in a shielded 3x3 NaI(Tl) well type crystal detector-multichannel analyzer system. An electromagnetic pump was used to provide a continuously circulating liquid mercury. All inside surfaces of the system were cleaned and degased until outgasing was not observed when the critical portions were closed off from the vacuum pump. The nonradioactive mercury in the liquid phase was double distilled, and the tagged mercury was decontaminated by standard distillation techniques upon its return from the Bulk Shield Facility (BSF) located at Wright-Patterson Air Force Base. Pressures in the vapor volume, as well as the liquid mercury flow rates, were determined with the aid of a differential pressure transducer-strip chart recorder system. Basically, the transducer consisted of a stainless steel pressure sensing diaphragm (range of ± 0.5 psi) whose deflection controlled the gap in each of two magnetic circuits. The gaps changed in opposite directions and produced corresponding changes in the inductance of two pickoff coils. This effect was utilized by the transducer indicator to provide a direct meter and recorder readout. The temperature distribution across the bulk vapor, liquid-vapor interface and into the bulk circulating liquid was measured with a movable probe consisting of three #28 gage chromel-alumel thermocouples. These thermocouples were mounted in a vertical tube whose displacement was 0.07 ± 0.02 cm. The thermocouples were so spaced (upper to lower separation distance was 1.012 cm) that it was possible to simultaneously measure the vapor space, liquid surface and circulating liquid temperatures. The probe

had sufficient displacement (9.71 ± 0.03 cm) so that all three thermocouples could either be in the vapor space or completely immersed in the liquid. Thermocouples were also placed at various locations around the heated-insulated system (Figure 13). These thermocouples were located between the tubing and alternate layers of asbestos textile tape and chromel heater ribbon. The use of a second layer of heater ribbon was intended to decrease the heat loss.

The procedure involves bringing the vapor volume, boiler and circulating system to operating temperatures with the exchange chamber being closed with the cover valve. The condensing surface is maintained close to the top of the exchange chamber as temperature controlled liquid mercury is pumped through the center tube (3 mm I.D.) in the exchange chamber, and effused into a 12 mm. I.D. overflow tube. When equilibrium is attained between the radiomercury in the boiler and that in the vapor volume (as determined by the vapor pressure of mercury), the cover valve is removed thereby allowing the vapor and liquid to come into contact. The differential pressure transducer is used to determine the vapor pressure and liquid flow rate. Samples are withdrawn periodically from the liquid circulating system and a constant monitoring Geiger-Mueller detector system is used for measuring the continuous count rate rise.

Production and Analysis of the Radiomercury

The production and analysis of the radiomercury was discussed in Chapter IV.

Figure 13. Thermocouple locations for the nonisothermal experimental system

Data and Results

Several studies relating to the performance of the nonisothermal experimental system are described. This is followed by a treatment of the results of the actual runs.

Calibration of the pressure transducer, used to establish the mercury vapor pressure, was obtained by first bringing the constant temperature bath and vapor volume to some predetermined temperature, and then increasing the voltage to the boiler (in a stepwise manner) until the pressure transducer reading was constant. An equilibrium condition was assumed to exist between the boiler and vapor volume when the change in the transducer reading first became constant. Under these conditions, the temperature inside the vapor volume is assumed to correspond to the saturation pressure of mercury. Studies conducted over a temperature range of 142°C to 193°C (Table 4) provided a calibration between vapor pressure and transducer reading which is slightly non-linear at the higher end.

TABLE 4

CALIBRATION OF THE PRESSURE TRANSDUCER TO MERCURY
VAPOR TEMPERATURE AND SATURATION PRESSURE

Bath Temperature (°C)	Mercury Vapor Pressure (mm of Hg)	Relative Change in Transducer Reading (divisions)
142	1.98	18.5 ± 0.5
164	4.91	45.0 ± 0.5
176	7.6	69.0 ± 0.5
193	13.7	117.0 ± 0.5

Another study involved the calibration of the pressure transducer used to establish the liquid mercury flow rate. In this study, the down-stream line leading to the exchange chamber (Figure 12) was removed. Liquid samples were collected at the sample port for measured time intervals and weighed. The transducer reading versus laminar mass flow rate was found to be linear with a slope of 18 grams per minute per division change in the reading. These studies employed mercury masses in the liquid system equivalent to those used during actual non-isothermal exchange studies. Under these conditions, measurable changes in flow rate with changes in head height do not exist.

Reproducibility studies of the ability to remove all of the mercury from the circulating liquid portion of the system revealed that this

can be accomplished to within 0.2% of the mass loaded into the system.

Visual examination of the liquid-vapor interface to determine the exchange area proved to be difficult. This difficulty resulted from the fact that the horizontal view is prevented by insulation. From vertical visual examinations, the circulating liquid surface appeared to be slightly rounded at the edges and rather flat in the central portion. Coupling the observed surface shape (approximately flat) with the known internal diameter of the exchange chamber (12 mm), the liquid-vapor exchange surface is calculated as 1.6 cm².

The calculated and experimentally determined data for the evaluation of the condensation and evaporation coefficients, and heat transfer coefficient are tabulated in Table 5. Three studies were conducted under nearly identical operating conditions. The mercury vapor pressure readings made prior to and following the exchange process were found to be identical. Furthermore, the circulating liquid mass flow rate was constant for each of the three studies. The movable chromel-alumel thermocouple readings for the three studies are shown in Figures 14 through 19. Circulating liquid samples collected prior to the removal of the cover valve for each run exhibited a counting rate equivalent to that of the background of the shielded 3x3 NaI(Tl) crystal detector-multichannel analyzer system. Count rate measurements of the tagged mercury in the boiler prior to the start of and after a run was completed provided identical (within the limits of experimental error) specific activity determinations, i.e., no dilution effect was noticeable during a run.

