CONDENSATION HEAT TRANSFER: COMMENTS ON NON-EQUILIBRIUM TEMPERATURE PROFILES AND THE ENGINEERING CALCULATION OF MASS TRANSFER

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Abstract—Two condensation heat transfer topics of relevance to nuclear reactor safety are critically reviewed. (1) The theory of non-equilibrium phase change developed by Bornhorst and Hatsopoulos is examined, and an anomaly in the vapor phase temperature profile noted. The source of the anomaly is clarified, and it is suggested that the theory is of doubtful validity. (2) The advisability of using simple engineering methods to calculate mass transfer phenomena during condensation from steam-air mixtures is discussed. A review of a suitable method is presented, with emphasis on the calculation of condensation rate, aerosol particle deposition rate and absorption of soluble gaseous species.

1. NON-EQUILIBRIUM VAPOR TEMPERATURE PROFILES

Bornhorst & Hatsopoulos (1967a) analyzed non-equilibrium liquid-vapor phase change using the methodology of irreversible thermodynamics, and concluded that during the condensation of saturated vapor the usual assumption of a uniform temperature profile is unrealistic. A dimensionless parameter $\Gamma = \phi(1 - T_s/T_i)$ was defined to characterize condensation rate, where $\phi = 2[1 - (P_s/P_i)(T_i/T_s)^{1/2}]$, P and T are pressure and temperature respectively; subscript *i* refers to vapor at the edge of the Knudsen layer, while T_s is the liquid temperature at the interface and P_s is the corresponding saturation pressure. Data was presented for the temperature difference $(T_i - T_{\infty})$, where T_{∞} is the vapor temperature some distance away from the interface, and is reproduced in table 1. Positive values of $(T_i - T_{\infty})$ imply the temperature profile shown in figure 1.

Such temperature profiles, as well as the large values of the temperature jump $(T_i - T_s)$ implied in table 1 are not in accord with physical intuition. Nevertheless, in a subsequent paper Bornhorst & Hatsopoulos (1967b) used these results in an analysis of bubble growth in various liquids, and concluded that non-equilibrium effects are very important at low pressure. Theofanous *et al.* (1969) also subsequently calculated bubble growth accounting for non-equilibrium effects, and claim agreement with experimental data for bubble growth in water and nitrogen. More recently the theory of Bornhorst and Hatsopoulos has been used by Baker (1978) to calculate sublimation of graphite subjected to intense radiation.

The purpose here is to show how the anomalies described above arise, and to question the validity of the underlying postulates. The nature of the dilemma can be most easily shown using



Figure 1. Non-equilibrium vapor phase temperature profile for condensation according to Bornhorst & Hatspoulos (1967a).

the approach of Schrage (1953) which is essentially an extension of the slip flow analysis of rarefied gas dynamics theory. Two streams of molecules are considered, one incident on the liquid surface and one leaving the liquid surface. A Knudsen layer adjacent to the interface is assumed wherein the effects of molecular collisions are ignored. The suffix i denotes conditions at the outer edge of the Knudsen layer, i.e. of the incident stream. The condensation and thermal accommodation coefficients will be assumed equal to unity. Then the incident flux is

$$\frac{n_i c_i}{4} [e^{-s^2} + \sqrt{\pi s (1 + \operatorname{erf} s)}], \qquad [1]$$

while invoking the Hertz hypothesis, the emitted flux is an equilibrium Maxwellian stream,

$$\frac{n_s c_s}{4}$$
, [2]

where *n* is number density, *c* is the average molecular speed $\left(\frac{8}{\pi}RT\right)^{1/2}$, $s = u/(2RT)^{1/2}$, and *u* is the vapor velocity normal to the interface. The net rate of condensation is

$$\dot{m}'' = \frac{n_i c_i}{4} [e^{-s^2} + \sqrt{\pi s (1 + \text{erf } s)}] - \frac{n_s c_s}{4}, \qquad [3]$$

$$=\frac{n_{i}c_{i}}{4}(1+\sqrt{\pi s})-\frac{n_{s}c_{s}}{4} \text{ for } \dot{m}''\to 0 \quad (s\to 0), \qquad [4]$$

