A REVIEW OF PARTICLE TRANSPORT
IN A CONDENSING STEAM ENVIRONMENT

T. W. Horst

JUNE 24, 1968

AEC RESEARCH &
DEVELOPMENT REPORT

BATTELLE NORTHWEST

BATTELLE MEMORIAL INSTITUTE PACIFIC NORTHWEST LABORATORY
BATTELLE BOULEVARD, P. O. BOX 500, RICHLAND, WASHINGTON 99352

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

PRINTED BY/ FOR THE U. S. ATOMIC ENERGY COMMISSION

3 3679 00061 1493

BNWL-848
UC-80, Reactor Technology

A REVIEW OF PARTICLE TRANSPORT IN A CONDENSING STEAM ENVIRONMENT

By

T. W. Horst

Particulate & Gaseous Waste Research
Atmospheric Sciences
ENVIRONMENTAL & RADIOLOGICAL SCIENCES

June 24, 1968

EPSC 00187-001
Date: 6/24/68

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

ABSTRACT

A review is made of aerosol transport and deposition in an environment consisting of a mixture of air and saturated steam adjacent to a heat transfer surface on which condensation is occurring. Basic information is drawn together regarding those phenomena which are unique to a condensing steam environment or are altered by the presence of condensing steam, with the view of prediction of particle transport from first principles. These phenomena include condensation in the presence of an inert gas, diffusiophoresis, thermophoresis, particle nucleation and growth, and Brownian and turbulent diffusion. Judgment is made of the probable relative importance of these various mechanisms for aerosol demobilization following a loss-of-coolant accident in a water cooled reactor. This compilation of background material is the base for an experimental and theoretical study to extend knowledge of particle transport in a condensing steam environment.

TABLE OF CONTENTS

	<u>PAGE</u>
<u>INTRODUCTION</u>	1
<u>SUMMARY AND CONCLUSIONS</u>	3
<u>DETERMINATION OF TEMPERATURE, VELOCITY, AND COMPOSITION PROFILES IN A CONDENSING STEAM SYSTEM</u>	5
<u>THE BOUNDARY VALUE PROBLEM</u>	5
<u>NUMERICAL BOUNDARY LAYER SOLUTION</u>	6
<u>EMPIRICAL SIMILARITY SOLUTION</u>	8
<u>A QUALITATIVE DESCRIPTION</u>	10
<u>DIFFUSIOPHORESIS AND THERMOPHORESIS</u>	10
<u>FREE MOLECULE REGIME</u>	15
<u><i>Diffusiophoresis</i></u>	15
<u><i>Thermophoresis</i></u>	17
<u><i>Simultaneous Diffusiophoresis and Thermophoresis</i></u>	18
<u><i>Experiments</i></u>	18
<u>TRANSITION REGIME</u>	18
<u>SLIP FLOW REGIME</u>	19
<u><i>Thermophoresis</i></u>	19
<u><i>Diffusiophoresis</i></u>	21
<u>CONDENSATION ON WATER DROPS AND AEROSOL PARTICLES</u>	22
<u>HOMOGENEOUS NUCLEATION</u>	23
<u>NUCLEATION BY INSOLUBLE PARTICLES</u>	23
<u>NUCLEATION BY SOLUBLE PARTICLES</u>	24
<u>CONDENSATION TO DROPS</u>	27
<u>PARTICLE SIZE DEPENDENCE OF DIFFUSION PROCESSES</u>	28
<u>MOLECULAR DIFFUSION</u>	28
<u>TURBULENT DIFFUSION</u>	29
<u>RESEARCH ON PARTICLE TRANSPORT IN CONDENSING STEAM SYSTEMS</u>	31
<u>THE WORK OF GOLDSMITH</u>	31
<u>PARTICLE REMOVAL IN STEAM SUPPRESSION SYSTEMS</u>	32
<u>PARTICLE REMOVAL IN CONTAINMENT VESSELS</u>	33
<u>ACKNOWLEDGEMENTS</u>	34
<u>NOMENCLATURE</u>	35
<u>REFERENCES</u>	37

LIST OF FIGURES

	<u>Page</u>
<u>Figure 1</u> Phenomena Accompanying Steam Condensation	11
<u>Figure 2</u> Diffusiophoresis During Steam Condensation	13
<u>Figure 3</u> Thermophoresis During Steam Condensation	14
<u>Figure 4</u> Saturation Ratio Required to Nucleate a Critical Drop in One Second on an Insoluble Particle Of Radius R_0 and with Surface Prop- erties Defined by $m = \cos \phi$. Ref. [36]	25
<u>Figure 5</u> Equilibrium Supersaturation as a Function of Droplet Radius for Water Droplets, Each Containing Mass m of Solute of Molecular Weight M and Van't Hoff Factor i . Ref. [38]	26

A REVIEW OF PARTICLE TRANSPORT IN A CONDENSING STEAM ENVIRONMENT

T. W. Horst

INTRODUCTION

Economic considerations must be reconciled with those of safety during the design and siting of nuclear reactors. However, a lack of definitive knowledge of the consequences of a reactor accident has forced the use in many cases of extremely conservative safety standards. The transport and deposition of the radioactive aerosol which is expected to be generated by fuel overheating following an accident, is one area where better understanding could lead to more efficient reactor design and more realistic predictions of post accident conditions. This report focuses particularly on the fundamental understanding of those removal mechanisms which owe their existence to or are altered by the presence of condensing steam.

The maximum credible accident for water cooled reactors is the loss-of-coolant accident. The events which would take place are rupture of the primary coolant circuit and loss of coolant, core heating by decay heat, cladding melting, fuel melting, fission product release, and fission product transport and dispersal throughout the containment system.^[1] The coolant is quickly vaporized subsequent to the rupture of the circulation system due to its rapid depressurization. This implies a post accident containment system atmosphere consisting of a steam/air mixture contaminated by radioactive gases and aerosols. The bulk atmosphere quickly comes to equilibrium with the unevaporated coolant and hence is a saturated mixture.

Condensation of steam occurs when this saturated atmosphere has access to a cooler surface, such as the walls of the containment vessel. As soon as a film of liquid condensate forms and a quasi-steady state obtains, the conditions adjacent to the wall are also those of saturation but at a lower temperature than in the bulk of the steam. Consequently gradients of temperature and composition (ratio of steam to air) are established normal to the surface.

These temperature and composition gradients are the direct cause of forces on the aerosol particles. A particle in a temperature gradient experiences a force proportional to the gradient which drives the particle away from the

warmer region. Likewise, a particle which is located in a region in which diffusion is taking place is driven with the molecular flux by a force proportional to the partial pressure gradient. These phenomena are known, respectively, as thermophoresis and diffusiophoresis.

The temperature and composition differences between the bulk of the mixture and that close to the wall would also imply differences in the density of the mixture. This density variation causes buoyancy forces and gives rise to natural convective flow down the walls. The convective flow would contribute to the mixing in the containment vessel and would enhance the supply of aerosol near the wall. Further, when this flow becomes turbulent it would be the source of an added deposition mechanism.

It is conceivable that the profiles of temperature and humidity are such that a supersaturated condition would occur in the boundary layer between the wall and the bulk of the system. If supersaturation occurs, condensation might also take place on particles present in this region. Even undersaturation could result in limited condensation to particles provided that they are soluble. Any increase in particle size and mass due to condensation could have an appreciable effect on the subsequent trajectory of the particle. Its gravitational settling velocity would increase. Due to its increased inertia, the particle would now move in a path of smaller curvature when accelerated. Inertial deposition to structures and spray drops would thus become more prominent, particularly if turbulent conditions were present.

In addition to natural condensation on the containment walls, engineered safeguards designed to suppress pressure build-up will also cause appreciable condensation. Condensation on spray drops would alter their efficiency for collecting aerosol particles due to the increased drop size and by providing additional collection forces. Condensation of steam/air mixtures upon injection into water pools could rapidly demobilize particles carried along with the mixture. Ice surfaces would provide a situation similar to that already discussed for the containment walls, the primary difference being a lower wall temperature.

Thus, we see that the condensing steam/air mixture plays an important role in the transport and deposition of radioactive aerosols subsequent to a reactor accident. All of the phenomena mentioned above have been studied both theoretically and experimentally, but little is known about what happens

in a system in which they are all occurring simultaneously or about which effects predominate under realistic circumstances. These effects of steam condensation need to be studied further. As a preliminary, a review is here made of current knowledge pertaining to particle removal in a condensing steam environment.

SUMMARY AND CONCLUSIONS

The ultimate goal of a study of particle transport in a condensing steam system would be the development of the ability to predict the rate at which particles are demobilized, given the boundary conditions of the system and a description of the particles. Understanding of the forces enhancing particle removal should permit optimum advantage to be taken of these processes during the design of reactor containment safeguards. This ability depends on a detailed knowledge of the phenomena occurring in a condensing steam system.

Research performed to date has demonstrated the effectiveness of condensing steam for removing particles and has given an order-of-magnitude understanding of the processes. However, detailed explanation of the measurements made and extrapolation of them to actual conditions will require more precise experimentation and improved modeling.

Except for the case of turbulent deposition, a fair understanding appears to exist of the forces acting on the particles. For example, given a temperature gradient, the thermophoretic velocity of a known aerosol particle can be predicted. The nucleation and growth of the particles due to condensation can also be calculated if the environmental conditions are known. However, there is little knowledge of the combined effect of all of these phenomena. Moreover, little is known of the values taken on in a condensing steam environment by the quantities, such as temperature gradients and convective velocities, that control particle transport.

The implied approach of a study of particle deposition in a condensing steam environment is an integration of all of those particle transport mechanisms important in such a system, but this review indicates that a necessary initial step must be an extension of the understanding of mass and heat transport and the resulting temperatures, vapor pressures, and fluid velocities in a condensing steam system. When the differences in density between the bulk

atmosphere and that next to the wall are such that the convective flow down the wall is laminar, the integrated boundary layer equations should give adequate profiles. However, conditions following a loss-of-coolant accident appear to be such that most of the boundary layer along the containment wall will be in turbulent flow. For this case it will probably be necessary to resort to experimental measurements and quasi-empirical theories. Similarly, the flow in the vicinity of a falling drop is not well known and an empirical approach is necessary to give more than an approximate description of phenomena occurring near it.

