MATERIAL REMOVAL ASSOCIATED WITH CONDENSATION ON A DROPLET IN MOTION

J. N. CHUNG[†] and P. S. AYYASWAMY University of Pennsylvania, Philadelphia, PA 19104, U.S.A.

(Received 27 February 1980; in revised form 29 September 1980)

Abstract—This paper treats a three-phase, multicomponent fluid mechanics-heat and mass transfer problem. Solutions to the nonlinear, coupled boundary layer equations that govern laminar condensation heat and mass transfer in the vicinity of the forward stagnation point of a spherical cold water drop translating in a saturated mixture of five components are presented. The environment surrounding the drop is composed of a condensable (steam), a noncondensable and nonabsorbable (air), a noncondensable but absorbable (e.g. elemental iodine), a chemically reactive component (e.g. methyl iodide), and a particulate substance. The dispersed and the continuous phases have been treated simultaneously. The effect of chemical reaction between a reactive component in the continuous phase and an additive (e.g. hydrazine) in the droplet liquid has also been considered. The droplet sizes, the thermodynamic range, and the nature of chemical constituents chosen for the illustrative calculations are closely related to the operating conditions that are likely to prevail in the containment spray atmosphere of a nuclear reactor following a loss of coolant accident. The choice of elemental iodine and methyl iodide has an added feature. The mass transfer resistance for elemental iodine is almost entirely in the gaseous phase while the absorption of methyl iodide could be regarded as a liquid-phase resistance-controlled process. Mass transport, in the presence of condensation, is seen to depend in a rather complicated manner on droplet radius.

1. INTRODUCTION

Material removal from the continuous phase by a translating liquid droplet experiencing laminar condensation has been considered earlier by Chung & Ayyaswamy (1978) (hereinafter referred to as Chung). The possibility of a chemical reaction between a component in the environment with one in the droplet liquid was also included there. However, in that study the effect of viscosity of the droplet liquid on the strength of the Hill's vortex inside the drop was not included. Also, the energy and mass transport equations appropriate for the thermal and concentration boundary layers inside the drop did not include the contribution of the tangential component of convection. The motivation for invoking these assumptions was primarily to reduce the complexity in the mathematical structure of the problem. With the inclusion of the viscosity effects for the droplet-inside, three separate regions of flow can, in general, be discerned: an interior core flow, a boundary layer type flow near the surface of the drop, and an internal wake flow. The extent and strength of each of the types of motion depends on: (a) the ratios of the dynamic viscosities and of the densities of that of the droplet liquid and of the outside medium, and (b) the Reynolds number characterizing the external flow. (See, e.g. Chung & Ayyaswamy 1981 for a detailed discussion.) In this paper, we present results based on a theory that simultaneously includes the effects due to the presence of hydrodynamic, thermal, and concentration boundary layers inside the drop coupled with the continuous phase boundary layers in the drop environment. The entire analysis will be restricted to the vicinity of the forward stagnation point of the moving droplet.

2. FORMULATION

Consider a single, cold water spherical droplet initially of uniform temperature T_0 , with fully developed and steady internal motion, that is translating at a constant velocity U_{∞} in a large content of an otherwise quiescent mixture of its own vapor (steam, condensable), air (noncondensable and nonabsorbable), and other components (noncondensable but absorbable). Elemental iodine, methyl iodide and a particulate material will form part of this multicomponent

mixture in the droplet environment. We shall take these materials to be present in trace amounts as in Chung. The droplet liquid will be assumed to contain a reactive reagent such as sodium thiosulfate or sodium hydroxide or hydrazine in extremely small amounts. Such additives serve two chemical functions: (a) they increase the solubility of certain materials in the drop liquid, and (b) they prevent the re-evolution of the dissolved material from the solution. Let the mixture be at $T_{\infty}(T_{\infty} > T_0)$ and the total pressure p_{∞} . The mass fractions of the noncondensable components in the bulk of the mixture are prescribed. The total pressure of the system is given by the sum of the partial pressures of the components in the bulk of the continuous phase. At the liquid and vapor-gas interface, the temperature T_i and mass fraction of each component are unknown *a priori* and are determined from two compatibility conditions that are explained below.