$$=\frac{n_i c_i}{4} + \frac{\dot{m}''}{2} - \frac{n_s c_s}{4},$$
 [5]

$$\dot{m}'' = 2\left[\frac{n_i c_i}{4} - \frac{n_s c_s}{4}\right] \text{ molecules/m}^2 \text{ s,}$$
[6]

or

$$\dot{m}'' = 2 \left[\frac{P_i}{(2\pi R T_i)^{1/2}} - \frac{P_s}{(2\pi R T_s)^{1/2}} \right] \, \text{kg/m}^2 \, \text{s} \,, \tag{7}$$

$$= 2[w^+ - w^-].$$
 [8]

Notice that the emitted flux does not contribute an additional $\dot{m}''/2$ since it was postulated to be an equilibrium Maxwellian stream: thus a number density "jump" results in order to have the r.h.s. of [5] equal to \dot{m}'' , in complete analogy with the temperature jump of slip flow.

For a monatomic vapor the incident energy flux is

$$E^{+} = w^{+}(2RT_{i}) + \frac{\dot{m}''}{2} \left(\frac{5}{2}RT_{i}\right) - \frac{1}{2}k \frac{\mathrm{d}T}{\mathrm{d}x}, \qquad [9]$$

where R is the gas constant/unit mass and k the thermal conductivity, and may be derived from Grad's thirteen moment distribution (upon dropping higher order and coupled terms). Alternatively, the first two terms come directly from integrating

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{0}^{\infty}f\xi_{x}\left(\frac{1}{2}m\xi^{2}\right)\mathrm{d}\xi,$$

with

$$f = \frac{n}{(2\pi RT)^{3/2}} \exp\left(-\frac{(\xi - \underline{u})^2}{2RT}\right)$$

and the third term may be simply added, following say Kennard (1938), who argued that it is the excess energy carried as the incident stream's contribution to heat conduction. The emitted energy flux is

$$E^{-} = w^{-}(2RT_s),$$
 [10]

so that the net energy transport across the Knudsen layer is

$$q = E^{+} - E^{-} = w^{+}(2RT_{i}) - w^{-}(2RT_{s}) + \frac{\dot{m}''}{2} \left(\frac{5}{2}RT_{i}\right) - \frac{1}{2}k\frac{\mathrm{d}T}{\mathrm{d}x}.$$
 [11]

In order to obtain the temperature "jump" $(T_i - T_s)$ this energy flux must be equated to the continuum expression for the energy flux at the edge of the Knudsen layer, which is

$$q = \dot{m}'' h_i - k \frac{\mathrm{d}T}{\mathrm{d}x} = \dot{m}'' \left(\frac{5}{2} R T_i\right) - k \frac{\mathrm{d}T}{\mathrm{d}x} \,. \tag{12}$$

Equating [11] and [12] and rearranging gives

$$-\frac{1}{2}k\frac{\mathrm{d}T}{\mathrm{d}x} = \left(w^{+} - \frac{\dot{m}''}{2}\right)(2R)(T_{i} - T_{s}) - \frac{\dot{m}''}{2}\left(\frac{1}{2}RT_{i}\right).$$
 [13]

The term $\left(w^{+} - \frac{\dot{m}''}{2}\right)$ is the average of w^{+} and w^{-} and could perhaps be set equal to $\mu/2\lambda$ where the bar indicates an average value over the Knudsen layer, and λ is the mean free path. Alternately we can assume $\dot{m}''/2 \ll w^{+}$ and thereby obtain the result of Bornhorst & Hatsopoulos (1967a), who used the methodology of irreversible thermodynamics to combine Schrage's expression for \dot{m}'' , with Kennard's temperature jump expression for $\dot{m}'' = 0$. Solving [13] for the temperature jump gives

$$T_{i} - T_{s} = \frac{-k(dT/dx)}{2\left(w^{+} - \frac{\dot{m}''}{2}\right)(2R)} + \frac{\dot{m}''T_{i}}{8\left(w^{+} - \frac{\dot{m}''}{2}\right)}.$$
[14]

When $\dot{m}'' = 0$, Kennard's temperature jump expression is recovered.