TABLE 5

CALCULATED AND MEASURED DATA AND RESULTS REQUIRED FOR THE
DETERMINATION OF THE CONDENSATION AND EVAPORATION
COEFFICIENTS, HEAT FLUX, AND INTERFACIAL HEAT
TRANSFER COEFFICIENT FOR FILM
CONDENSATION OF MERCURY

Study No.	1	2	3
L_o (g)	1006	923	998
$L_o + \Delta L$ (g)	1238	1107	1229
Exposure Time (min)	46,5	38,0	46.0
T_v (°C)	181	181	181
T_s (°C)	176	176	177
$\left[\frac{d\left(\frac{a_1}{a_b}\right)}{dt} \right]_{t=0}$ (min ⁻¹)	2.2×10^{-2}	2.5×10^{-2}	2.7×10^{-2}
$\Gamma \sigma_c$	6.2×10^{-1}	6.6×10^{-1}	7.1×10^{-1}
σ_c	0.58	0.61	0.66
σ_e	0.90	0.94	0.98
q (g-cal/cm ² min)	2.2×10^2	2.1×10^2	2.2×10^2
h_i (g-cal/cm ² min °C)	4.4×10^1	4.3×10^1	5.4×10^1
G_e (g/cm ² min)	18.0	18.0	18.5
G_{net} (g/cm ² min)	3.1	3.0	3.1
G_c (g/cm ² min)	21.3	21.3	21.3

Figure 14. Movable chromel-alumel thermocouple readings for nonisothermal experimental study number 1

Figure 15. Movable chromel-alumel thermocouple readings for nonisothermal experimental study number 2

Figure 16. Movable chromel-alumel thermocouple readings for nonisothermal experimental study number 3

Figure 17. Movable chromel-alumel thermocouple readings at the liquid-vapor interface for nonisothermal experimental study number 1

Figure 18. Movable chromel-alumel thermocouple readings at the liquid-vapor interface for nonisothermal experimental study number 2

Figure 19. Movable chromel-alumel thermocouple readings at the liquid-vapor interface for nonisothermal experimental study number 3

Discussion

Calculations of the evaporation and condensation coefficients are made using the specific activities in the boiler and liquid phases. This approach is based on the assumption that the liquid-vapor transfer process for this experimental system is described by equations (23), (66) and (67). The fact that during each study the vapor pressure, vapor temperature profile (see Figures 14 through 16), and the circulating liquid mass flow rate are constant implied that a steady-state condition existed. On the other hand, the interfacial exposure area is estimated, and the slope of the " (x_1/x_b) versus exposure time" curve at time $t = 0$ (for the GM Detector-Ratemeter-Recorder system) is determined after first correcting the radiation detector-ratemeter-strip chart recorder reading for changes in the full scale range of the ratemeter, background radiation, and time lag of the condensed radiomercury in the circulating liquid. As an alternate method, one can simply take the initial slope of the raw data as accurately as possible. This method gave results consistent with those values reported in Table 5. The tabulated values are those of the more lengthy method involving the corrections and time smoothing, as this procedure is considered a little more accurate. The condensation and evaporation coefficients tabulated in Table 5 are considered to be valid to within an experimental error of 10 percent. These tabulated results show that the evaporation and condensation coefficients are essentially unity and 0.65, respectively.

According to the Fedorovich-Rohsenow nonequilibrium transport model, the liquid and vapor phases are separated by an interphase zone approximately ten mean-free-path lengths thick. The existence of this zone is indicated by a temperature greater than that of the bulk liquid and less than that of the bulk vapor. Thermocouple readings within 5 mm on either side of the liquid-vapor interface are shown in Figures 17 through 19. The circles in these figures have diameters equivalent to the measured diameters of the thermocouple beads. The overlap of the circles at the liquid-vapor interface (in each study) is within the error of the diameter measurements. The thermocouple bead diameter is equivalent to the thickness of the Fedorovich-Rohsenow interphase zone. From an examination of Figures 17 and 19, the bulk vapor temperature exists up to within less than 10 mean-free-path lengths of the liquid surface. In Figure 18, one of the three thermocouples provided a reading slightly less than the average value attributed to the bulk vapor. In light of the readings shown in Figures 17 and 19, coupled with the fact that this "intermediate" reading is essentially equivalent to the average bulk vapor temperature, it is implied that the value is not related to the existence of an interphase zone of ten mean-free-path lengths thick. These thermocouple measurements are in agreement with those of Subbotin et al. (33). Employing a moving thermocouple, they observed only a temperature jump at the liquid-vapor interface of condensing potassium. Implied from Figures 17 through 19 is that the " $T_i = T_v$ " condensation model is valid for this experimental system.