A more definitive answer to the question of the relative importance of the various particle transport mechanisms which occur will only come from further research, but this study gives some indications of what will be found. Thermophoresis appears to be negligible compared to diffusiophoresis, including in this term the effect of the bulk flow of water vapor towards a condensing surface. In laminar flow the dominant mechanism for deposition to containment walls will thus be diffusiophoresis. Gravitational settling will also be important for horizontal surfaces. In turbulent flow the inertial deposition of the particles may completely overshadow all other effects, except possibly the movement of the particles with the bulk flow of water vapor.

All of these forces depend on particle size to some extent, most prominently the last two. Thus, particle growth due to condensation must be taken into account. Nucleation and condensation will occur most readily on soluble particles and on insoluble particles with a small contact or wetting angle.

Due to the fact that the quantities governing the forces on the particles are all closely interrelated, it may be possible to correlate particle deposition with a single parameter of the condensing steam system. The most obvious and easily measured of these is the condensation rate. The force that governs the convective flow is that of gravity and depends on the fluid density. This in turn depends on the temperature and composition of the steam/air mixture. Obviously the composition and temperature profiles are unique functions of the boundary conditions, which in turn depend on the water vapor and heat fluxes. Since the heat flux at the wall is mainly due to the latent heat given up by the condensing vapor, the condensation rate parameterizes everything that is occurring in the bulk. If it also parameterizes particle deposition, the condensation rate may be useful for extrapolation of particle deposition rates to systems other than those in which they are measured.

DETERMINATION OF TEMPERATURE, VELOCITY, AND COMPOSITION
PROFILES IN A CONDENSING STEAM SYSTEM

THE BOUNDARY VALUE PROBLEM

Knowledge of the velocity, temperature, and composition profiles normal to a surface on which steam is condensing is essential for calculation of the forces acting on particles in the system and for calculation of the rates of condensation to the surface and to the particles. These profiles are uniquely determined by the laws of conservation of mass, momentum, and energy, and by the conditions at the boundaries of the system. The conservation laws lead to the familiar partial differential equations of transport theory^[2]: the continuity equation,*

$$\frac{\partial}{\partial t} \rho + \nabla \cdot (\rho \vec{u}) = 0, \quad (1)$$

the equation of motion,

$$\rho \left(\frac{\partial}{\partial t} \vec{u} + \vec{u} \cdot \nabla \vec{u} \right) = \rho \vec{g} - \nabla p - \nabla \cdot \vec{\tau}, \quad (2)$$

the diffusion equation,

$$\frac{\partial}{\partial t} \rho_1 + \nabla \cdot \rho_1 \vec{u} = \nabla \cdot \rho D_{12} \nabla W_1, \quad (3)$$

and the conduction equation,

$$\rho C_p \left(\frac{\partial T}{\partial t} + \vec{u} \cdot \nabla T \right) = \nabla \cdot \lambda \nabla T. \quad (4)$$

The boundary conditions can be obtained by visualizing the system as one in which the temperature and composition are held constant in the bulk of the steam/air mixture, the temperature is held constant on the cooler surface on which condensation occurs, and the total pressure is held constant throughout the system. In the steady state a film of condensate is assumed to form and flow down the wall. The thickness of the film, δ , and the condensation rate, m'' , are related by using a mass balance and the simple theory of the flow of thin liquid films due to Nusslet.^[3] This theory neglects the effects of fluid acceleration and interfacial shear. The film thickness is found to be a function of x , the distance down the wall,

* Symbols are defined on pages 35-36.

$$\delta^3(x) = \frac{3}{g} \left(\frac{v}{\rho} \right)_L \int_0^x m''(\xi) d\xi, \quad (5)$$

and so also is the velocity of the gas-liquid interface,

$$u(x) = g\delta^2/2 v_L. \quad (6)$$

A heat balance can be used to also relate the heat flux out of the gas, q'' , to the film thickness,

$$q''(x) = \lambda_L (T_i - T_w)/\delta. \quad (7)$$

Here we have neglected energy convection in the liquid. Specification that the steam/air mixture at the surface of the liquid film is saturated, then, completes the boundary conditions by relating the surface composition to the surface temperature.

It is, however, far from trivial to now proceed to solve this boundary value problem since the transport equations are coupled to each other. This coupling occurs both due to the convective terms in the diffusion and conduction equations and due to the fact that temperature and composition differences give rise to buoyancy forces in the equation of motion. There is, in addition, a contribution to the diffusion due to temperature gradients and a contribution to the energy flux due to composition differences, both of which have been neglected in writing equations (3) and (4). Therefore, in order to predict the behavior of a condensing steam system, we must make approximations which reduce the set of equations to a tractable form but which are hopefully also realistic approximations.

NUMERICAL BOUNDARY LAYER SOLUTION

Sparrow and Lin^[4] have solved this problem for a vertical surface and laminar flow of the steam/air mixture by partial decoupling of the equations, use of a similarity transformation of variables, and numerical integration of the resulting simplified equations. Their method of analysis closely parallels that of standard boundary layer theory. The results are non-dimensional profiles normal to the wall of air and water vapor concentrations, of convective velocity down the wall, and of bulk flow velocity towards the wall. The simple

theory of Nusselt^[3], as outlined above, is used by Sparrow and Lin along with the neglect of all heat transfer to the film excepting that due to the release of the latent heat of condensation. These authors justified the use of Nusselt's theory by referring to the results of previous analyses^[5,6] which showed that the quantities neglected are truly small for the system being studied.

The transport equations applying in the steam/air mixture are decoupled by "retaining fluid property variations only where they play an essential role in establishing the transport process", i.e., in the buoyancy forces, and by further neglecting the effect of temperature differences compared with the effect of composition differences on buoyancy. This decoupling removes the conduction equation from the problem; and the continuity equation is identically satisfied by use of a stream function. Thus only equations (2) and (3) remain to be solved simultaneously. This is accomplished by using a transformation of variables to reduce them to ordinary differential equations and by then numerically integrating them.

Some of the restrictions of this analysis were lifted in a later paper by Minkowycz and Sparrow^[7] in which conductive heat transfer at the interface is accounted for and the fluid properties are allowed to retain their complete variability in both the liquid and gas phases. The conduction equation was solved simultaneously with the equation of motion and the diffusion equation, and the temperature profile normal to the wall is an added result. Another notable result comes from the solution of the boundary value problem in the liquid film. The authors find a reference temperature, T^* , at which to evaluate the physical properties of the liquid film,

$$T^* = T_w + 0.31 (T_i - T_w). \quad (8)$$

Using these constant properties in Nusselt's analysis gives the same value for the heat flux as in the variable property analysis.

The results of both of Sparrow's analyses, expressed as the ratio of heat transfer to the wall in a steam/air mixture to that in pure steam, agree well with an experimental study using horizontal cylinders. The only data presented were for very small percentages of air in the bulk atmosphere. Unfortunately no meaningful comparison is made between the analysis which more fully accounts for heat conduction and variable fluid properties and the simpler analysis which neglects these phenomena.

EMPIRICAL SIMILARITY SOLUTION

A good discussion of the standard empirical solution to this problem can be found in Jakob's^[8] or other heat transfer and mass transfer texts. Jakob analyzes the similarity between heat transport and mass transport and shows that with some approximations there is also similarity between pure heat transfer and simultaneous heat and mass transfer. The conditions at the surface are still related by Nusselt's theory.^[3] These surface conditions are used in conjunction with experimental measurements of heat transfer to predict the behavior of the condensing steam system.

In the absence of mass transfer, the heat transfer to a surface can be empirically correlated with the boundary conditions of the system through the Nusselt number. The Nusselt number for free convection is found to be a function of the Grashof number and the Prandtl number. For the laminar regime ($10^4 < N_{Gr} N_{Pr} < 10^9$) the local Nusselt number is^[9]

$$N_{Nu} = \frac{3}{4} (0.59) [N_{Gr} N_{Pr}]^{1/4}, \quad (9)$$

and for the turbulent regime ($10^9 < N_{Gr} N_{Pr} < 10^{12}$) it is^[9]

$$N_{Nu} = (0.13) [N_{Gr} N_{Pr}]^{1/3}. \quad (10)$$

The total heat flux to the wall of a condensing steam system is the sum of the heat flux due to the release of the latent heat of condensation and that occurring in the absence of mass transfer,

$$q''(x) = \Lambda m''(x) + \frac{\lambda}{x} N_{Nu} (T_{\infty} - T_i). \quad (11)$$

In the presence of simultaneous temperature and concentration gradients, however, the factor in the usual Grashof number due to buoyance, $g(T_i - T_{\infty})/T_{\infty}$, must be replaced by $g(M_{\infty} T_i - M_i T_{\infty})/M_i T_{\infty}$, in order to get the appropriate Nusselt number. The physical properties of the steam/air atmosphere are evaluated at the film temperature,

$$T_f = (T_{\infty} - T_i)/2. \quad (12)$$

The Nusselt number for mass transfer, the Sherwood number, can be obtained by utilizing the similarity between heat transfer and mass transfer,

$$N_{Sh}(N_{Gr}, N_{Sch}) = N_{Nu}(N_{Gr}, N_{Pr}). \quad (13)$$

This equation tells us to replace the Prandtl number by the Schmidt number in equations (9) and (10). Jakob warns, however, that the contribution of the bulk flow to the mass transport has no analogue in heat transfer. It must be a post-hoc addition to the similarity analysis, as is the contribution of the latent heat of condensation to the heat flux. The resulting mass flux of condensate to the wall is

$$m''(x) = \left(\frac{p_1}{p_2} + 1 \right)_i \left(\frac{D_{12}}{T} \right)_f \frac{m_1}{k} \frac{N_{Sh}}{x} [(p_1)_\infty - (p_1)_i] \quad (14)$$

where the numbered subscripts refer to the water vapor (1) and air (2).

The basis of the similarity approach is that by non-dimensionalizing the temperature and concentration variables, the conduction equation and equation of motion in a one-component system become identical with the diffusion equation and equation of motion in an isothermal system. Thus if the non-dimensionalized boundary conditions are also the same for both systems, the similarity relation, equation (13), holds true. For simultaneous heat and mass transfer we must also have identical systems of equations to use a similarity argument. For this to be true the heat and mass transfer must also be independent of each other. However, even if we neglect the effect of temperature gradients on diffusion and the effect of concentration differences on energy transport, the two processes are coupled through their mutual effect on buoyancy. Fortunately, as Sparrow and Lin contend^[4], the influence of temperature may be small compared with that of composition and we have satisfied the requirements for similarity of the differential equations. Another difficulty arises from the non-similar boundary conditions, however: the finite velocity normal to the wall caused by the mass transfer does not exist in pure heat transfer and alters the profiles of the quantities being transported. Bird, Stewart, and Lightfoot show how the Nusselt and Sherwood numbers may be corrected for this.^[10]

As indicated by equations (9) and (10), this empirical approach may be used to predict the heat and mass fluxes in both laminar and turbulent flow. In the laminar flow regime, knowledge of the physical transport coefficients will then give the gradients of temperature and composition at the interface and the bulk flow velocity towards the wall. These should be adequate to uniquely solve

the boundary layer equations for the appropriate profiles. For turbulent flow of the boundary layer, however, theory for the prediction of more than the fluxes at the interface is almost non-existent.