It is convenient to begin the analysis by separate considerations of each region in the model. We will also list the various assumptions where appropriate.

Region I: liquid region

(a) Inside the droplet the flow field is taken to consist of a thin hydrodynamic boundary layer near the droplet surface, an internal wake near the central axis of the droplet, and a Hill's vortex core flow with a reduced strength compared to that of a complete inviscid solution. This Hill's vortex is represented by:

$$\psi_{\rm core} = -(1/2)Ar^2(R^2 - r^2)\sin^2\theta.$$
 [1]

Following Prakash & Sirignano (1978), $(U_{\infty}/AR^2) = 10$, and the strength of the vortex is thus known. We have assumed that any reduction in the strength of the Hill's vortex is due only to the drop viscosity, i.e. the effect of condensation on the strength of the vortex is negligible (Chung & Ayyaswamy 1981).

The thin hydrodynamic boundary layer will be assumed to be fully developed, steady and laminar (Chao 1969). (b) The thermal boundary layer within the droplet will be assumed to be fully developed, thin, and can be treated as quasi-steady. (c) Since the flow velocity range to be considered is moderate, viscous dissipation, compressibility effects and expansion work can be neglected. (d) No droplet oscillation. (e) The concentration boundary layer appropriate for describing methyl iodide transport will be assumed to be thin and can be treated as quasi-steady. The methyl iodide-reagent chemical reaction can be considered to be of first order. When the methyl iodide is in dilute form and there is a large excess of reagents, this assumption is valid (Parsly 1971). (f) All of the elemental iodine mass transfer resistance is in the continuous phase (Parsly 1971). (g) With regard to particulate removal, we shall assume, following Suneja (1973) that if a particle has a contact with the droplet surface, it will be captured and so will vanish from the flow field. Therefore, for particulate capture, the mass transfer resistance is entirely derived from the continuous phase. The Stokes-Einstein relationship will be used for calculating the diffusion coefficients of particles (Friedlander 1967).

The governing equations for the liquid boundary layers are (see figure 1):

$$\frac{\partial}{\partial x}(\rho_L u_L r_0) + \frac{\partial}{\partial y}(\rho_L v_L r_0) = 0, \qquad [2]$$

$$\rho_L u_L \frac{\partial u_L}{\partial x} + \rho_L v_L \frac{\partial u_L}{\partial y} = \frac{\partial}{\partial y} \left(\mu_L \frac{\partial u_L}{\partial y} \right) + \rho_{L\infty} K_L^2 x - L(\rho_L - \rho_\infty),$$
^[3]

$$\rho_L u_L \frac{\partial T_L}{\partial x} + \rho_L v_L \frac{\partial T_L}{\partial y} = \frac{1}{C_{\rho_L}} \frac{\partial}{\partial y} \left(k_L \frac{\partial T_L}{\partial y} \right), \tag{4}$$



Figure 1. Geometry and coordinate system.

and

$$\dot{u}_{L}\frac{\partial C_{5L}}{\partial x} + v_{L}\frac{\partial C_{5L}}{\partial y} = \frac{\partial}{\partial y}\left(\mathcal{D}_{12}\frac{\partial C_{5L}}{\partial y}\right) - \tilde{K}C_{5L},$$
[5]

where $K_L = (U_{\infty}/10R)$, L = (gx/R), and \overline{K} is the rate constant for chemical reaction. The corresponding boundary conditions are:

$$u_{L} = (U_{\infty}/10R), T_{L} = T_{L,0}, C_{5L} = C_{5L,0} \text{ as } y \to -\infty$$

$$u_{L,i} = u_{vg,i}, \mu \frac{\partial u_{vg,i}}{\partial y} = \mu_{L} \frac{\partial u_{L,i}}{\partial y}, T_{L} = T_{i}, C_{5L} = HC_{5}, y = 0.$$
[6]

Here H is the distribution coefficient, C_{5L} is the concentration of methyl iodide in the liquid phase, while C_5 is in the continuous phase. We note that the appropriate boundary condition at the inside edge of the hydrodynamic boundary layer corresponds to the Hill's vortex, with strength $A = U_{\infty}/10R^2$, evaluated at the surface of the droplet r = R. The temperature at the edge of the thermal boundary layer is taken to be the instantaneous temperature of the outermost isothermal streamline of the internal thermal core.

