

STUDY OF TWO PHASE SYSTEMS OF LIQUID METALS

BY

MARAYAN SWAROOP SINGHAL

Thesis submitted to the Indian Institute of
Technology, Delhi for the Award of the
Degree of

DOCTOR OF PHILOSOPHY

Department of Physics
Indian Institute of Technology, Delhi

JULY 1976

PREFACE

Because of their important implications in the processes of vapour deposition of thin films, molecular distillation, absorption and innumerable other aspects of chemical, aerospace, and nuclear technology, the phenomenon of mass and heat transfer resulting from the evaporation or condensation of a pure substance has always been of interest to chemists, physicists, and engineers. During the last decade the liquid metals have been used as coolants in advanced nuclear reactors. This is due to their higher heat transfer rates at low pressures as compared to those of the conventional coolants. It is apparent that in the design of heat transfer equipments which use processes involving evaporation and condensation a knowledge of parameters which effect the heat transfer coefficients is needed. However, previous investigators have found that heat transfer coefficients associated with film condensation of saturated liquid metal vapours have values which are much lower than those predicted by Nusselts theory¹. Sukhatme² attributes these discrepancies to an interfacial resistance which becomes significant at low pressures. The presence of non-condensable gases during condensation of vapours can considerably reduce the heat transfer rates.

Molecules undergoing condensation or evaporation strike the liquid-vapour interphase from both directions. Some of these molecules are reflected back into their original phase. Only a fraction of these impinging molecules which pass from one phase into the other contribute to the net rate of

condensation or evaporation as the case may be. The processes of condensation and evaporation occur in two phase systems which are not in thermodynamic equilibrium. There is, however, a dilemma concerning the study of such systems; because most of the mathematical expressions used for non-equilibrium liquid-vapour systems have been derived assuming the systems to be at thermodynamic equilibrium. In order to bring about an agreement between these expressions and the experimental results, it was found necessary to introduce appropriate coefficients (known as evaporation and condensation coefficients). No work is available in the literature which analyses critically the above approach. This motivated the author to (a) study experimentally evaporation coefficient, (b) design and perform an experiment for systems in thermodynamical equilibrium and hence to check the theories given for such systems.

Numerous experimental methods for the determination of condensation and evaporation coefficients are known. The variation of evaporation and condensation coefficient with pressure for various substances have been reported previously. However, no satisfactory explanation for the variation of evaporation coefficient with vapour pressure is available. This is because in these studies the effect of temperature gradient which was always present in the vapour phase, has been ignored. In the present investigations the experiments were performed near isothermal conditions eliminating thereby the effect due to temperature gradient. The results so obtained represent the true variation of evaporation coefficient with pressure. The results are discussed in the thesis.

There are two basic theories on evaporation and condensation reported in the literature, one is based on kinetic theory of gases and the other on statistical approach. The kinetic theory of gases, in the present work, has been analysed from the very beginning and its validity has been examined very carefully. The formulae based on this theory have been checked for their limitations. Some important steps for the derivation of the formulae for the rate of evaporation based on statistical approach are also critically examined. A brief discussion on the theory based on reaction rates (transition state theory) used for analysing evaporation mechanism is also reported in this thesis.

In previous studies isothermal conditions were assumed during the evaporation process. In practice however, even if the experimental conditions are fairly isothermal, there is a net heat loss at the surface of the sample, which causes a temperature change at the vapour-liquid interface. The rate of evaporation or condensation depends on the temperature of the liquid surface. A good knowledge of the surface temperature is therefore essential in such studies. The surface temperature can not be measured very accurately by conventional methods, however, approximate estimate can be made by a theoretical analysis. In this work such theoretical details have been worked out. The results for mercury surface temperature are presented in this thesis.

During the course of above work, the system was accidentally contaminated by mineral oil which was used as a hot-bath. This contamination resulted in an interesting observation. It was

observed that in the presence of an oil contamination, the amount of the saturated vapours was much more than the theoretically calculated value. An investigation of this problem is reported in this work. For these studies mercury was evaporated in an oil contaminated closed system under isothermal conditions. It was observed that the surface of the evaporating mercury was covered by a very thin layers of oil and the rate of evaporation was observed to reduce, i.e., the value of the evaporation coefficient decreased. The contamination of the surface of the vapour chamber also influences the evaporation process. The mercury molecules are adsorbed on this surface during evaporation process and then are de-adsorbed during the collection of vapours. The amount of the adsorbed mercury molecules depends on the thickness of the oil layer.

At thermodynamic equilibrium the rate of evaporation is equal to the rate of condensation, i.e., there is an exchange of equal number of molecules between the liquid phase and the vapour phase. This process is known as Molecular Exchange Phenomenon. The exchange rate can be calculated provided exchange coefficient α is known. In this work an experimental set-up was designed to study this phenomenon. A mathematical expression to obtain the value of α was derived for the system used. Experimental results for mercury are given in this thesis. Factors which can influence this phenomenon were studied and results are reported in this work.