A QUALITATIVE DESCRIPTION

Figure 1 shows the qualitative picture obtained from the solution of this boundary value problem. Condensation to the cool wall causes depletion of the water vapor supply in its immediate vicinity. The resulting diffusion of water vapor down the water vapor gradient carries air with it to the wall. Since the air cannot condense, its concentration then increases at the wall, leading to a diffusion of air away from the wall. In the steady state the requirements of constant total pressure and zero air velocity normal to the interface lead to a unique bulk flow or Stefan flow towards the wall. This flow is such that the air is convected toward the wall at the same rate that it diffuses away from the wall. If we define the Stefan velocity to be the average molecular velocity, the molecular diffusion fluxes of air and water vapor are equal in a reference frame moving with this velocity. [See equations (22) and (23)]. The net flux of water vapor towards the wall due to diffusion and convection is equal to the condensation rate.

The difference in density between the bulk atmosphere and the boundary layer adjacent to the wall causes free convection in the system. Since the principal constituents of air (nitrogen, oxygen, argon, and carbon dioxide) all have molecular weights heavier than that of water vapor, the build-up of air adjacent to the wall increases the density of the atmosphere in the boundary layer and leads to convective flow down the wall. The cooler temperatures in the vicinity of the wall also lead to higher densities there and augment the effects of composition in causing flow down the wall.

DIFFUSIOPHORESIS AND THERMOPHORESIS

A recent review by Waldmann and Schmidt^[11] covers well the theoretical and experimental investigations of both thermophoresis and diffusiophoresis up to 1965 and parts of this section are a synopsis of that review. Diffusio-

STEAM CONDENSATION

FIGURE 1

PHENOMENA ACCOMPANYING STEAM CONDENSATION

phoresis is defined as the movement of a particle in an isothermal gas mixture due to concentration gradients of the component gases. Thermophoresis occurs when a particle moves due to thermal gradients in the gas in which it is suspended.

A qualitative understanding of diffusiophoresis and thermophoresis can be obtained from Figures 2 and 3. Here these phenomena are depicted as they would occur in a condensing steam system. Due to the concentration gradient of air, more air molecules strike the particle from the left than from the right and it experiences a resultant force to the right, in the direction of the diffusion of the air. The opposite is true for the water molecules whose collisions with the particle impart a net force to the left. Relative to a reference frame in which the diffusion fluxes are equal, the resulting velocity of the particle will be away from the wall since the air molecules are heavier than the water molecules and transfer more momentum per collision. However, since the velocity of this reference frame is towards the wall, the total particle velocity is also towards the wall.

Thermophoresis is similarly explained on the basis of molecular collisions. Since temperature is actually a measure of molecular velocities, a particle in a thermal gradient experiences a net thermophoretic force away from the region of higher temperature. The faster traveling molecules transfer larger amounts of momentum to the particle per collision and the net force on the particle will be towards the cool wall.

It is convenient in discussing diffusiophoresis and thermophoresis to recognize the existence of three dynamic regimes, each characterized by its Knudsen number (the dimensionless ratio of the molecular mean free path of the molecules of the surrounding fluid to the particle's size). When $N_{Kn} = \ell/R \gg 1$ we are in the free-molecule regime and here the particle is so small or the mean free path is so large that the particle has no influence on the velocity distribution of the fluid molecules. In the slip-flow regime, $N_{Kn} < 1$, the fluid is treated as a continuum and is materially influenced by the presence of the particle. Molecular effects are accounted for in the boundary conditions at the particle surface. Most difficult to analyze, finally, is the transition region where $N_{Kn} \sim 1$.

DIFFUSIOPHORESIS

FIGURE 2

DIFFUSIOPHORESIS DURING STEAM CONDENSATION

THERMOPHORESIS

FIGURE 3

THERMOPHORESIS DURING STEAM CONDENSATION

FREE MOLECULE REGIMEDiffusiophoresis

Both diffusiophoresis and thermophoresis are analyzed in the free molecule regime by calculating the momentum transferred per unit time to the particle by molecular collisions. The gas molecules are assumed to have the Chapman-Enskog^[12] velocity distribution. Waldmann^[13] and Bakanov and Derjaguin^[14] independently performed these calculations and found that for diffusiophoresis,

$$\vec{F} = -\frac{8}{3} R^2 \sum_i n_i \sqrt{2\pi m_i kT} (1 + \frac{\pi}{8} a_i) (\vec{v} - \vec{u}_i). \quad (15)$$

The assumption is made here that a fraction, a_i , of the gas molecules (the index i referring to a component of the mixture) is reflected thermally with a Maxwellian distribution and the remaining fraction $(1 - a_i)$ is reflected specularly. This diffusiophoretic force, $\vec{F}$, is simply the sum of the drag forces (as originally derived by Epstein^[15]) due to each of the constituent gases of the mixture. In the steady state, $\vec{F} = 0$, and

$$\vec{v} = \sum_i \alpha_i \vec{u}_i. \quad (16)$$

where

$$\alpha_i \equiv (1 + \frac{\pi}{8} a_i) \gamma_i \sqrt{m_i} / \left[\sum_K (1 + \frac{\pi}{8} a_K) \gamma_K \sqrt{m_K} \right], \quad (17)$$

$$\gamma_i \equiv n_i / \sum_j n_j = n_i / n. \quad (18)$$

For the special case of a vapor (1) diffusing through resting air (2),

$$\begin{aligned} \vec{u}_1 &= \vec{m}'' / \rho_1 \\ \vec{u}_2 &= 0, \end{aligned} \quad (19)$$

where $\vec{m}''$ is the mass flux of vapor (gm/cm²/sec) and is equal to the condensation rate for a steam/air condensation system in the steady state. Using equations (16) and (17) then,

$$\vec{v} = \frac{\gamma_1 \sqrt{m_1}}{\gamma_1 \sqrt{m_1} + \gamma_2 \sqrt{m_2}} (\vec{m}''/\rho_1), \quad (20)$$

where it is assumed that $a_1 = a_2$ in order to better display the functional dependence on the molecular masses and concentrations. According to equation (20) the particle is carried in the direction of the vapor flux but at some fraction of the velocity of the vapor molecules. Evaluation of this equation for typical steam/air condensation conditions gives

$$\vec{v} \sim 10^3 \vec{m}'' \text{ cm/sec.} \quad (21)$$

Equation (20) may be analyzed in the following manner.^[16] Mutual diffusion is defined relative to a reference frame moving with the average molecular velocity, $\vec{u}$, by the equation,

$$\gamma_1 \vec{u}'_1 = -\gamma_2 \vec{u}'_2 = -D_{12} \nabla \gamma_1, \quad (22)$$

where

$$\vec{u} = \gamma_1 \vec{u}'_1 + \gamma_2 \vec{u}'_2, \quad (23)$$

and velocities measured relative to $\vec{u}$ are denoted by primes, i.e.,

$$\vec{u}_i = \vec{u} + \vec{u}'_i. \quad (24)$$

Since $\vec{u}'_2$, the velocity of the air relative to a stationary reference frame, is zero, equations (22) and (24) give

$$\vec{u} = -\vec{u}'_2 = -\frac{1}{\gamma_2} D_{12} \nabla \gamma_1. \quad (25)$$

The air is moving towards the wall with the bulk motion of the gas at velocity $\vec{u}$, and is diffusing away from the wall at an effective velocity $\vec{u}'_2$ relative to $\vec{u}$. Using equation (22), now, in equation (16) gives the velocity of a particle relative to the bulk motion of the gas,

$$\vec{v}' = - \frac{\sqrt{m_1} - \sqrt{m_2}}{\gamma_1 \sqrt{m_1} + \gamma_2 \sqrt{m_2}} D_{12} \nabla \gamma_1. \quad (26)$$

This result implies that the particle moves in the direction of the diffusion flux of the heavier component, the air in the case of a steam/air mixture. Thus, the particle can be thought of as being carried with the bulk flow at a velocity $\vec{u}$, and as being retarded by the net effect of diffusion (See Figure 2), with the result that its diffusiophoretic velocity is given, as in equation (20), by

$$\vec{v} = \vec{u} + \vec{v}' = - \frac{\sqrt{m_1}}{\gamma_1 \sqrt{m_1} + \gamma_2 \sqrt{m_2}} \frac{1}{\gamma_2} D_{12} \nabla \gamma_1. \quad (27)$$

From this equation it can also be seen that the velocity is independent of particle size in the free molecule regime.

Thermophoresis

For thermophoresis in the free molecule regime Waldmann^[13] and Bakanov and Derjaguin^[14] calculate:

$$\vec{F} = - \frac{8}{3} R^2 \sqrt{2\pi m k T} [n(1 + \pi a/8)(\vec{v} - \vec{u}) + \frac{1}{5} \frac{\lambda_t}{kT} \nabla T]. \quad (28)$$

The first term is again the drag force and the second is the thermophoretic force. $\lambda_t = (15\mu \text{ k}/4m)$ is the heat conductivity for a monatomic gas. In the steady state, then, the particle velocity relative to a stationary gas is

$$\vec{v} = \frac{1}{5(1 + \pi a/8)} \frac{\lambda_t}{nkT} \nabla T. \quad (29)$$

The velocity is again independent of the particle size and is in the direction of decreasing temperature. For a condensing steam/air system this would be a velocity towards the cool wall. Assuming that the steam/air system is everywhere saturated, equation (29) can be evaluated for conditions identical to those of equation (21), giving

$$\vec{v} \sim 10 \frac{\text{cm}}{\text{m}} \text{ cm/sec.} \quad (30)$$

This is two orders of magnitude smaller than the diffusiophoretic velocity given by equation (21) and thus thermophoresis would be negligible for transport to a condensation surface in cases of interest to this study.