Finally we consider a one-dimensional situation with the vapor at infinity at T_{∞} : solving the appropriate form of the energy conservation equation gives

$$-k\frac{\mathrm{d}T}{\mathrm{d}x}=\dot{m}''C_p(T_x-T_i). \tag{15}$$

For a polyatomic vapor we replace (2R) in [14] by $C_v + (1/2)R$, and taking $w^+ - (\dot{m}''/2) \simeq w^+$, we equate [12] and [15] to obtain

$$\frac{1}{2}\dot{m}''C_{p}(T_{\infty}-T_{i})=w^{+}\left(C_{v}+\frac{1}{2}R\right)(T_{i}-T_{s})-\frac{\dot{m}''}{2}\left(\frac{1}{2}RT_{i}\right),$$
[16]

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or

$$\frac{T_i - T_{\infty}}{T_i} = \frac{R}{2C_p} - \frac{2w^+}{\dot{m}''} \left(\frac{C_v + \frac{1}{2}R}{C_p}\right) \left(\frac{T_i - T_s}{T_i}\right).$$
[17]

Substituting $C_p - C_v = R$, and $C_p/C_v = \gamma$, and defining $\phi = \dot{m}''/w^+$, gives

$$T_{i} = \frac{T_{\infty}}{1 - \frac{\gamma - 1}{2\gamma} + \frac{\gamma + 1}{\gamma} \frac{1}{\phi}} + \frac{T_{s}}{1 + \frac{\gamma}{\gamma + 1}\phi - \frac{\gamma - 1}{2(\gamma + 1)}\phi},$$
[18]

and [7] may be rewritten

$$\boldsymbol{\phi} = 2 \left[1 - \frac{P_s}{P_i} \left(\frac{T_i}{T_s} \right)^{1/2} \right]$$
[19]

$$P_s = P_s(T_s)$$
 from equilibrium data. [20]

For prescribed P_i , T_x and ϕ , [18]–[20] can be solved for T_s , T_i and P_s . It is also of interest to calculate

$$T_{I}^{*} = \frac{T_{\infty}}{1 + \frac{\gamma + 1}{\gamma} \frac{1}{\phi}} + \frac{T_{s}}{1 + \frac{\gamma}{\gamma + 1} \phi}, \qquad [21]$$

which is the value of T_i obtained if the phase change term in [14] is ignored.

Table 2 shows some typical results for condensation of saturated steam. The main feature of the results is that T_i proves to be higher than T_{∞} . Associated with this anomaly is a larger than expected temperature jump at high rates of condensation. For example, in case 3, $(T_i - T_s)$ is 2.05 K, while $(T_i^* - T_s)$ is only 0.05 K (the usual temperature jump of slip flow theory). On the other hand the temperature jumps are small compared to those suggested by Bornhorst & Hatsopoulos (1967a); even in case 3 where the vapor velocity towards the interface is of the order of the sonic velocity, $(T_i - T_{\infty})/T_i$ is only 0.004. The presentation of results by Bornhorst and Hatsopoulos in terms of dimensionless parameters characterizing the condensation process is quite misleading.

There is no direct experimental evidence to confirm the temperature profiles suggested by Bornhorst and Hatsopoulos: such temperature profiles are simply too difficult to measure. Thus it is suggested that the postulates used in the analysis should be critically examined before accepting such anomalous results. First let us examine carefully how values of $T_i > T_x$ come about. In the absence of phase change [11] and [12] become

$$q = w^{+}(2RT_{i}) - w^{-}(2RT_{s}) - \frac{1}{2}k\frac{\mathrm{d}T}{\mathrm{d}x}, \qquad [11a]$$

$$q = -k\frac{\mathrm{d}T}{\mathrm{d}x}\,.\tag{12a}$$

							$T_{\rm c} - T$	
Case	<i>P_i</i> bar	φ	T∗ °C	\mathcal{C}^{T_i}	<i>T₅</i> °C	<i>T</i> * ℃	$\frac{T_i}{T_i}$	u m/s
1	0.035	0.001	26.79	26.80	26.78	26.78	0.0000	3.17
2	0.035	0.01	26.79	26.92	26.70	26.70	0.0004	31.7
3	0.035	0.1	26.79	27.92	25.87	25.92	0.004	325.0
4	1.0	0.001	100.05	100.06	100.03	100.03	0.0000	7.53
5	1.0	0.01	100.05	100.16	99.9 0	99.9 0	0.0001	75.3