The interfacial heat transfer coefficient between the vapor and liquid phases (see Table 5) for this experimental configuration is approximately $47 \text{ (g-cal)/(cm}^2 \text{ min } ^\circ\text{C)}$. To provide a basis for the comparison of various researchers' results, the experimentally determined value is normally compared to the Nusselt coefficient. Although Nusselt's theoretical heat transfer analysis (topic 1, Figure 1) has been applied to geometries such as vertical and horizontal cylinders, banks of horizontal cylinders, and flat plates at any orientation, none exists for the experimental system described in this paper. This experimental system might be approximated to that of the film type condensation on a horizontal flat plate. The flow from a flat plate and the crude similarity to this experimental configuration is shown in Figure 20. The disadvantage to this approach is that the flat plate theory has a horizontal limit of zero because of a $\sin^4 \theta$ dependency. It is of academic interest to examine the value of the Nusselt coefficient as the angle of the plate is decreased. The vertical plate local coefficient [equation (1)] at "one-half diameter" ($x = 0.6 \text{ cm}$) down from the top of the plate is $1.39 \times 10^4 \text{ (g-cal)/(cm}^2 \text{ min } ^\circ\text{C)}$, decreasing to $4.98 \times 10^3 \text{ (g-cal)/(cm}^2 \text{ min } ^\circ\text{C)}$ at 1° off horizontal, to $1.80 \times 10^3 \text{ (g-cal)/(cm}^2 \text{ min } ^\circ\text{C)}$ for 1 sec off horizontal, and approaching zero for the horizontal flat plate. The vertical case value mentioned above corresponds to a film thickness of $8.6 \times 10^{-5} \text{ cm}$, while the experimental coefficient of $40 \text{ (g-cal)/(cm}^2 \text{ min } ^\circ\text{C)}$ corresponds to $3.2 \times 10^{-2} \text{ cm}$, which is still a small number. A ratio of h_i/h_{Nu} is meaningless since the Nusselt coefficient is not really

Figure 20. The flow from a flat plate and the crude similarity to the nonisothermal experimental configuration

known. On the other hand, for the case of vapor condensing on a horizontal circulating liquid, it is reasonable to assume that a small finite value of h_{Nu} for the horizontal limit (other than zero) could exist.

By contrast, the condensation and evaporation coefficients are meaningful and important. As mentioned above, the evaporation and condensation coefficients for mercury vapor condensing on a circulating liquid mercury are essentially unity and 0.65, respectively.

In one analysis by Wilhelm (19) to ascertain the numerical value of the condensation coefficient, he required that the condensation and evaporation coefficients be equal to each other. By arbitrarily requiring that the coefficients be equal for the three nonisothermal studies reported in this paper, it is found that the slope of the " a_1/a_v versus time" curve at time $t = 0$ must be two-fold greater than experimentally observed. From another viewpoint, this means that employment of the observed experimental data and the "equality" restriction leads to a transfer coefficient of 1.7, but the upper limit of the transfer coefficient is known to be unity.

It is clear that the condensation and evaporation coefficients are not identical except at equilibrium. Furthermore, the results strongly imply that the evaporation coefficient is unity. The mean-free-path (using the billiard-ball model) for a mercury molecule evaporated into a vapor at 200°C is of the order of 10^5 molecular diameters. The mean-free-path is decreased to 10^4 molecular diameters for a vapor temperature corresponding to a pressure of one atmosphere,

The ratio of the volume occupied by all vapor molecules to total vapor volume is 10^{-11} and 10^{-8} for the 200°C and one atmosphere cases, respectively. Based upon these calculations, it is highly improbable that the occupied space presented to the evaporated molecules is sufficient to reduce the evaporation coefficient to a value of less than unity in this vapor pressure range, which is as observed.

Wilhelm (19) recomputed all available data in the literature assuming an evaporation coefficient of unity, and arrived at a plot which implied that the condensation coefficient rapidly decreased from unity at isothermal equilibrium to a very low value at high thermal driving forces (Figure 21). The condensation coefficients reported in Table 5 are consistent with this, but are at a relatively low value of the driving force. From an examination of the condensation coefficient curve shown in Figure 21, it is obvious that studies at higher driving forces are needed to ascertain the validity of the observed rapid decrease. Because of the complexity of operating the experimental system and limitations of the "boil-up" rate from the boiler, it is impossible for us to conduct steady-state studies in excess of the temperature jump ($T_v - T_s$) employed. Therefore, a complete study of the condensation coefficient as a function of the thermal driving force is prohibited by the existing experimental system.

As a result of not being able to perform studies at higher temperature jumps, it is impossible to verify or disprove the rapid decrease in the condensation coefficient with increased driving force. Now as

in the past, these low experimental values are questioned. Calculations of the experimental drift velocity correction factor Γ shows that it decreases with increasing driving force (Figure 21). This results from a minor change in the net mass transfer flux with increasing driving force and is without physical interpretation, since it is logically expected to be an increasing function. Theoretical representations of the correction factor Γ are presented in Figure 21 where the condensation coefficient is held constant at a value of 0.65 and unity. The maximum possible value of Γ is represented by the latter curve, while the former is related to the present experimental work. The shape of these theoretical Γ curves indicate that a condensation coefficient whose value is constant or undergoes a less drastic change with increased driving force than reported in the literature, yields a more logically shaped Γ function.

In summary, it is clear that the evaporation coefficient is unity while the condensation coefficient is less than unity and is approximately 0.65 under the nonequilibrium conditions of this research. Prior to this research, these facts have never been experimentally established. Furthermore, this research has confirmed the work of Subbotin et al. (33) in that a temperature gradient does not exist at the liquid-vapor interface. Directly related to this result is the fact that the Fedorovich-Rohsenow interphase zone (vapor barrier) does not exist. This hypothetical zone presented in the Fedorovich-Rohsenow liquid-vapor interface model is a device that forces the condensation coefficient to be unity.