Simultaneous Diffusiophoresis and Thermophoresis

Waldman^[13] and Bakanov and Derjaguin^[14] also analyzed simultaneous diffusiophoresis and thermophoresis in the free molecule regime. The resultant velocity is that due to the sum of the drag forces and the thermophoretic forces for each component plus additional forces depending on the concentration gradients, $\nabla \gamma_i$. When the components have equal molecular masses these additional terms can be easily handled, but for the general case it is necessary to know the values of coefficients in the Chapman-Enskog velocity distribution function for an unequally heated gas mixture.

Experiments

Experiments have confirmed well the respective theories for $\ell/R \gg 1$ by agreeing with the predicted results within about 10% or better in most cases. Schmitt and Waldmann^[17] measured the diffusiophoretic velocities of oil drops in several gas mixtures and got good agreement for $\ell/R \sim 2$, their largest N_{Kn} . They used a modified form of Millikan's oil drop apparatus for their experiments. Schmitt^[18] also found thermophoretic velocities of oil drops (low conductivity) which agreed with free molecule theory when "a" was taken to be unity. Goldsmith and May^[19] obtained excellent agreement with theory for the diffusiophoretic velocity of an aerosol produced by passing air or oxygen over a heated nichrome wire, again corresponding to $N_{Kn} \sim 1$. They performed experiments for water vapor diffusing through air and helium, measuring the velocity through the deposition it caused on a plate normal to the concentration gradient.

It is noted here that experiments have shown the theory for the free molecule regime gives reasonably accurate results for a Knudsen number as low as two to five. This is useful since outside of the free molecule regime there has been a good deal of disagreement over both theoretical and experimental results.

TRANSITION REGIME

In the transition regime, $\ell/R \sim 1$, Brock^[20] has made a momentum transfer calculation for the thermophoretic force on a particle in a monatomic gas. Since the particle will now materially influence the velocity distribution of

the gas molecules, the velocity distribution must be found as the appropriate solution of the inhomogeneous steady-state Boltzmann equation. Brock was able to do this by using an approximate form for the Boltzmann equation which follows from the BGK^[21] model. His result can be expressed as a correction to the velocity in the free molecule regime,

$$\vec{v} = \vec{v}_{(eq. 29)} \exp(-\tau R/\ell), \quad (31)$$

where

$$\tau = 0.06 + 0.09 a + 0.28 a[1 - a\lambda/2\lambda_p]. \quad (32)$$

Here λ and λ_p are the thermal conductivities of the gas and particle, respectively. The alteration of the distribution function of the gas caused by the presence of the particle is seen to result in a lowering of the thermophoretic velocity with decreasing N_{Kn} . This relationship matches that measured experimentally for $0.2 < \ell/R < \infty$. Brock^[22] later extended the analysis to polyatomic gases with the relation,

$$\tau = 0.06 + 0.09 a + 0.28 a[1 - b\lambda/2\lambda_p], \quad (33)$$

where b , the thermal accommodation coefficient, has replaced a , the momentum accommodation coefficient, in the last term of equation (32). Comparison with the experiments of both Schmitt^[18] and Jacobsen and Brock^[23] indicates that τ includes the correct functional dependence on the parameters. Small errors in the numerical constants are possible, however, due to the inappropriate use of the BGK model in the case of simultaneous heat and momentum transfer.

No theoretical analyses in the transition regime have been reported for diffusiophoresis.

SLIP FLOW REGIME

Thermophoresis

In the slip flow regime, Epstein^[24] was the first to solve the hydrodynamic boundary value problem for thermophoresis, including in the analysis thermal creep at the surface of the particle. Brock^[25] has corrected Epstein's equation by additionally accounting for temperature and tangential velocity

discontinuities at the particle surface (temperature jump and velocity slip). Brock's result is

$$\vec{v} = - \frac{[1 + A\ell/R + B\ell/R \exp(-C\ell/R)] \cdot 2(\lambda + C_t\ell/R \lambda_p)}{(1 + 3C_m\ell/R)(2\lambda + \lambda_p + 2C_t\ell/R \lambda_p)} \frac{\lambda}{5nkT} \nabla T, \quad (34)$$

where A, B, and C are numerical constants obtained experimentally by Millikan^[26]. C_t and C_m are constants associated with the temperature jump and velocity slip and depend on the nature of the gas-particle interaction. In order to account for large departures from local equilibrium in the vicinity of a particle of very high thermal conductivity, Jacobsen and Brock^[23] later extended the analysis further. By using the third approximation to the solution of the Boltzmann equation, they derived a thermophoretic velocity which can be expressed in terms of the previous result, equation (34), and a multiplicative correction factor,

$$\vec{v} = \vec{v}_{(eq. 34)} [1 + 4/3 a_3 C_m \ell/R - (4/3 a_3 C_m \ell/R)/(\lambda/\lambda_p + C_t \ell/R)]. \quad (35)$$

where a_3 is a third order constant which they set equal to 2.4. Since this expression is highly sensitive to the values of the slip coefficients, Jacobsen and Brock limit its present usefulness to curve fitting.

Bakanov and Derjaguin^[27], on the other hand, claim that the method of Epstein and Brock is invalid. They calculated that Maxwell's expression for thermal creep is too large by a factor of 35 and hence this phenomenon is too small to account for thermophoresis. Instead they considered a term in the equation for the heat flux in the bulk of the gas which follows from the third order Chapman-Enskog theory. Applying Onsager's principle of symmetry of kinetic coefficients they calculated from this an expression for the thermophoretic velocity. Derjaguin and Yalamov^[28] later improved this by accounting for velocity slip and temperature jump and got:

$$\vec{v} = - 2/3 \frac{8\lambda + \lambda_p + (2C_t\ell/R) \lambda_p}{2\lambda + \lambda_p + (2C_t\ell/R) \lambda_p} \frac{\lambda}{5nkT} \nabla T. \quad (36)$$

In general this formula gives a velocity about twice that of Brock's. Both formulas reduce in the limit of $\ell/R \rightarrow \infty$ to $\sim \lambda/\lambda_t$ times the thermophoretic velocity for the free molecule regime. Thus, they roughly interpolate throughout the range of the Knudsen number.

Experiments by Schmitt^[18] with oil drops and by Schadt and Cadle^[29] with NaCl particles and with tricresyl phosphate drops (high conductivity, $0.05 < \ell/R < 1$) agree within about 20% with the predictions of Brock's theory, equation (34); and the measurement of Jacobsen and Brock^[23] with NaCl particles can be fit well with their second order slip flow equations, equation (35), for $0.06 < \ell/R < 0.7$ by choosing values of slip coefficients which differ slightly from the theoretical. Derjaguin, Storozhilova, and Rabinovich^[30], on the other hand, pointed to convective currents as an uncontrolled source of error in the Millikan condenser apparatus used by the previous researchers, devised new experiments, and obtained data for $0.15 < \ell/R < 0.5$ which confirms the Russian theory, equation (36). Measurements of thermophoresis by Goldsmith and May^[19] with the Nichrome aerosol ($N_{Kn} \sim 1$) appear to be bracketed by the opposing theories and are actually matched best by the free molecule theory, equation (29).

Brock^[20] feels that the origin of some of this disagreement in the slip-flow regime is due to ignoring the effect of the accommodation coefficient, a , and to incorrectly comparing results for monatomic and polyatomic cases. He concludes that surface effects are most noticeable in the transition regime and that hydrodynamic effects become important for $N_{Kn} < 0.2$.

Diffusiophoresis

Diffusiophoresis in the slip-flow regime is analyzed by Schmitt and Waldmann^[17] as a hydrodynamic problem, accounting for a finite gas velocity at the particle surface due to diffusion slip. For the case of a vapor (1) diffusing through a resting gas (2), they got

$$\vec{v} = - (1 + \sigma_{12} \gamma_2) \frac{D_{12}}{\gamma_2} \nabla \gamma_1, \quad (37)$$

where σ_{12} is a numerical constant called the diffusion slip factor. Kramers and Kistemaker^[31] and Brock^[32] have derived expressions for this factor, but for good agreement with experiment^[17] an empirical relation must be used,

$$\sigma_{12} = 0.95 \left(\frac{m_1 - m_2}{m_1 + m_2} \right) - 1.05 \left(\frac{d_1 - d_2}{d_1 + d_2} \right), \quad (38)$$

where d is the diameter of the gas molecule.

Derjaguin, Yalamov, and Storozhilova^[33] perform an analysis similar to the Russian theory for thermophoresis in the slip-flow regime. They get for diffusiophoresis,

$$\vec{v} = - \frac{\rho_1}{\rho} \frac{D_{12}}{\gamma_1} \nabla \gamma_1. \quad (39)$$

This equation is matched fairly well by their own experimental measurements for $\ell/R < 0.5$.

In summation, then, an aerosol particle in a condensing steam environment has forces acting on it due to thermophoresis and diffusiophoresis which enhance deposition on the condensation surface. Since for all N_{Kn} thermophoresis is proportional to $\nabla \ln T$ and diffusiophoresis is proportional to $\nabla \gamma_1$, the former is negligible compared to the latter in cases of interest to this study. The particles are carried toward the wall at a velocity which is less than that of the water vapor flux. The proportionality factor depends on the ratios of the molecular masses and the concentrations of the air and water vapor comprising the atmosphere of the system. Prediction of this proportionality factor can be made to an accuracy of about 10% in the free-molecule regime and perhaps to within a factor of two in the slip-flow regime. In the absence of better information, a smooth interpolation would be a first approximation in the transition regime.

CONDENSATION ON WATER DROPS AND AEROSOL PARTICLES

Water vapor may self nucleate and condense into drops in the absence of condensation nuclei if the degree of supersaturation is great enough. At lower supersaturations, however, a nucleating surface is necessary for condensation. The conditions necessary for condensation to occur on a surface are determined by applying equilibrium thermodynamics.^[34] A system is in stable (unstable) equilibrium at constant temperature when the Helmholtz free energy is minimized (maximized). This fact is used to find the equilibrium saturation ratio for a surface exposed to the vapor. (The saturation ratio is the ratio of the vapor pressure at the point in question to that of a plane water surface.) If, then, the saturation is greater than required for equilibrium, condensation occurs and will continue until equilibrium is again attained.