In the evaporation phenomenon there are two processes which take place simultaneously, transfer of molecules from liquid

phase to vapour phase, and the transfer of molecules from vapour phase to liquid phase. An attempt was made to set up an experiment which would separate out these two processes. For this purpose a liquid was taken in two containers, one containing radio-active form and the other stable form. The change in concentration of radio-active molecules were investigated in their common vapour phase. The results obtained are presented in this thesis. Necessary theory was developed to explain these results. Some theoretical calculations were also done to find the optimum mass of the radio-active liquid needed for molecular exchange phenomenon studies.

It was thought that the vibration in the liquid phase would greatly influence the molecular exchange phenomenon. An experimental setup was designed and used to study this effect. It was observed that the rate of molecular exchange in the presence of vibrations was always less than the value without vibrations. This trend is quite different from what is usually expected in such situations.

Another important aspect of the present work was to study the effect of the presence of foreign gas in the vapour phase. An inert gas was chosen as foreign gas because of its use, in conjunction with liquid metals, as coolants in nuclear power reactors. The value of the exchange coefficient at various pressures of the inert gas was determined. The value of the exchange coefficient decreases with the increase of the inert gas pressure and the extrapolated results show that it is zero when the pressure of the gas is equal to the saturated vapour pressure of the liquid used.

The thesis is written in four chapters, Chapter I deals with theoretical aspects of two phase systems and specific statement of the problem. Chapter II contains the experimental and theoretical studies on evaporation phenomenon. Chapter III discusses the studies related with molecular exchange phenomenon. Chapter IV records summary, conclusions and recommendations. A few appendices follow at the end.

The work to be reported in this thesis has resulted in the following publications:

1. "Evaporation of mercury into vacuum under isothermal conditions", Indian J. Phys., 46(1972), 499.
2. "Molecular exchange in liquid-vapour system", Indian J. Phys., 47 (1973), 89.
3. "Theoretical analysis on cooling of the surface during evaporation in a closed system", Indian J. Pure & Appl. Phys., 11(1973), 560.
4. "Effect of vapour phase diffusion on molecular exchange phenomenon", Indian J. Pure & Appl. Phys., 11 (1973), 728.
5. "Effect of liquid phase diffusion on molecular exchange phenomenon", Indian J. Pure & Appl. Phys., 11(1973), 731.
6. "A Study of molecular exchange phenomenon using radioactive mercury vapours", Indian J. Chem., 12, (1974), 167.
7. "Molecular exchange in liquid-vapour system in presence of inert gas", Indian J. Phys., 48 (1973), 744.
8. "Evaporation of mercury in an oil contaminated closed system", Indian J. Phys. 48 (1974), 983.
9. "A Study of molecular exchange phenomenon in presence of vibrations in the liquid phase", Indian J. Chem., 13(1975), 508.

10. "Theoretical analysis of molecular exchange phenomenon",
Indian J. Pure & Appl. Phys., 13 (1975), 846.

REFERENCES

1. Nusselt, W., Zeitschrift des Vereines Deutscher
Ingenieure, 60(1916), 541 & 569.
2. Sukhatme, S.P. and Rohsenow, W.M., Tech. Report No.
9167-27, Dept. of Mech. Eng., MIT, USA, (1964).

TABLE OF CONTENTS

Chapter		Page
	PREFACE ..	1
I	THEORETICAL ASPECTS OF TWO PHASE SYSTEMS ..	8
1.1	Introduction ..	8
1.2	Theories of Two Phase Systems ..	9
	References ..	18
II	EVAPORATION STUDIES ..	21
2.1	Introduction ..	21
2.2	Effect of Vapour Pressure on Evaporation Coefficient ..	22
2.3	Cooling of the Liquid Surface During Evaporation ..	31
2.4	Evaporation of Mercury in an Oil Contaminated System ..	38
	References ..	43
III	MOLECULAR EXCHANGE PHENOMENON ..	44
3.1	Introduction ..	44
3.2	Study of Molecular Exchange Phenomenon Using Radio-active Mercury as Liquid Phase ..	45
3.3	MEP in Presence of Vibrations in the Liquid Phase ..	59
3.4	MEP in Presence of Inert Gas ..	64
3.5	Effect of Concentration of Liquid Phase on MEP ..	67
3.6	Effect of Vapour Phase Diffusion on MEP ..	74

Chapter		Page
3.7	Effect of Liquid Phase Diffusion on MEP	79
	References	85
IV	CONCLUSIONS AND RECOMMENDATIONS	86
	Appendix-I	90
	Appendix-II	91
	Appendix-III	98