Figure 21. Condensation coefficients and drift velocity correction factors from data based upon the evaporation coefficient being equal to unity

These statements are based upon experimental studies conducted with a small temperature jump, and it might be argued that the interphase zone exists at a higher driving force. According to the model under consideration, the vapor barrier is approximately ten mean-free-path lengths thick. The mean-free-path decreases with increasing vapor temperature (and corresponding pressure) and thus the thickness of the zone is decreased as the driving force is increased. If the interphase zone does not exist at the lower temperature, then it would be unlikely to be observed at a higher temperature.

This discussion is confined to liquid metals, and mercury in particular. As previously stated, the transfer coefficient associated with water is essentially unity. In making such a statement, it must be remembered that the researchers assumed that the condensation and evaporation coefficients are equal to each other and did not use a model which possessed an interphase zone. Since their experimental results provided a numerical value of unity rather than one substantially different from this value, it is reasonable to assume that the condensation and evaporation coefficients of water are unity. These results imply that the condensation coefficient of water is not necessarily numerically equal to that associated with liquid metals. This difference cannot be attributed to the free volume in the liquid phase of the two materials because they are nearly equal. The fact that the self-diffusion of water molecules in water is five-fold greater than that of mercury molecules in mercury may partially explain the fifteen-fold difference between the results obtained for mercury and that of water.

At present, a physically consistent explanation does not exist for the difference.

CHAPTER VI

CONCLUSIONS

The following conclusions are made for a pure, saturated mercury vapor condensing on its liquid phase.

Isothermal Equilibrium. At isothermal equilibrium of the liquid and vapor phases, the condensation and evaporation coefficients are equal to each other and are unity.

Nonisothermal Conditions. Under nonisothermal conditions of the liquid and vapor phases, the condensation coefficient is less than unity and the evaporation coefficient is equal to unity. All previously conducted film type condensation studies of liquid metals have suffered because of the assumption that the evaporation and condensation coefficients are at all times identical. A temperature jump exists at the liquid-vapor interface. The bulk vapor temperature exists up to a distance of less than ten mean-free-path lengths from the liquid surface, and the " $T_i = T_v$ " condensation model is valid. The Fedorovich-Rohsenow interphase zone of ten mean-free-path lengths thick and the associated temperature gradient do not exist.

Finally, this work has clearly demonstrated the feasibility of using radioactive tracers for the evaluation of evaporation and condensation coefficients for both isothermal and nonisothermal conditions.

CHAPTER VII

RECOMMENDATIONS

It is recommended that a new experimental system be fabricated with which the condensation and evaporation coefficients can be studied over a significantly large range of liquid-vapor interfacial temperature jumps for liquid metals and other materials. The number of materials to be studied is restricted to those where the radioactive tracer technique can be used. Maintaining a steady-state condition at high driving forces requires that many portions of the system be automatically controlled, and the boiler possess an extensive capacity. Furthermore, the fabrication of such a system requires the services of an outstanding shop facility. Owing to the complexity of the experimental system and the numerous radionuclides to be used, it is recommended that the work be performed at one of the national laboratories.

The isothermal equilibrium studies are relatively simple and an obvious extension should be to study the effect of noncondensable gases on the coefficients.

APPENDIX A

DATA AND SAMPLE CALCULATIONS FOR THE ISOTHERMAL EQUILIBRIUM STUDY

The data for the six isothermal equilibrium studies and a sample calculation are presented below.

Data

TABLE 6. DATA FOR THE ISOTHERMAL EQUILIBRIUM STUDIES

Column (1)	Study Number
(2)	Date of Study
(3)	Amount of Sample Collected - g
(4)	Type of Sample - L for Liquid; V for Vapor
(5)	Net Total Count
(6)	Clock Time Counting Period - min.

TABLE 6

(1)	(2)	(3)	(4)	(5)	(6)
1	9-25-67	0.0261	L	144183	10
		0.0078	V	306112	100
2	9-25-67	0.0261	L	143995	10
		0.0078	V	383241	100
3	9-29-67	0.0261	L	116928	20
		0.0204	V	371601	100
4	9-29-67	0.0261	L	117131	20
		0.0211	V	421931	100
5	10-4-67	0.0261	L	225001	100
		0.0357	V	243743	100
6	10-4-67	0.0261	L	224972	100
		0.0355	V	95863	100

Sample Calculation

To illustrate how the tabulated transfer coefficient values in Table 3 (Chapter IV) are determined, a sample calculation is performed below using the data for study number 1 (9-25-67).

Determination of τ . Employing equation (59), where the ratio a_v to a_1 is equal to the ratio x_v to x_1

$$\begin{aligned} - (1/\tau) &= (9 \text{ sec})^{-1} \left\{ \ln [1 - (3.92 \times 10^5 \text{ cpm/g}) / (5.52 \times 10^5 \text{ cpm/g})] \right\} \\ - (1/\tau) &= - 1.37 \times 10^{-1} \text{ sec}^{-1} \end{aligned}$$

Determination of σ . Employing equation (9), where

$$P = 1.78 \times 10^{-1} \text{ mm of Hg}$$

$$T = 92.1^\circ\text{C}$$

gives a mass transfer flux (G) of $7.77 \times 10^{-3} \text{ g}/(\text{cm}^2)(\text{sec})$. Substitution of this value for G as well as

$$A = 1.41 \times 10^{-1} \text{ cm}^2$$

$$V = 5.02 \times 10^3 \text{ cm}^3$$

$$c_T = 1.56 \times 10^{-6} \text{ g}/\text{cm}^3$$

$$(1/\tau) = 1.37 \times 10^{-1} \text{ sec}^{-1}$$

into equation (49) gives $\sigma = 0.98$.