HOMOGENEOUS NUCLEATION

The equilibrium saturation ratio over a curved surface of pure water is given by the Kelvin equation,

$$S = \exp (2\sigma/Rn_L kT), \quad (40)$$

where R is the radius of curvature. Since this is a decreasing function of R it can be seen that, for a given saturation, water droplets grow by condensation if they are larger than a critical size,

$$R^* = 2\sigma/n_L kT \ln S. \quad (41)$$

As an example, a saturation ratio of 3.5 requires a critical drop of radius $10^{-3}\mu$ for equilibrium, while a saturation of 1.001 requires a drop of 1μ radius. The rate at which drops of this size form spontaneously from the vapor (nucleate) and begin to grow is given approximately by:^[35]

$$J = KN \exp [-\Delta G(R^*)/kT]. \quad (42)$$

Here N is the number of water molecules in the system and K is a kinetic factor describing the net effect of chance collisions on the aggregation of the molecules. The increase in free energy in going from the vapor to the liquid is

$$\Delta G(R^*) = 4/3 (\pi R^{*2} \sigma) = \frac{16\pi\sigma^3}{3[n_L kT \ln S]^2}. \quad (43)$$

Numerical evaluation of equation (42) for reasonable rates of formation shows that saturations exceeding 200% to 400% are necessary for self nucleation.

NUCLEATION BY INSOLUBLE PARTICLES

If an insoluble surface is present, drops of the critical radius will form on this surface at a higher rate than they will self nucleate. This is due to the fact that they have less volume and require fewer molecules than for the case of homogeneous nucleation; hence the free energy increase is smaller. Their volume is smaller because they can form as a partial sphere resting on the surface rather than the complete sphere necessary in the absence of the surface. Fletcher^[36] has derived an expression for this free energy increase as a function of the radius, R_0 , of the nucleating particle and of the contact angle, ϕ , between the water/air interface and the water/particle interface,

$$\Delta G(R^*) = \frac{16\pi\sigma^3}{3[n_L kT \ln S]^2} f(\cos \phi, R_O/R^*). \quad (44)$$

Here $f < 1$. In the presence of insoluble particles, the nucleation rate, equation (42), is proportional to the number of water molecules adsorbed per unit area of surface. Fletcher's calculations of the saturations required for a given particle to nucleate a drop of critical size in one second are shown in Figure 4. Since for the same rate of nucleation, lower saturations are required in the presence of particles than in their absence, this process will dominate that of homogeneous nucleation.

NUCLEATION BY SOLUBLE PARTICLES

Finally, if the particle is soluble, nucleation will occur even more readily. Raoult's law states that the equilibrium vapor pressure over a solution is less than that over pure water by a factor equal to the mole fraction of water in the solution. Using the van't Hoff factor, i , to take into account the dissociation of inorganic salts, Fletcher^[37] gets an expression for the equilibrium saturation ratio over a solution droplet composed of water of gram-molecular weight, M_O , and containing a mass, m , of a solute of gram-molecular weight M ,

$$S = \left(\exp \frac{2\sigma'}{n_L' kTR} \right) \left[1 + \frac{i m M_O}{M(4/3 \pi R'^3 \rho_L' - m)} \right]^{-1}. \quad (45)$$

Here the primes refer to the properties of the solution. Figure 5 is a numerical evaluation of this equation as presented graphically by Fletcher.^[38]

From Figure 5 it is apparent that soluble particles can act as nucleation sites without having to depend on chance agglomerations of water molecules to reach a critical size. If, for a given particle, the ambient supersaturation is below the peak of the equilibrium saturation curve, two equilibrium drop sizes are possible. The point to the left of the peak represents stable equilibrium. Growth by condensation will occur only if the ambient saturation increases or if statistical fluctuations in size cause the drop to reach the point on the curve to the right of the peak. If, however, the ambient saturation is above the peak of the curve, spontaneous nucleation, condensation, and growth will occur. Thus, soluble particles will serve as nucleation sites at very low supersaturation and will form drops much more readily than insoluble particles.

FIGURE 4

SATURATION RATIO REQUIRED TO NUCLEATE A CRITICAL DROP IN ONE SECOND ON AN INSOLUBLE PARTICLE OF RADIUS R_0 AND WITH SURFACE PROPERTIES DEFINED BY $m = \cos \phi$. Ref. [35]

FIGURE 5

EQUILIBRIUM SUPERSATURATION AS A FUNCTION OF DROPLET RADIUS FOR WATER DROPLETS, EACH CONTAINING MASS m OF SOLUTE OF MOLECULAR WEIGHT M AND VAN'T HOFF FACTOR i . Ref. [37]

CONDENSATION TO DROPS

After a drop of critical size has been nucleated, it grows at a rate determined by the ambient environment and the conditions at its surface, just as the condensation flux to a flat wall depends on these boundary conditions. The conditions far from the drop are taken from knowledge of the temperature and water vapor concentration profiles within the condensing steam environment. The temperature and partial pressure of the water vapor at the drop surface are related by assuming equilibrium saturation there and by making a balance between heat stored in the drop, heat brought to the drop by condensation, and heat transported by convection and conduction. In this case natural convection is negligible compared to forced convection, but the partial differential equations (1)-(4) cannot readily be solved due to their non-linearity and the geometry of the boundary conditions. Therefore empirical Nusselt and Sherwood numbers must again be used to calculate the fluxes of heat and mass to the drop.

For the case of a drop nucleated on a particle, the additional assumption of a quasi-steady state may be made and heat stored in the drop may be neglected. The results show that the aerosol grows at a rate roughly proportional to the inverse of its radius.^[39] Growth continues until the ambient vapor pressure is lowered to equilibrium due to condensation of the available water vapor or due to movement of the particle to a different region.

As noted earlier in this work, particle growth is important since gravitational settling and inertial deposition both increase with particle size. It is also important to the extent that diffusiophoresis and thermophoresis depend on particle size; and this becomes significant outside of the free molecule regime.

Spray drops, on the other hand, enter the system at a low temperature and conditions at their surface will change rapidly with time. Since, unlike the aerosol particles, it is not probable that they will enter a supersaturated region, their growth will be limited by their heat storage capacity and condensation will cease when they reach the temperature of the environment. Thus the change of the surface conditions with time and the heat stored within the drops must both be taken into account when determining their growth.

The final size of the spray drops determines their efficiency for collecting aerosols. Diffusiophoresis and thermophoresis during spray drop

growth also contribute to their washout of particles, but exact calculation of these phenomena is hindered again by our inability to solve equations (1) - (4). An approximate calculation of these phenomena can be made by assuming that the drop is stationary and by relating the particle velocity to the condensation rate as done in equations (21) and (30).

PARTICLE SIZE DEPENDENCE OF DIFFUSION PROCESSES

Up to this point we have considered only the ordered motions of a particle due to forces which can be attributed directly to vector quantities such as fluid velocity, temperature gradients, and water vapor gradients. The motion of a particle due to its diffusion is a random process and becomes ordered only when a large number of particles is considered. When the latter is the case, the force acting on the particles can be attributed to a gradient in the particle concentration. The random motion becomes ordered in the sense that the net movement of particles is away from a region of high concentration. This net flux of particles due to diffusion, $\vec{N}$, is expressed by Fick's law as,

$$\vec{N} = -K \nabla n. \quad (46)$$

MOLECULAR DIFFUSION

When the random force on a particle is due to the Brownian motion of the molecules of the fluid in which it finds itself, the coefficient of proportionality, K , is the molecular diffusivity, D . Einstein^[40] showed this to be,

$$D = kTL = \frac{kT}{6\pi\mu R} [1 + A\ell/R + B\ell/R \exp(-C R/\ell)]. \quad (47)$$

L is the mobility of the particle and is the ratio of the velocity of a particle to the drag force. (The factor in brackets is an experimental correction to the mobility due to Millikan.^[26] It is identical to that found in equation (34).) The mobility decreases with the particle size and hence condensation to a particle will decrease its motion due to Brownian diffusion. Brownian diffusion generally becomes negligible for micron size particles. For air at 23°C and normal atmospheric pressures, typical values for the molecular diffusivity are shown in Table I.^[41]

TABLE I
DEPENDENCE OF MOLECULAR DIFFUSIVITY AND RELAXATION
TIME ON PARTICLE SIZE

Particle Radius (μ)	Molecular Diffusivity ($\text{cm}^2\text{sec}^{-1}$)	Relaxation Time (sec)
10^{-3}	1.3×10^{-2}	1.3×10^{-9}
10^{-2}	1.4×10^{-4}	1.4×10^{-8}
10^{-1}	2.2×10^{-6}	2.3×10^{-7}
1	1.3×10^{-7}	1.3×10^{-5}
10	1.4×10^{-8}	1.2×10^{-3}

It is expected that Brownian diffusion will be of minor importance compared to the phenomena already discussed when considering deposition to structural surfaces. However, it may be of importance for the scavenging of submicron particles by spray drops.^[16]

TURBULENT DIFFUSION

Brownian diffusion will further be negligible compared to turbulent diffusion once turbulent flow becomes established. The random force on a particle in this instance is due to the fluctuating eddy velocity of the turbulent flow. A measure of the response of a particle to the drag force of a suddenly applied fluid velocity is its relaxation time,

$$\tau = mL = \frac{2}{9} \frac{R^2 \rho_p}{\mu} [1 + A\ell/R + B\ell/R \exp(-C\ell/\ell)], \quad (48)$$

where m is the mass of the particle and ρ_p is its density. This is the time necessary after sudden application of a force for a particle to attain a fraction $(1 - 1/e)$ of its terminal velocity for that force.^[42] Typical values of τ are given in Table I for unit density particles. Using this, the stop distance, d , of a particle with initial velocity, v_o , can be written as,

$$d = v_o \tau. \quad (49)$$

This is the distance which the particle would travel before it was brought to a complete stop by the fluid drag of a resting medium. The relaxation time, and hence also the stop distance, increases with particle size. If the scale

of the turbulent eddies are large compared with the stop distance, the particle velocities will closely conform to fluid velocities except near boundaries.^[43]

In turbulent flow the eddy velocities also transfer momentum, giving rise to turbulent stresses,

$$\tau_{xy} = - \overline{u'_x u'_y} \equiv - \rho \epsilon_{xy} \frac{\partial \bar{u}_x}{\partial y} \quad (50)$$

where the overbar indicates a time average and the primes indicate an instantaneous deviation from the average, i.e.,

$$u = \bar{u} + u'. \quad (51)$$

When the particle velocities correspond closely to the fluid velocities it is assumed that the eddy viscosity, ϵ , is identical to the turbulent diffusivity.^[43-47] Hence for turbulent diffusion,

$$\vec{N} = - \epsilon \nabla n. \quad (52)$$

Turbulent diffusion becomes important and the eddies normal to the wall are the dominating mechanisms for transport to the wall when ϵ is large compared to D . The effect of this is to keep the particles well mixed to within a very short distance of the wall, depending on the scale of turbulence. At the wall, however, $\epsilon_{xy} \rightarrow 0$ and some other mechanisms must be responsible for actual deposition on the wall. The identity of this mechanism depends on the stop distance. Once the particle attains a proximity to the wall equal to its local stop distance (determined by its local velocity normal to the wall) deposition is assumed inevitable. However, if the distance from the wall at which D becomes comparable to ϵ is larger than this free flight distance, Brownian diffusion will also be important for deposition. In any case, turbulent deposition is larger than that caused by Brownian diffusion alone, since the concentration gradient near the wall is greater due to the action of the turbulent eddies in bringing the bulk particle concentration closer to the wall.