Table 2. Vapor temperatures during saturated steam condensation

Equating these Knudsen layer and continuum expressions for q allows evaluation of $(T_i - T_s)$, the usual temperature jump or slip flow. $(T_i - T_s)$ is not zero because $\frac{1}{2}k(dT/dx)$ is missing from [11a] as a result of the emitted molecules having a postulated Maxwellian distribution. On the other hand the incident molecules are recognized to have a non-equilibrium distribution to the extent that they must carry excess energy as the incident stream's contribution to heat conduction. Now considering phase change an analogous situation exists: [11] is missing a $\frac{1}{2}(\dot{m}''_2RT_i)$ because the emitted molecules were postulated to be an equilibrium Maxwellian stream of zero bulk velocity. Thus in equating [11] and [12] the enthalpy flow $\dot{m}''(\frac{1}{2}RT_i)$ in the continuum expression for q cannot be balanced and the temperature difference $(T_i - T_s)$ is forced to become a function of \dot{m}'' . There is an effective heat source at the boundary of the Knudsen layer owing to the mismatch of enthalpy flow terms.

There is a serious inconsistency in the postulates leading to this anomaly. The energy flux in the Knudsen layer can only be written as $q = E^+ - E^-$ if E^+ and E^- are evaluated at the same plane. If we take the edge of the Knudsen layer to be about one mean free path from the interface then the evaluation of E^+ is suspect: if a temperature jump exists then a quasiequilibrium distribution at temperature T_i , one mean free path from the interface is impossible, since on an average about 1/2 the molecules there arrive with temperature T_s and 1/2 with a temperature a little lower than T_i . On the other hand if we take the edge of the Knudsen layer to be a few mean free paths from the interface it may well be a good assumption to evaluate E^+ based on a quasi-equilibrium distribution at temperature T_i , but then it is untenable to evaluate E^- at another plane, i.e. as a stream of molecules emitted from the surface. Again, if a temperature jump does exist, then the E^- stream will change substantially over a few mean free paths. As one further point, even the postulate of the emitted stream being in equilibrium with the liquid surface should be questioned since there is a substantial energy flux through the interface.

A more refined kinetic theory approach to the problem allowing for non-equilibrium distributions would possibly yield an acceptable vapor temperature profile. In fact there have been a number of attempts in this direction, e.g. Shankar (1968), Patton & Springer (1969), Sloat *et al.* (1971), and Cippolla, *et al.* (1974), but these have been mainly concerned with obtaining corrections to the mass flux expression, [3], and have not attempted to resolve anomalies in the temperature profile. Until such time the results of Bornhorst and Hatsopoulos are confirmed by such analysis it is suggested that they be disregarded.

2. CONDENSATION FROM STEAM-AIR MIXTURES

2.1 Introduction

Nuclear reactor containment-vessel design and accident analysis often requires calculations of heat and mass transfer rates for steam condensing from steam-air mixtures. For example, Hales *et al.* (1972) were required to calculate condensation rates on the walls of a containment vessel in order to predict radioactive aerosol-particle deposition rates. The gas phase heat and mass transfer problem was idealized as the laminar natural convection boundary layer associated with laminar film condensation on a vertical surface, and exact numerical solutions of a set of coupled non-linear ordinary differential equations obtained. The advisability of using such solutions for this purpose should be questioned since the engineering problem involves in general a non-stagnant steam-air mixture, various geometrical shapes, and a poorly wetting surface giving rise to mixed rather than film-wise condensation. But most importantly, the rate at which heat is removed from the condensate will depend on the thermal properties of the vessel wall and an appropriate thermal network must be set up and solved. In such a network numerical data from exact solutions are cumbersome to use, and simple correlations are more appropriate.