Alternative Analysis. An alternative approach to the analysis of the data is provided by a plot of $[1 - (a_v/a_1)]$ versus the exposure time (Figure 22). From an examination of this figure it is seen that the six isothermal equilibrium studies are relatively insensitive to temperature; study number 6 is apparently in error; and the transfer coefficient is essentially unity.

Figure 22. An alternative approach to the display of the six isothermal equilibrium studies

APPENDIX B

DATA AND SAMPLE CALCULATIONS FOR THE NONISOTHERMAL STUDY

The data for the three nonisothermal studies and a sample calculation are presented below. The format used to present the data consists of: listing the amount of mercury in the system at the start and end of the study, etc.; tabulation of the counting rates (measured with the shielded 3x3 NaI(Tl) crystal-multichannel analyzer system), and those rates measured with the Geiger-Müller (GM) detector-ratemeter-recorder system after the cover valve is closed and the radioactive circulating liquid mercury is well mixed; displaying the GM detector-recorder trace obtained during the study (Figures 23 through 28); and tabulation of the "observed and reduced" counting rates (measured with the GM detector-ratemeter-recorder system) obtained from the GM detector-recorder trace.

Data

Some Basic Data. Some of the basic data related to the nonisothermal study are tabulated below.

TABLE 7. BASIC DATA FOR THE NONISOTHERMAL STUDIES

Column (1) Study Number and Date of Study

TABLE 7--continued.

- Column (2) Circulating Liquid-Vapor Exposure Time - min
- (3) Amount of Mercury in Circulating Liquid System at
Start of Study - g
- (4) Amount of Mercury in Circulating Liquid System at
End of Study - g
- (5) Amount of Mercury in Boiler and Vapor Volume at
Start of Study - g
- (6) Amount of Mercury in Boiler and Vapor Volume at
End of Study - g
- (7) Mass Flow Rate in Circulating Liquid System During
the Study - g/sec

(1)	(2)	(3)	(4)	(5)	(6)	(7)
1 12-26-69	46.5	1009	1241	1011	778	~45
2 1-23-70	38.0	927	1111	924	738	~45
3 1-24-70	46.0	1001	1232	1003	774	~45

Counting Rates. The counting rates measured with the shielded 3x3 NaI(Tl) crystal-multichannel analyzer system and those with the GM detector-ratemeter-recorder system (after the cover valve is closed and the radioactive circulating liquid mercury is well mixed) are tabulated below.

TABLE 8. COUNTING RATES FOR THE NONISOTHERMAL STUDIES

Column (1)	Study Number and Date of Study
(2)	Amount of Sample Collected - g
(3)	Time at which Sample Removed - B for Before Cover Valve Opened; A for After Cover Valve Closed
(4)	Type of Sample - N for None (Used to Measure Background Count); C for Circulating Liquid; B for Boiler
(5)	Type of Detector Used - X for Shielded 3x3 NaI(Tl) Crystal-Multichannel Analyzer; GM for GM Detector-Ratemeter-Recorder
(6)	Total Count
(7)	Type of Count - N for Net; G for Gross
(8)	Live Time Counting Period for Samples Counted in the Shielded 3x3 NaI(Tl) Crystal-Multichannel-Analyzer - sec
(9)	Net Specific Activity of Sample - cpm/g

TABLE 8--continued.

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)
1 12-26-69	0.0000	B	N	X	1396	G	100	---
	3.1149	B	C	X	1357	G	100	0
	4.8240	A	C	X	215633	N	100	2.68×10^4
	5.8291	A	C	X	260565	N	100	2.68×10^4
	4.3239	B	B	X	4341217	N	1000	6.00×10^4
	3.1926	A	B	X	3192284	N	1000	6.00×10^4
	---	A	C	GM	---	N	---	9.06
2 1-23-70	0.0000	B	N	X	1353	G	100	---
	3.9127	B	C	X	1358	G	100	0
	4.4731	A	C	X	140396	N	200	9.44×10^3
	4.0916	A	C	X	127943	N	200	9.44×10^3
	3.7145	B	B	X	272937	N	200	2.21×10^4
	3.0141	A	B	X	221946	N	200	2.21×10^4
	---	A	C	GM	---	N	---	4.06
3 1-24-70	0.0000	B	N	X	1417	G	100	---
	2.7647	B	C	X	1401	G	100	0
	3.3774	A	C	X	116221	N	200	1.03×10^4
	3.1467	A	C	X	108353	N	200	1.03×10^4
	3.6219	B	B	X	264993	N	200	2.20×10^4
	2.9112	A	B	X	213104	N	200	2.20×10^4
	---	A	C	GM	---	N	---	4.30

GM Detector-Ratemeter-Recorder Response. The GM Detector-Ratemeter-Recorder responses for the nonisothermal studies numbered 1, 2, and 3 are shown in Figures 23 and 24, 25 and 26, and 27 and 28, respectively. From an examination of Figures 23, 25, and 27 it is seen that the radioactive mercury does not reach the detector until approximately one minute after the cover valve has been raised, and that an axial mixing problem exists during the first few minutes. It is seen in Figures 24, 26, and 28 that the radioactive mercury does not become "well mixed" until after the cover valve is closed.