The turbulent deposition rate depends strongly on the particle stop distance, and hence turbulent deposition, in contrast to Brownian diffusion, will increase with particle size and condensation on the particles can markedly alter their deposition. This relation between particle size and turbulent deposition rate has been well documented by experiment.^[43,44,47] Goldsmith and May^[19] use particle condensation to interpret experiments on turbulent

deposition in the presence of water vapor and temperature gradients. (This work will be discussed in the following section.)

Actual calculation of the diffusional deposition rates depends on knowledge of the particle size, concentration gradient, and, in the case of turbulent flow, the eddy viscosity. Particle size has been seen to depend on condensation to it. The particle concentration profile is a function of the diffusion and of the other forces acting on it. It must be determined by a relation similar to equation (3). Finally, the eddy viscosity may be determined by direct experiments, such as those of Laufer^[48], or by analysis of transport data, such as done by Lin, Moulton, and Putnam.^[49] However, it must be noted that no existing theory of turbulent deposition has had its general validity proven by experiment.

RESEARCH ON PARTICLE TRANSPORT IN CONDENSING STEAM SYSTEMS

Limited experimental data on the transport of particles in condensing steam systems is available, but the work which has been done does not quantitatively isolate the many phenomena which are occurring. With a few exceptions, the common approach has been to measure the efficiency of a system at removing particles as a function of system geometry, flow rates, or composition of the steam/air mixture, usually with the aim of optimizing the design of a certain piece of hardware.

THE WORK OF GOLDSMITH

The most definitive experiments are those of Goldsmith and coworkers. As a continuation of their experiments on particle deposition in thermal and water vapor gradients, they investigated the combined effect of thermophoresis and diffusiophoresis. Goldsmith and Stinchcombe^[50] report an experiment using NaCl solutions at different temperatures on opposite walls of a chamber to produce simultaneous temperature and vapor pressure gradients. These experiments showed the thermophoretic and diffusiophoretic velocities to be additive. Subsequent use of pure water surfaces at different temperatures, however, revealed that as the thermal gradient exceeded a critical value the deposition velocity increased to many times the sum of the velocities caused by diffusiophoresis and thermophoresis. Goldsmith and May^[19] explained this enhanced effect as

being due to the fact that the linear temperature and water vapor gradients are such that a layer of supersaturated vapor will be found within the chamber. In this region condensation on the submicron particles would cause them to grow to a size for which gravitational settling would be the predominant deposition mechanism. No direct confirmation of this hypothesis is presented.

Extension of these experiments^[50] to turbulent flow ($N_{Re} = 2 \times 10^4$) again revealed an enhanced effect when supersaturation was possible. These results were attributed^[19] to the increase in inertia accompanying the growth in size of the particles. The deposition rate was shown^[50] to be directly proportional to the rate of condensation,

$$\vec{v} \approx 6 \times 10^4 \frac{\text{m}}{\text{sec}}, \quad (53)$$

and may be compared with that calculated in equation (21) for diffusiophoresis alone.

The efficiency of a condensor scrubber apparatus for removing aerosols from a gas stream was also shown^[50] to increase with the condensation rate. Steam was added to the gas before it flowed through a tube with cooled walls and the particle concentration was monitored at both ends of the condensor tube. To a lesser degree, the removal efficiency also increased with the flow rate and the degree of cooling.*

Goldsmith and Stinchcombe^[50] also demonstrated qualitatively the efficiency of steam condensation for removing particles from the air. They monitored the particle concentration in an ordinary bathroom with a Pollak counter. Less than an hour after starting to run hot water into the bath tub, the particle concentration dropped to less than 30% of its original value.

PARTICLE REMOVAL IN STEAM SUPPRESSION SYSTEMS

Condensation on the particles was used to explain results obtained when investigating particle removal in steam pressure suppression systems. In the system in question, steam enters a cooling pond through a submerged tube known as a lute. This steam would result from a loss-of-coolant accident and hence could contain radioactive particles. Ashworth, Barton, and Robbins^[58] reported that zinc sulfide particles with a mean size of 2μ were reduced in number

* References [51] through [57] also deal with devices for the removal of aerosol contaminants from gas streams.

by a factor of 10^8 between the reactor vessel preceeding the lute and the air above the cooling pond. British researchers also reported high decontamination factors for a 0.06μ aerosol. Diffey, Rumary, Smith, and Stinchcombe^[59] reported reductions by factors of 50 to 100 with air/steam ratios of 0.1 to 0.4 by weight. The percentage removed decreased as the proportion of air to steam increased, but was independent of the total air volume. They hypothesized one of the controlling factors to be particle growth due to condensation. Rumary^[60] further demonstrated that the air/steam ratio and the depth of lute immersion were the only variables on which removal depended, the effects of lute diameter, flow rate, and temperature being insignificant. For pure air flowing through the lute, 60% to 80% of the particles were not removed. Hillary, Taylor, Abbey and Diffey^[61] had results which conflicted with the above in that their largest decontamination (> 500) was found with the largest air/steam ratio (75%). This was explained as being due to the use of cold air, thus causing the condensation on the particles to be proportional to the quantity of air through its capacity for cooling.

PARTICLE REMOVAL IN CONTAINMENT VESSELS

Projects at Battelle-Northwest (BNW) and at Oak Ridge National Laboratory (ORNL) have been designed to simulate as closely as practical a loss-of-coolant accident. Due to the complexity of the phenomena produced, however, little can be said about the exact nature of the removal processes or the degree to which the condensing steam environment contributes to particle demobilization. Limited theoretical expressions have been formulated to predict removal rates measured in these experiments.

The Nuclear Safety Pilot Plant (NSPP) represents the main effort at ORNL to measure fission product removal in a simulated containment vessel. The results of six runs in steam-air atmospheres have been reported.^[62-64] In these runs an initial steam-air atmosphere and charge of radioactive aerosol are allowed to decay. Early runs gave data primarily on iodine transport, the latest reported run giving the best data on the removal of particles.

Watson, Perez, and Fontana^[65] were able to predict satisfactorily the removal of molecular iodine in the first of the above mentioned runs with their "Condensing Steam Model". This postulates that the iodine is carried to the wall chiefly by the flux of condensing steam and hence would have the same removal rate as the steam. (Their analyses also included the idea of using

forced convection as a model for natural convection. The free-stream velocity to be used in the forced convection analysis is set equal to the local maximum velocity due to natural convection.) An analysis which parallels the work of Sparrow and Lin^[4] was also made for the removal of molecular iodine in NSPP, but calculations were not made for values of the governing parameters which corresponded with those of the experimental runs.^[66] The results from the last reported run^[64] for particle removal were in reasonable agreement with the correlation reported by Goldsmith and Stinchcombe^[50], equation (53).

The Containment Systems Experiment (CSE) at BNW has thus far reported two runs in their containment vessel which have simulated natural removal in a steam-air atmosphere.^[67] An analysis similar to that described earlier in the report under Empirical Similarity Solution was used and predicted well the removal rate of molecular iodine.^[68] Previous runs in much smaller tanks showed that the analysis of Watson, Perez, and Fontana could be greatly improved by including a term which accounted for the contribution of diffusion to deposition.^[69] This term empirically correlated the diffusion to the convective velocity through the square root of the condensation rate.

From the research reported it is apparent that steam condensation plays an important role in the removal of contaminants. However, it is also apparent that there is little more than an order of magnitude quantitative understanding of the phenomena occurring. Precise studies of these mechanisms need to be carried out in order to explain the measurements being made in these simulation experiments and to extrapolate them to actual conditions in a loss-of-coolant accident.

ACKNOWLEDGEMENTS

I would like to thank A. K. Postma for his assistance through the preliminary literature search which he carried out on this subject, and also L.C. Schwendiman and Mr. Postma for their helpful criticism of this review.

NOMENCLATURE

a	= accommodation coefficient for momentum
b	= accommodation coefficient for energy
C_m	= constant associated with the velocity slip
C_p	= specific heat capacity (per unit mass)
C_t	= constant associated with the temperature jump
d	= molecule diameter
D_{12}	= mutual diffusion coefficient for species 1 and 2
$\vec{e}$	= unit vector
$\vec{F}$	= force
$\vec{g}$	= gravitational acceleration
G	= free energy
k	= Boltzmann constant
ℓ	= mean free path
L	= mobility
M	= gram molecular weight
m	= molecular mass
$\vec{m}''$	= mass flux of vapor
$\vec{N}$	= number flux
n	= molecular number density
N_{Gr}	= Grashof Number = $g x^3 (M_\infty T_i - M_i T_\infty) / M_i T_\infty \nu^2$
N_{Kn}	= Knudsen Number = ℓ / R
N_{Nu}	= Nusselt Number = $q'' x / \lambda (T_\infty - T_i)$
N_{Pr}	= Prandtl Number = ν / λ
N_{Re}	= Reynolds number = $2RV / \nu$
N_{Sch}	= Schmidt Number = ν / D
N_{Sh}	= Sherwood Number = $m'' x kT / D_{12} m_1 [(p_1)_\infty - (p_1)_i]$
p	= fluid pressure
q''	= heat flux
R^*	= critical radius
R	= particle radius, drop radius, radius of curvature
S	= saturation ratio
t	= time
T	= temperature
$\vec{u}$	= fluid velocity
$\vec{v}$	= particle velocity