Similar comments can be made about the work of Chung & Ayyaswamy (1978a, b) who were concerned with condensation on droplets of a containment spray emergency core cooling

system, with associated deposition rates of radioactive aerosol particles, and absorption into the liquid phase of fission products such as elemental iodine and methyl iodide. The effect of liquid phase reagents added to increase absorption rates was also investigated. Numerical and perturbation solutions were obtained for the vapor phase boundary layer in the stagnation region of a translating rigid sphere. But the engineering problem requires calculation of condensation rates and absorption rates for the whole droplet, and the absorption problem may require simultaneous consideration of the liquid phase. Most importantly the droplet temperature changes rapidly over its trajectory so that the engineering problem requires solution of coupled ordinary differential equations so that auxiliary functions should preferably be in the form of simple engineering correlations. It should be further noted that in both of the above discussed studies, the numerical solution of the vapor phase transport problem required considerable effort and expense, and perhaps served to obscure features of the complete engineering problem.

It therefore seems appropriate to review existing methods for the engineering analysis of heat and mass transfer during condensation from steam-air mixtures. The desired level of analysis should be equivalent to using Newton's law of cooling, $q = h\Delta T$, for convective heat transfer, but must be more complex owing to the presence of simultaneous heat and mass transfer, and the necessity to properly account for finite (and high) mass transfer rates. Good chemical engineering texts, for example that of Bird *et al.* (1960), contain the necessary ingredients for such analysis, but are seldom combined into a form convenient for problem solving. On the other hand Spalding (1960, 1963; Kays 1966) has presented a standard formulation of the steady state convective heat and mass transfer problem which is particularly convenient for engineering problem solving. Spalding's approach will be followed here with a view of (i) demonstrating its use, and (ii) as a vehicle for presenting some correlations appropriate for mass transfer during steam condensation.

2.2 The simultaneous heat and mass transfer problem

First we will consider the calculation of condensation rates from steam-air mixtures: figure 2 depicts a steam-air gas phase adjacent to a water liquid phase. Irrespective of the nature of gas phase flow, i.e. whether it be laminar or turbulent, forced or natural, the steam condensation rate m'' can be calculated from

$$\dot{m}'' = g_m B_m = g_h B_h, \tag{22}$$

$$B_m = \frac{m_{1,e} - m_{1,s}}{m_{1,s} - 1}; \qquad B_h = \frac{C_{pe}(T_e - T_s)}{h_{fg} - (\dot{q}''_{u}/\dot{m}'')},$$
[23]

$$g_m = \frac{\rho D_{12}}{L} \operatorname{Nu}_m; \qquad g_h = \frac{k/C_p}{L} \operatorname{Nu}_h, \qquad [24]$$



Figure 2. Notation for condensation from a steam-air mixture.

$$\frac{\mathrm{Nu}}{\mathrm{Nu}^*} = F(B) \,, \tag{25}$$

$$Nu_m^* = f(Re, Sc); \quad Nu_h^* = f(Re, Pr) \quad (forced \ convection), \qquad [26]$$

$$Nu_m^* = f(Gr, Sc); \quad Nu_h^* = f(Gr, Sc, Pr) \quad (\text{free convection}).$$
[27]

In the above equations g is the conductance; B is the driving force; and subscripts m and h refer to based on mass or heat transfer respectively, m_1 is the mass fraction of steam with subscript s denoting the gas phase adjacent to the interface and e denoting the free stream or bulk condition. Properties include h_{fg} , the latent heat of condensation; ρ , k and C_p , the mixture density, thermal conductivity and specific heat, respectively; D_{12} is the binary diffusion coefficient for the steam-air mixture. Nu, Re, Gr, Sc and Pr are the dimensionless Nusselt, Reynolds, Grashof, Schmidt and Prandtl numbers, respectively. The superscript * refers to the limit of zero net mass transfer $(m'' \rightarrow 0)$.