Figure 23. GM-Ratemeter-Recorder response for nonisothermal study number 1 (-8 to 25 minutes)

Figure 24. GM-Ratemeter-Recorder response for nonisothermal study number 1 (25 to 56 minutes)

Figure 25. GM-Ratemeter-Recorder response for nonisothermal study number 2 (-5 to 22 minutes)

Figure 26. GM-Ratemeter-Recorder response for nonisothermal study number 2 (22 to 51 minutes)

Figure 27. GM-Ratemeter-Recorder response for nonisothermal study number 3 (-8 to 25 minutes)

Figure 28. GM-Ratemeter-Recorder response for nonisothermal study number 3 (25 to 57 minutes)

GM Detector Counting Data. The GM Detector-Ratemeter-Recorder trace obtained during the nonisothermal study and the observed and reduced counting rates determined from the recorder trace are presented in this section.

TABLE 9. GM DETECTOR COUNTING DATA FOR THE
NONISOTHERMAL STUDIES

Row	(1) Study Number
	(2) Date of Study
Column	(1) Time - min
	(2) Average Net Value from Recorder Trace Related to a Full Scale Deflection of 10K cpm Corrected for the Volume of the Coil (51.1 cm^3) and the Density of Circulating Liquid Mercury - net cpm/g of circulating liquid mercury
	(3) "Smooth Curve" Value Obtained by Fitting a Smooth Curve to a Plot of the Average Values in Column (2) versus Time (and Correcting for the Fact that the Values in Column (2) Represent a Circulating Radioactive Liquid which is Not Well Mixed) - net cpm/g
	(4) a_1/a_b Value for the GM Detector System (Obtained by Dividing Each Value in Column (3) by the Calculated Value of a_b Related to the GM Detector System)

TABLE 9--continued.

(1)	1			2			3		
(2)	12-26-69			1-23-70			1-24-70		
(1)	(2)	(3)	(4)	(2)	(3)	(4)	(2)	(3)	(4)
0	0.01			0.00	0.00	0.000	0.04	0.00	0.000
1	0.05	0.00	0.000	0.01	0.12	0.013	0.00	0.06	0.007
2	0.17	0.40	0.020	0.05	0.35	0.037	0.00	0.32	0.035
3	0.85	0.82	0.041	0.73	0.55	0.058	0.41	0.55	0.060
4	1.34	1.25	0.062	0.92	0.78	0.082	1.02	0.74	0.081
5	1.85	1.63	0.080	1.18	0.98	0.104	1.02	0.89	0.097
6	2.03	2.00	0.098	1.26	1.17	0.123	1.34	1.12	0.122
7	2.61	2.32	0.114	1.31	1.35	0.142	1.32	1.28	0.140
8	2.80	2.66	0.131	1.50	1.50	0.158	1.55	1.42	0.155
9	2.95	2.95	0.145	1.61	1.64	0.173	1.66	1.54	0.168
10	3.55	3.22	0.159	1.87	1.79	0.188	1.88	1.66	0.180
11	3.78	3.51	0.173	2.01	1.91	0.201	1.97	1.78	0.194
12	3.87	3.76	0.185	2.05	2.04	0.214	2.21	1.91	0.208
13	4.16	4.02	0.198	2.14	2.16	0.227	2.25	2.03	0.222
14	4.72	4.27	0.210	2.34	2.27	0.239	2.41	2.14	0.234
15	4.86	4.55	0.224	2.57	2.37	0.250	2.51	2.25	0.246
16	4.97	4.78	0.235	2.60	2.48	0.261	2.80	2.35	0.256
17	5.29	5.00	0.246	2.67	2.58	0.272	2.88	2.45	0.268
18	5.59	5.21	0.257	2.77	2.70	0.284	2.83	2.56	0.280
19	5.86			2.94	2.79	0.294	3.11	2.64	0.288
20	5.92	5.60	0.276	3.05	2.89	0.304	3.21	2.73	0.298
21	5.92			3.13	2.98	0.314	3.27		

TABLE 9--continued.

(1)	1	2	3	(1)	1	2	3
(2)	12-26-69	1-23-70	1-24-70	(2)	12-26-69	1-23-70	1-24-70
(1)	(2)	(2)	(2)	(1)	(2)	(2)	(2)
22	6.13	3.29	3.36	41	9.27	4.12	4.71
23	6.61	3.36	3.43	42	9.36	4.08	4.63
24	6.53	3.43	3.48	43	9.36	4.06	4.71
25	6.57	3.38	3.66	44	9.40	4.10	4.89
26	6.78	3.58	3.79	45	9.32	4.08	4.86
27	7.21	3.71	3.81	46	9.57	4.07	4.92
28	7.49	3.74	3.85	47	9.66	4.06	4.97
29	7.52	3.86	3.90	48	9.72	4.06	4.87
30	7.27	3.89	3.81	49	9.66	4.06	4.67
31	7.57	3.86	3.94	50	9.43	4.06	4.53
32	8.06	4.01	4.02	51	9.13	4.06	4.38
33	8.02	4.03	4.09	52	9.06		4.28
34	7.93	4.09	4.16	53	9.06		4.28
35	8.23	4.14	4.23	54	9.06		4.27
36	8.51	4.17	4.25	55	9.06		4.28
37	8.42	4.21	4.25	56	9.06		4.29
38	8.51	4.24	4.31	57			4.33
39	8.96	4.24	4.39	58			4.33
40	8.98	4.20	4.58				

Sample Calculation

To illustrate how the values tabulated in Table 5 (Chapter V) are determined, sample calculations are performed in this section employing Study Number 1 (12-26-69).