- w_A = mass fraction, ρ_A/ρ
- x = distance down vertical surface
- γ = number fraction, n_i/n
- δ = thickness of condensate film
- ϵ = eddy viscosity
- λ = thermal conductivity
- λ_t = thermal conductivity of monatomic gas
- μ = dynamic viscosity
- ν = kinematic viscosity
- ξ = distance down vertical surface
- ρ = mass density
- σ = surface tension
- σ_{12} = constant associated with diffusion slip
- τ = relaxation time
- $\vec{\tau}$ = stress tensor
- ϕ = contact angle
- ∇ = gradient operator = $\vec{e}_1 \frac{\partial}{\partial x_1} + \vec{e}_2 \frac{\partial}{\partial x_2} + \vec{e}_3 \frac{\partial}{\partial x_3}$
- Λ = latent heat of condensaton (per unit mass)

SUBSCRIPTS

- f = film
- i = liquid/gas interface
- i, j, k = species of gas mixture
- L = liquid
- p = particle
- w = wall
- ∞ = bulk of fluid far from boundaries
- 1 = water vapor
- 2 = air

REFERENCES

- [1] Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1966, ORNL-4071, Oak Ridge National Laboratory, Oak Ridge, Tenn., March, 1967.
- [2] R.B. Bird, W.E. Stewart and E.N. Lightfoot. Transport Phenomena, John Wiley & Sons, Inc., New York, New York, 1960, pp 75, 79, 557, 316.
- [3] W. Nusselt. Zeitschrift Vereines Deutscher Ingenieur, Vol. 60, pp 541-546, 569-575, (1916).
- [4] E.M. Sparrow and S.H. Lin. "Condensation Heat Transfer in the Presence of a Noncondensable Gas," J. Heat Transfer, Trans. ASME, Vol. 86, Series C, pp 430-436, (1964).
- [5] J.C.Y. Koh, E.M. Sparrow and J.P. Hartnett. "The Two Phase Boundary Layer in Laminar Film Condensation," Intern. J. Heat Mass Transfer, Vol. 2, pp 69-82, (1961).
- [6] M.M. Chen. "An Analytical Study of Laminar Film Condensation: Part I -- Flat Plates," J. Heat Transfer, Trans. ASME, Vol. 83, Series C, pp 48-54, (1961).
- [7] W.J. Minkowycz and E.M. Sparrow. "Condensation Heat Transfer in the Presence of Noncondensables, Interfacial Resistance, Superheating, Variable Properties and Diffusion," Intern. J. Heat Mass Transfer, Vol. 9, pp 1125-1144, (1966).
- [8] M. Jakob. Heat Transfer, Vol. I, John Wiley & Sons, New York, New York, Chapters 28 and 30, (1949).
- [9] W.H. McAdams. Heat Transmission, McGraw-Hill Book Co., Inc., New York New York, 1954, 3rd Ed., p 172-173.
- [10] See pp 636-676 of Reference [2].
- [11] L. Waldmann and K.H. Schmitt. "Thermophoresis and Diffusiophoresis of Aerosols," in Aerosol Science, (C.N. Davies, Editor), Academic Press, Inc., New York, New York, pp 137-162, (1966).
- [12] S. Chapman and T.G. Cowling. The Mathematical Theory of Nonuniform Gases, Cambridge Univeristy Press, Cambridge, Mass., 1953, 2nd Ed.
- [13] L. Waldmann. "Uber die Kraft eines Inhomogenen Gases auf Kleine Suspendierte Kugeln," Z. Naturforsch., Vol. 14a, pp 589-599, (1959).
- [14] S.P. Bakanov and B.V. Derjaguin. "The Motion of a Small Particle in a Nonuniform Gas Mixture," Discussions Far. Soc., Vol. 30, pp 130-138, (1960).
- [15] P.S. Epstein. "On the Resistance Experienced by Spheres in their Motion Through Gases," Phys. Rev., Vol. 23, pp 710-733, (1924).
- [16] P. Goldsmith, H.J. Delafield and L.C. Cox. "The Role of Diffusiophoresis in the Scavenging of Radioactive Particles from the Atmosphere," Quart. J. Roy. Meteorol. Soc., Vol. 89, pp 43-61, (1963) (also see Ref. 19).

- [17] K.H. Schmitt and L. Waldmann. "Untersuchungen an Schwebstoffteilchen im Diffundierenden Gasen," Z. Naturforsch., Vol. 15a, pp 843-851, (1960).
- [18] K.H. Schmitt. "Untersuchungen an Schwebstoffteilchen im Temperaturfeld," Z. Naturforsch., Vol. 14a, pp 870-881, (1959).
- [19] P. Goldsmith and F.G. May. "Diffusiophoresis and Thermophoresis in Water Vapor Systems," in Aerosol Science, (C.N. Davies, Ed.), Academic Press, Inc., New York, New York, pp 163-194, (1966).
- [20] J.R. Brock. "The Thermal Force in the Transition Region," J. Colloid Interface Sci., Vol. 23, pp 448-452, (1967).
- [21] P.L. Bhatnagar, E.P. Gross and M. Krook. "A Model for Collision Processes in Gases. I. Small Amplitude Processes in Charged and Neutral One-Component Systems," Phys. Rev., Vol. 94, pp 511-525, (1954).
- [22] J.R. Brock. "Experiment and Theory for the Thermal Force in the Transition Region," J. Colloid Interface Sci., Vol. 25, pp 392-395, (1967).
- [23] S. Jacobsen and J.R. Brock. "The Thermal Force on Spherical Sodium Chloride Aerosols," J. Colloid Sci., Vol. 20, pp 544-554, (1965).
- [24] P. Epstein. "Zur Theorie des Radiometers," Z. Physik., Vol. 54, p. 537, (1929).
- [25] J.R. Brock. "On the Theory of Thermal Forces Acting on Aerosol Particles," J. Colloid Sci., Vol. 17, pp 768-780, (1962).
- [26] R. Millikan, Phys. Rev., Vol. 22, p. 1, (1923).
- [27] B.V. Derjaguin and S.P. Bakanov. Dokl. Akad. Nauk SSSR. (Phys. Chem.) Vol. 147, p. 139, (1962).
- [28] B.V. Derjaguin and Y. Yalamov. "Theory of Thermophoresis of Large Aerosol Particles," J. Colloid Sci., Vol. 20, pp 555-570, (1965) [See, also, Vol. 21, pp 256-258, (1965).]
- [29] C.F. Schadt and R.D. Cadle. "Thermal Forces on Aerosol Particles." J. Phys. Chem., Vol. 65, pp 1689-1694, (1961).
- [30] B.V. Derjaguin, A.I. Storozhilova and Y. I. Rabinovich. "Experimental Verification of the Theory of Thermophoresis of Aerosol Particles," J. Colloid Interface Sci., Vol. 21, pp 35-58, (1966).
- [31] H.A. Kramers and J. Kistemaker. Physica, s' Grav., Vol. 10, p. 699, (1943).
- [32] J.R. Brock. "Forces on Aerosols in Gas Mixtures," J. Colloid Sci., Vol. 18, pp 489-501, (1963).
- [33] B.V. Derjaguin, Y.I. Yalamov and R.J. Storozhilova. "Diffusiophoresis of Large Aerosol Particles," J. Colloid Interface Sci., Vol. 22, pp 117-125, (1966).
- [34] F.F. Abraham. "A Re-Examination of Homogeneous Nucleation Theory: Thermodynamic Aspects," J. Atmospheric Sci., Vol. 25, pp 47-53, (1968).

- [35] J.E. McDonald, "Homogeneous Nucleation of Vapor Condensation. II. Kinetic Aspects," Am. J. Phys., Vol. 31, pp 31-41, (1963).
- [36] N.H. Fletcher. "Size Effect in Heterogeneous Nucleation," J. Chem. Phys., Vol. 29, pp 572-576, (1958). (See, also, Vol. 31, p. 1136).
- [37] N.H. Fletcher. The Physics of Rain Clouds, Cambridge University Press, London, pp 58-61, (1962).
- [38] See p. 60 of Reference [36]. Originally from W.E. Howell, "The Growth of Cloud Drops in Uniformly Cooled Air," J. Meteorology, Vol. 6, p. 134, (1949).
- [39] See pp 122-127 of Reference [37].
- [40] A. Einstein, Ann. Physik, Vol. 17, p. 549, (1905).
- [41] N.A. Fuchs. The Mechanics of Aerosols, Pergamon Press, New York, New York, p. 184, (1964).
- [42] See P. 71 of Reference [41].
- [43] A.C. Wells and A.C. Chamberlain. "Transport of Small Particles to Vertical Surfaces," Brit. J. Appl. Phys., Vol. 18, pp 971-977, (1967).
- [44] S.K. Friedlander and H.F. Johnstone. "Deposition of Suspended Particles From Turbulent Gas Streams," Ind. Eng. Chem., Vol. 49, pp 1151-1156, (1957).
- [45] P.R. Owen. "Dust Deposition from a Turbulent Airstream," Intern. J. Air-Water Pollution, Vol. 3, pp 8-25, 50-53, (1960).
- [46] C.N. Davies. Aerosol Science, Academic Press, Inc., New York, New York, pp 425-440, (1966).
- [47] G.A. Sehmel. Aerosol Deposition from Turbulent Airstreams in Vertical Conduits, BNWL-578, Pacific Northwest Laboratory, Richland, Washington, March, 1968.
- [48] J. Laufer. "The Structure of Turbulence in Fully Developed Pipe Flow," National Advisory Committee for Aeronautics Research, Report 1174, National Bureau of Standards, (1954).
- [49] C.S. Lin, R.W. Moulton and G.L. Putnam. "Mass Transfer Between Solid Walls and Fluid Streams," Ind. Eng. Chem., Vol. 45, pp 636-640, (1953).
- [50] P. Goldsmith and R.A. Stinchcombe. The Clean-Up of Submicron Particles By Condensing Steam, AERE-M-1214. Atomic Energy Research Establishment, Harwell, Great Britain, May, 1963 (See also Reference [19].)
- [51] G.I. Stuber. Detailed Project Status Report, Decontamination of Waste Gases, KLX-55. The Kellogg Corporation, New York, New York, (1949).
- [52] P.J. Schauer. Removal of Submicron Aerosol Particles from a Moving Gas Stream by the Condensation Center Effect, MLM-490, Mound Laboratory, Miamisburg, Ohio, 1950.
- [53] W.A. Bain. Project Summary Gas Decontamination, KLX-1380, The Kellogg Corporation, New York, New York, (1952).