The evaluation of the conductive heat flux across the *u*-surface into the water, \dot{q}''_{u} , depends on the particular problem. For example, if condensation on a condenser tube is under consideration, then $\dot{q}''_{u} = -U(T_s - T_c)$ where U is an overall heat transfer coefficient which accounts for the thermal resistances of the condensate film, tube wall, coolant flow, etc. and T_c is the coolant temperature; a sample calculation for such a problem is given by Edwards *et al.* (1979). In the case where condensation takes place on a containment vessel wall, \dot{q}''_{u} is the input to a thermal network calculation of the thermal response of the condensate layer and wall structure. For condensation on a droplet there are two possibilities. (i) Most likely a lumped thermal capacitance model of the droplet thermal response is adequate and \dot{q}''_{u} is used in a droplet energy balance to determine its time rate of change of bulk temperature. (ii) If liquid side transport is to be accounted for then the appropriate model equation, e.g. based on a Hill's vortex flow, can be solved once and for all, and a correlation of the interior heat transfer coefficient, h_i , developed. Then $\dot{q}''_{u} = -h_i(T_s - T_b)$ and the bulk droplet temperature T_b is calculated from a droplet energy balance as before.

The mass transfer driving force B_m is essentially exact. The heat transfer driving force B_h is exact only for unity Lewis number, or for equal specific heats of vapor and noncondensable; neither are quite true for steam-air mixtures. The formulation is easily modified to remove this restriction, Edwards *et al.* (1979), but such a refinement is of little value, since there are larger sources of error elsewhere. The mass and heat transfer Nusselt numbers in the limit $\dot{m}'' \rightarrow 0$, Nu^{*}_m and Nu^{*}_h are found from standard correlations for the particular flow situation and geometry.

The "blowing factor" F, which accounts for the effect of finite mass transfer rates on concentration and temperature profiles, can as a first approximation be taken as the simple logarithmic function of stagnant film or Couette flow model theory,

$$F = \frac{\ln\left(1+B\right)}{B}.$$
 [28]

However, more refined correlations of F are available for specific flow situations. Exact numerical solutions of boundary layer equations can be used to determine a weighting factor for B in [28] to conveniently account for variable properties: Mills & Wortman (1972) demonstrate such an approach. Exact numerical solutions for condensation from steam-air mixtures are given by Minkowycz & Sparrow (1966), Denny *et al.* (1971) and South (1972). Other useful exact and approximate solutions of the noncondensable gas problem are given by Sparrow & Lin (1964), Turner *et al.* (1973), and Rose (1969). When air is present in very small concentrations, condensation rates are very high and the vapor phase boundary layer is under A. F. MILLS

strong suction. Acrivos (1960) has developed useful asymptotic formulae for F in this limit, applicable to laminar boundary layers; for forced flow the formulae are of the form

$$F_m = C \frac{\mathrm{Sc}^n}{[2(1+B_m)(1+\mathrm{Sc}^{-1})]^{1/2}},$$
[29]

where the constants C and n are functions of geometry only. Equation [29] was used as the basis of correlation of experimental data for condensation from steam-air mixtures flowing down over a horizontal tube by Mills *et al.* (1974),

$$F_m = \left[1 + \left(\frac{-1.18B_m}{(1+B_m)^{1/2}}\right)^{3/2}\right]^{2/3}.$$
 [30]

When correlations such as [30] are developed from experimental or numerical data, secondary effects such as the effect of vapor drag on the condensate film may also be approximately accounted for.