Determination of a_1/a_b . The GM Detector-Ratemeter-Recorder trace for Study Number 1 is shown in Figures 23 and 24. From this recorder trace, the average counting rates observed and recorded at predetermined time intervals are corrected for the background counting rate and the amount of circulating liquid mercury in the coil around the GM detector (see Figure 11 or 12). A smooth curve drawn for a plot of these net counting rates per gram of circulating liquid mercury (tabulated in Column 2 of Table 9), as a function of time, exhibits the same general shape as that of the original recorder trace. As with the original recorder trace, a time lag exists between the time that the cover valve is removed and the radioactivity is detected by the GM tube. Also, a "well-mixed" circulating liquid is observed after the cover valve is closed. From this smooth curve, the specific activities observed and recorded at predetermined time intervals are reduced to a "well-mixed" condition by multiplying each by the ratio of the "well-mixed" value to that observed at the instant the cover valve is closed (Column 3, Table 9). These specific activities tabulated in Column 3 of Table 9 represent a_1 for the GM detector.

Weighed samples of the circulating liquid collected after the cover valve is closed are counted in the shielded 3x3 NaI(Tl) Crystal-

Multichannel Analyzer (see Table 8). The specific activity tabulated in Column 9 of Table 8 represents a_1 for the crystal detector. A conversion from one detector system to the other is obtained from the ratio of " a_1 for the GM detector" to " a_1 for the crystal detector," or

$$[(9.06 \text{ cpm/g}) / (2.68 \times 10^4 \text{ cpm/g})] = 3.38 \times 10^{-4}$$

Weighed samples of the mercury in the boiler collected before and after the exchange process are counted in the shielded 3x3 NaI(Tl) Crystal-Multichannel Analyzer (see Table 8). The specific activity tabulated in Column 9 of Table 8 represents a_b for the crystal detector. A specific activity a_b for the GM detector is obtained by multiplying a_b for the crystal detector by the "GM to Crystal" conversion factor

$$(6.00 \times 10^4 \text{ cpm/g}) (3.38 \times 10^{-4}) = 2.03 \times 10^1 \text{ cpm/g}$$

This value of a_b for the GM detector is used to determine the a_1/a_b values tabulated in Column 4 of Table 9.

Slope Determination. The slope of the a_1/a_b versus time curve at time $t = 0$ (for the GM detector) shown in Figure 29 is $2.20 \times 10^{-2} \text{ min}^{-1}$.

Figure 29. $\left\{ \frac{a_1}{a_b} \right\}_{GM \text{ net}}$ as a function of time for nonisothermal study number 1

Alternate Slope Determination. Since $\left\{ [d(a_1/a_b)]/[dt] \right\}_{t=0}$ is desired, and the corrections cited affect the magnitude of a_1 and not its time rate of change, a simplified method was used as a check. This involved taking the slope of the initial portion of the GM-Ratemeter-Recorder response (Figure 23, 3K Full Scale).

The "base-line" is established by drawing a horizontal straight line through the averaged background counting rate (-8 to 0 minutes). This curve is intersected by an "average" curve drawn through the "3K Full Scale" response. The slope of the tangent drawn at the point of intersection is

$[(9.04 - 1.64) \text{ divisions}]/[(9 - 2) \text{ minutes}] = 1.058 \text{ divisions/minute}$ relative to a full scale response of 3K (for the circulating liquid mercury), where

$$(1 \text{ division for 3K}) = (0.3 \text{ division for 10K}) = 300 \text{ cpm/division}$$

Accounting for the mass of the circulating liquid mercury in the tube coiled around the GM detector

$$(300 \text{ cpm/division})/[(51.1 \text{ cm}^3)(13.2 \text{ g/cm}^3)] = 0.445 \text{ cpm/(g division)}$$

The specific activity (a_b) related to the GM detector is 20.3 cpm/g, and

$$[0.445 \text{ cpm/(g division)}]/[20.3 \text{ cpm/g}] = 0.02192 \text{ per division}$$

Hence, the slope of the tangent drawn at the point of intersection is

$$\left\{ [d(a_1/a_b)]/[dt] \right\}_{t=0} = (0.02192/\text{division})(1.058 \text{ division/min}) =$$

$$= 0.02319/\text{min}$$

The percent difference between this value and that calculated in the previous section is -5.4%.

Determination of $\Gamma\sigma_c$. Employing equation (20) where

$$T_v = 181^\circ\text{C}$$

and

$$P_v = 9.14 \text{ mm of Hg}$$

the condensing mass flux (G_c) is found to be $2.13 \times 10^1 \text{ g/(cm}^2 \text{ min)}$.

Substitution of this value and

$$L_o = 1006 \text{ g}$$

$$\left[\frac{d(a_1/a_b)}{dt} \right]_{t=0} = 2.20 \times 10^{-2} \text{ min}^{-1}$$

A is 1.6 cm^2 and $a_v/a_b = 1 + r/G^*A^* = 1 + (0.011)/(21.3)(4.93) = 1.048$ (found in Appendix C) into equation (66) gives $\Gamma\sigma_c = 6.2 \times 10^{-1}$.

Determination of σ_c . To determine σ_c it is necessary to evaluate Γ . The only unknown term in equation (23) is G_{net} which is given by

$$G_{\text{net}} = \frac{\Delta L}{A \Delta t}$$

where

$$\Delta L = (L_o + \Delta L) - (L_o) = (1238 - 1006) \text{ g} = 232 \text{ g}$$

$$\Delta t = 46.5 \text{ min}$$

$$A \approx 1.6 \text{ cm}^2$$

Hence

$$G_{\text{net}} = 3.1 \text{ g/(cm}^2 \text{ min)}$$

Using this value for G_{net} as well as those previously mentioned for T_v and P_v gives a value for Γ of 1.07.