- [54] F. Fahnoe, A.E. Lindross, and R.J. Abelson. "Aerosol Build-up Techniques," Ind. Eng. Chem., Vol. 43, pp 1336-1346, (1951).
- [55] A.L. Bralove. "Radioactive Dust Separation Equipment," Nucleonics, Vol. 8, pp 37-51, 60-67, (1951).
- [56] Rama. "Condensation Process for the Extraction of Radioactivity from Air," Nature, Vol. 184, pp 1789-1790, (1959).
- [57] K.T. Semrau, C.W. Marynowski, K.E. Lunde, and C.E. Lapple. "Influence of Power Input on Efficiency of Dust Scrubbers," Ind. Eng. Chem., Vol. 50, pp 1615-1620, (1958).
- [58] C.P. Ashworth, D.B. Barton and C.H. Robbins. "Pressure Suppression," Nucl. Eng., Vol. 7, pp 313-321, (1962).
- [59] H.R. Diffey, C.H. Rumary, M.J.S. Smith, and R.R. Stinchcombe. Iodine Clean-up in a Steam Suppression System, AERE-R-4882, Atomic Energy Research Establishment, Harwell, Great Britain, (1965).
- [60] C.H. Rumary. Decontamination Factors for a Submicron Ni/Cr Aerosol, AERE-PR/CE 10, pp 11-12, Atomic Energy Research Establishment, Harwell, Great Britain, (1966).
- [61] J.J. Hillary, J.C. Taylor, F. Abbey and H.R. Diffey. Iodine Removal By A Scale Model of the S.G.H.W. Reactor Vented Steam Suppression System, TRG Report 1256(W). United Kingdom Atomic Energy Authority, Risley, Great Britain, (1966).
- [62] L.F. Parsly and T.H. Row. Study of Fission Products Released from Trace-Irradiated UO₂ into Steam-Air Atmospheres (Nuclear Safety Pilot Plant Runs 8 and 9), ORNL-TM-1588, Oak Ridge National Laboratory, May 1966.
- [63] L.F. Parsly and T.H. Row. Behavior of Fission Products Released from Synthetic High-Burnup UO₂ in Steam Atmospheres (Nuclear Safety Pilot Plant Runs 10-12), ORNL-TM-1698, Oak Ridge National Laboratory, Feb. 1967.
- [64] L.F. Parsly and T.H. Row. Behavior of Fission Products Released into a Steam-Air Atmosphere from Overheated UO₂ Previously Irradiated to 20,000 MWD/T (Nuclear Safety Pilot Plant Run No. 14, Part I), ORNL-TM-1908, Oak Ridge National Laboratory, September, 1967.
- [65] G.M. Watson, R.B. Perez, and M.H. Fontana. Effects of Containment System Size on Fission Product Behavior, ORNL-4033, Oak Ridge National Laboratory, January, 1967.
- [66] N. Ozisik and D. Hughes, "Boundary Layer Analysis of the Effect of Condensation on Fission Product Deposition," Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1967, ORNL-4228, pp 90-92, Oak Ridge National Laboratory, April, 1968.
- [67] R.K. Hilliard, L.F. Coleman, and J.D. McCormack. "Large Scale Aerosol Transport Test," Nuclear Safety Programs Quarterly Report, July-October 1967, BNWL-754, pp 1-36, Pacific Northwest Laboratory, Richland, Washington, (to be published).

- [68] R.K. Hilliard and J.G. Knudsen. "Mass Transfer Model for Natural Transport," Nuclear Safety Programs Quarterly Report, November 1967 - January 1968, pp 27-33, Pacific Northwest Laboratory, Richland, Washington (to be published).

- [69] R.K. Hillard, L.F. Coleman, and J.D. McCormack. Comparisons of the Containment Behavior of a Simulant with Fission Products Released from Irradiated UO_2 , BNWL-581, Pacific Northwest Laboratory, Richland, Washington, March, 1968.

DISTRIBUTION

No. Of
Copies

- 1 AEC, Chicago Operations Office
 9800 South Cass Avenue
 Argonne, Illinois 60439
 G.H. Lee, Director, Patent Division
- 2 AEC, Chicago Patent Group
 Federal Bldg., 700 Area
 Richland, Washington 99352
 R.K. Sharp
- 13 AEC Library, Washington, D.C. 20545
 Division of Reactor Development & Technology
 A.J. Pressesky, Chief, Analysis & Evaluation Board (5)
 J.A. Lieberman, Assistant Director, Nuclear Safety (5)
 H.L. Hamester
 I.C. Roberts
 S.A. Szawlewicz
- 267 AEC, Division of Technical Information Extension
 P.O. Box E
 Oak Ridge, Tennessee
- 1 AEC RDT Senior Site
 Representative
 P.O. Box 550, Area 300, Bldg. 3760, Richland, Wash. 99352
 P.G. Holsted/A.D. Toth
- 2 AEC Richland Operations Office
 Federal Bldg., Richland, Washington 99352

 C.L. Robinson, Director, Research & Development Division
 Technical Library
- 3 USAEC, Scientific Representative
 c/o Atomic Energy of Canada, Ltd.
 Chalk River, Ontario, Canada

 R.W. Ramsey
- USAEC/AECL COOPERATIVE PROGRAM
 TAC AND SUBCOMMITTEES
- 5 Technical Advisory Committee (TAC) - United States

 A.N. Tardiff, U.S. Chairman of TAC, AEC Headquarters, Washington, D.C.
 H.J. Reynolds, U.S. Secretary of TAC, c/o Atomic Energy 20545
 of Canada, Ltd., Sheridan Park, Ontario, Canada

 D.F. Babcock, U.S. Member of TAC, E.I. duPont de Nemours & Co.,
 Wilmington, Delaware

H. Harty, U.S. Member of TAC, Heavy Water Reactor Program Office,
Battelle-Northwest, Richland, Washington 99352

E.A. Mason, U.S. Member of TAC, Massachusetts Institute of Technology
Cambridge, Massachusetts, 02139

5 Technical Advisory Committee (TAC) - Canadian

D.D. Stewart, Canadian Chairman of TAC, Atomic Energy of Canada, Ltd.,
Chalk River Nuclear Laboratories, Chalk River, Ontario, Canada

W.R. Livingston, Canadian Secretary of TAC, Atomic Energy of Canada, Ltd.,
Chalk River Nuclear Laboratories, Chalk River, Ontario, Canada

L. Pease, Canadian Member of TAC, Atomic Energy of Canada, Ltd.,
Toronto, Ontario, Canada

R.F.S. Robertson, Canadian Member of TAC, Atomic Energy of Canada, Ltd.,
Whiteshell Nuclear Research Establishment, Pinawa, Manitoba, Canada

W.R. Thomas, Canadian Member of TAC, Atomic Energy of Canada, Ltd.,
Chalk River Nuclear Laboratories, Chalk River, Ontario, Canada

9 TAC Subcommittee - SAFETY

A.J. Pressesky, Analysis & Evaluation Branch, Division of Reactor
Development & Technology, U.S. Atomic Energy Commission,
Washington, D.C. 20545

G. Hake, Atomic Energy of Canada, Ltd., Chalk River Nuclear Laboratory,
Chalk River, Ontario, Canada

H. Morewitz, Atomics International, P.O. Box 309,
Canoga Park, California 91305

R.K. Hilliard, Battelle-Northwest, Richland, Washington 99352

F.C. Boyd, Atomic Energy Control Board, Chalk River, Ontario, Canada

S. Davies, Canadian General Electric Co., Ltd., 107 Park Street N.,
Petersborough, Ontario, Canada

Manager, Water Reactor Safety Program Office, Phillips Petroleum Co.,
Idaho Falls, Idaho 83401

J.J. Billington, Engineering & Physics, Dilworth, Secord, Meagher &
Associates, 4195 Dundas St., West, Toronto, Ontario, Canada

J.D. Sainsbury, Atomic Energy of Canada, Ltd., Sheridan Park,
Ontario Canada

1 Atlantic Richfield Hanford Co., Richland, Washington

P.W. Smith

- 4 Atomic Energy Research Establishment, Harwell, England
 A.E.J. Eggleton
 W.J. Megaw
 P. Goldsmith
 R.A. Stinchcombe
- 1 Atomics International, Canoga Park, California
 R. Kountz
- 1 Babcock & Wilcox Co., Lynchburg, Virginia
 D.A. Nitti
- 3 Battelle Memorial Institute, Columbus, Ohio
 J.M. Genco
 J.A. Gieske
 D.L. Morrison
- 1 Brookhaven National Laboratory
 A.W. Castleman, Jr.
- 1 California Institute of Technology
 S.K. Friedlander
- 4 Douglas-United Nuclear, Richland, Washington
 T.W. Ambrose
 N.R. Miller
 J.W. Riches
 J.R. Spink
- 1 duPont, Savannah River Laboratory, Aiken S. Carolina
 A.H. Peters
- 2 General Atomics, San Diego, California
 D. Busch
 H.H. Van den Bergh
- 2 General Electric Co., APED, San Jose, California
 H.V. Clukey
 M. Siegler
- 1 Georgia Institute of Technology
 C. Orr
- 1 Harvard University
 F.J. Viles, Jr.
- 1 National Center for Atmospheric Research
 R.D. Cadle

1 North Carolina State University

M.N. Ozisik

9 Oak Ridge National Laboratory, Oak Ridge, Tennessee

R.E. Adams
W.E. Browning, Jr.
W.B. Cottrell
M.H. Fontana
G.N. Keilholtz
L.F. Parsely, Jr.
G.W. Parker
T.H. Row
G.M. Watson

1 Oregon State University

J.G. Knudsen

1 Reactor Development Laboratory, Windscale, England

D.A. Collins

2 University of Minnesota

K.T. Whitby
B.Y. Liu

1 University of Pittsburgh

M. Corn

1 University of Texas

J.R. Brock

2 Westinghouse Electric Co., (APD)

W.D. Fletcher
J.D. McAdoo

Battelle-Northwest, Richland, Washington

J.M. Batch
G.E. Benedict
D.W. Bolme
M.T. Dana
C.E. Elderkin
J.J. Fuquay
J.M. Hales
T.W. Horst (20)
R.L. Junkins
J.D. McCormack
H.M. Parker
A.K. Postma (10)
G.J. Rogers
L.C. Schwendiman (50)
C.L. Simpson
G.A. Sehmel

W.G.N. Slinn
E.C. Watson
K.R. Wise
N.G. Wittenbrock
M.A. Wolf
Technical Information Files(5)
Technical Publications