A further simplification of [22]-[27] is often justifiable: to good approximation

$$\frac{g_m}{g_h} = Le^{2/3}; \quad Le = \Pr/Sc,$$
 [31]

and [27] becomes

$$\frac{\dot{m}''}{g_m} = \frac{m_{1,e} - m_{1,s}}{m_{1,s} - 1} = \mathrm{Le}^{-2/3} \frac{C_{pe}(T_e - T_s)}{h_{fg} - (\dot{q}''_w/\dot{m}'')}.$$
[32]

2.3 Deposition of radioactive aerosol particles

After the simultaneous heat and mass transfer problem is solved for the condensation rate \dot{m}'' the calculation of the aerosol particle deposition rate is relatively straightforward; with subscript p denoting an aerosol particle we can also write

$$\dot{m}'' = g_p B_p$$
; $B_p = \frac{m_{p,e} - m_{p,s}}{m_{p,s} - m_{p,t}}$, [33]

provided we are prepared to ignore the effects of thermophoresis and diffusiophoresis. Contrary to the impression given by Hales *et al.* (1972), such effects are small for the containment vessel steam-air condensation problem, as has been shown by Lau (1973). Essentially convection dominates except for very low particle Schmidt numbers, where Brownian motion is dominant; when phoretic effects are large compared to Brownian motion, convection controls the particle deposition rate. Again we have

$$Nu_{p}^{*} = f(Re, Sc_{p}) \qquad (forced convection),$$
$$= f(Gr, Sc, Sc_{p}) \qquad (free convection), \qquad [34]$$

where $Sc_p = \nu/D_p$ and D_p is the Brownian diffusion coefficient in the steam-air mixture. The simple logarithmic blowing factor, [28] should be particularly appropriate in this large Sc limit. The transferred state mass fraction $m_{p,t}$ is equal to $n_{p,s}/\dot{m}''$. It is usually assumed that all particles impinging on a condensed phase surface are captured, so that $m_{p,s} = 0$. Substituting in [33] and rearranging gives the particle flux

$$n_{p,s} = g_p m_{p,e}.$$
 [35]

Equation [35] can be rewritten in terms of number density and particle flux if desired.

Since aerosol particles have rather large Schmidt numbers, we are interested in correlations for $\operatorname{Nu}_{p}^{*}$ valid in the limit $\operatorname{Sc}_{p} \to \infty$. For forced convection such correlations are readily available. For free convection the appropriate limit forms are more complex, and the forms used by Hales *et al.* (1972) are incorrect. For example Lightfoot (1968) used an analysis of laminar free convection on a vertical wall to develop a correlation valid in the limit $\operatorname{Gr}_{m}/\operatorname{Gr}_{h} \to 0$, and $\operatorname{Pr/Sc} \to 0$,

$$Nu_m^* = 0.50 Gr_h Sc (Sc/Pr)^{1/12},$$
[37]

a result which has been subsequently confirmed numerically by Wassel & Mills (1975). Equation [37] suggests that the appropriate correlation of Nu_{ρ}^{*} for the same flow condition of steam-air mixtures is

$$Nu_{p}^{*} \simeq 0.55 Gr^{1/4} Sc_{p}^{-1/3}.$$
 [38]

2.4 Absorption of soluble gaseous species

Turning now to absorption into the liquid phase of soluble fission products such as elemental iodine and methyl iodine, we denote such species as species 3 and can write

$$\dot{m}'' = g_{m3}B_{m3} = g_{m3}\frac{m_{3,e} - m_{3,s}}{m_{3,s} - m_{3,t}},$$
[39]

where \dot{m}'' is the already calculated steam condensation rate. Effective binary diffusion of species 3 in the steam-air mixture has been assumed, and again care must be taken in the correct development of free convection correlations for g_{m3}^* . For most effective scrubbing when using a droplet spray it is desirable to add a suitable reagent to the injected water. For example, use of hydrazine to increase the solubility of methyl iodide has been suggested. Ideally the solubility should be so increased that gas side control with $m_{3,s} = 0$ is obtained. Then with $m_{3,t} = n_{3,s}/\dot{m}''$, [39] becomes

$$n_{3,s} = g_{m3}m_{3,e}.$$
 [40]

More generally $m_{3,s}$ will be related to $m_{3,u}$ through solubility data, perhaps in the form of Henry's law, and transport of species 3 in the liquid phase must be considered. As was the case for heat transport in section 2.2, interior mass transfer coefficient correlations can be developed once and for all and an overall mass transfer coefficient used to describe transport of the solute into the bulk liquid. An overall species balance on the droplet then gives the rate of change of bulk concentration of the solute.

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