Hence,

$$\sigma_c = [(6.2 \times 10^{-1})/\Gamma] = [(6.2 \times 10^{-1})/(1.07)]$$

$$\sigma_c = 0.58$$

Determination of σ_e . The numerical value of the evaporation coefficient is obtained through the use of equation (67). The evaporation mass flux given by equation (21) is 1.80×10^1 g/(cm² min), where

$$T_s = 176^\circ\text{C}$$

and $P_s = 7.68$ mm of Hg.

Hence, $\sigma_e = 0.90$.

Determination of the Heat Flux. The heat flux (q) for interfacial energy transfer is computed from equation (69), where the values of T_v , T_s and G_{net} are given above. The latent heat of condensation and specific heat are 70.5 (g-cal)/g and 0.025 g-cal/g^{°C}, respectively.

Hence

$$q = 2.2 \times 10^2 \text{ (g-cal)/(cm}^2 \text{ min)}$$

Determination of h_i . The interfacial heat transfer coefficient between the vapor and liquid phases is obtained from equation (70). Using the values of G_{net} , λ , T_v and T_s listed above gives a value of

$$h_i = 4.4 \times 10^1 \text{ (g-cal)/(cm}^2 \text{ min } ^\circ\text{C)}$$

Calculation of Complete Curve of Figure 29. A calculation of the complete a_1/a_b curve shown in Figure 29 is accomplished by coupling a mass balance on the radioactive mercury in the boiler (see Appendix C) with a mass balance on the radioactive mercury in the circulating liquid phase given by equation (64). Integration of equation (64) under the assumption that the mass fraction in the vapor phase is constant, yields

$$\frac{x_1}{x_v} = \left[\frac{\alpha_c}{r - \alpha_e} \right] \left[1 - \left(1 + \frac{rt}{L_o} \right)^{\frac{\alpha_e - r}{r}} \right]$$

where $r = G_{net} A_2$
 $\alpha_e = \sigma_e G_e A_2$
 $\alpha_c = \Gamma \sigma_c G_c A$

and A_2 refers to the circulating liquid-vapor exposure area.

From Appendix C, assuming that $x_v = x_b = \text{constant}$

$$(x_v/x_b) = 1 + (r/G^*A^*)$$

Substitution of numerical values from Table 5 for Study Number 1

(remembering the sign convention, and using $\sigma_e = 1.00$) into the equations listed above when $t = 40$ minutes gives an (x_1/x_b) ratio of 0.44.

Calculations employing this approach provide results which satisfy the entire a_1/a_b curve, as indicated by the points marked "4" in Figure 29.

APPENDIX C

RADIOMERCURY IN THE BOILER FOR THE NONISOTHERMAL STUDY

Radiomercury in the Boiler. A mass balance on the radiomercury in the boiler is given by

$$\frac{d}{dt} (m_b x_b) = \Gamma \sigma_c G_c A^* x_v + \sigma_e G_e A^* x_b$$

The liquid in the boiler at any time $t \geq 0$ is

$$m_b = m_{b0} - rt$$

where m_{b0} represents the initial amount of liquid in the boiler, A^* represents the boiler liquid-vapor volume exposure area, and t is the elapsed time $t \geq 0$. Under the experimental conditions that the boiler and vapor volume are maintained in isothermal equilibrium

$$G_c = G_e = G^*$$

and $\Gamma \sigma_c = \sigma_e = \sigma = 1.00$

Thus,

$$(m_{b0} - rt) \dot{x}_b + x_b \dot{m}_b = G^* A^* x_v + G^* A^* x_b$$

Integration of this equation under the assumption that $x_v = \text{constant}$ leads directly to that shown on page 68.

$$(x_v/x_b) = 1 + (r/G^* A^*)$$

Dilution in the Boiler. Count rate measurements of the tagged

mercury in the boiler prior to the start of and after a run was completed provided identical (within the limits of experimental error) specific activity determinations, i.e., no dilution effect was noticeable during a run. Solution of the "mass balance on the radiomercury in the boiler" equation (see the preceding section) for the ratio x_b/x_{b0} as a function of time (where $x_b = x_v \neq \text{constant}$), resulted in an approximate 20% dilution of the tagged mercury at the end of a run. In an attempt to resolve this apparent anomaly, the molecular flow from the vapor volume to the boiler and to the circulating liquid is examined.

As a crude similarity to the nonisothermal experimental configuration it is assumed that the molecular flow from the vapor volume to the boiler and to the circulating liquid can be represented as "free-molecule" flow. Thus, the conductance is given by

$$F = K A v_a/4$$

where K , A , and v_a represent the Clausing's factor, cross-sectional area, and average molecular velocity, respectively. The inverse of conductance is the resistance to flow. The "boiler" (see Figure 12) is a long tube (I.D. of 3.5 cm) with an elbow located ~ 6 cm from the entrance to the vapor volume and ~ 25 cm above the liquid mercury surface in the boiler. Clausing's factors for the vapor volume to elbow distance, elbow, and elbow to liquid mercury surface distance are calculated to be 0.248, 0.0287, and 0.0695, respectively. Since the vapor-circulating liquid surface is represented by an aperture (I.D. of 1.2 cm), the Clausing's factor is taken to be unity.

The resistance to flow of a molecule leaving the vapor volume and reaching the liquid surface in the boiler is ~6 fold greater than that of a molecule leaving the vapor volume and reaching the circulating liquid. This crude approximation implies that a preferential flow direction existed, and it tends to explain the experimental fact that no dilution effect was noticeable during a run.

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