The interfacial tangential velocity is governed by (i) matching of shear stress, and (ii) continuity of tangential velocity. Thus, for the interfacial condensate layer, $U_{L,i} \rightarrow 0$ as $x \rightarrow 0$ as in Chung. Also, in view of the thinness of the condensate layer, it will be assumed that the thermal resistance across this layer is negligibly small such that $T_s = T_i$ (Chung & Ayyaswamy 1981).

Region II: continuous phase boundary layer

(a) The flow field in the vicinity of the forward stagnation point will be taken to be a laminar boundary layer. (b) Since the flow velocity range to be considered is moderate, viscous dissipation, compressibility effects and expansion work will be ignored. (c) The interfacial resistance, the thermal-diffusion and diffusion-thermo effects will be assumed to be neglibible. The continuous phase boundary layer equations are then as follows:

$$\frac{\partial}{\partial x} \left(\rho u r_0\right) + \frac{\partial}{\partial y} \left(\rho v r_0\right) = 0, \qquad [7]$$

$$\rho u \frac{\partial u}{\partial x} + \rho v \frac{\partial u}{\partial y} = \frac{\partial}{\partial y} \left(\mu \frac{\partial u}{\partial y} \right) + \rho_{\infty} K^2 x - L(\rho - \rho_{\infty}), \qquad [8]$$

$$\rho u \frac{\partial T}{\partial x} + \rho v \frac{\partial T}{\partial y} = \frac{1}{C_p} \frac{\partial}{\partial y} \left(k \frac{\partial T}{\partial y} \right) + \frac{\rho}{C_p} \left\{ \sum_{n=1}^{4} \left(\sum_{m=1}^{5} D_{mn} C_{p_m} \right) \frac{\partial w_n}{\partial y} \right\} \frac{\partial T}{\partial y},$$
[9]

and

$$\rho \,\frac{\partial w_m}{\partial x} + \rho v \,\frac{\partial w_m}{\partial y} = \sum_{n=1}^4 \frac{\partial}{\partial y} \,\rho D_{mn} \,\frac{\partial w_n}{\partial y}, \, m = 1, 2, 3 \text{ and } 4.$$
^[10]

Here $K = (3/2) (U_{\infty}/R)$, L = (gx/R), and subscripts 1, 2, 3, 4 and 5 refer to air, steam, elemental iodine, particulate and methyl iodide, respectively. The diffusion coefficients which are highly sensitive to composition are calculated from expressions given by Taitel & Tamir (1974). The corresponding boundary conditions are:

i-surface:
$$y = 0$$

$$\mu \frac{\partial u_{vg,i}}{\partial y} = \mu_L \frac{\partial u_{L,i}}{\partial y}, v_{L,i} = 0$$

$$T = T_L = T_i, w_1 = w_{1,i}(T_i, p_{\infty}), w_2 = w_{2,i}, w_3 = w_4 = 0,$$

$$w_5 = w_{5,i}, u_{vg,i} = u_{L,i} = GU_{\infty} \frac{x}{R}.$$
[11]

Here G is a measure of the surface mobility. This is an unknown and has to be determined through the use of the shear stress continuity condition. At e-surface: $y \rightarrow \infty$

$$u = \frac{3}{2} \left(\frac{U_{\infty} x}{R} \right), T = T_{\infty}, w_1 = w_{1,\infty}, w_2 = w_{2,\infty},$$

$$w_3 = w_{3,\infty} = \epsilon_3, w_4 = w_{4,\infty} = \epsilon_4, w_5 = w_{5,\infty} = \epsilon_5,$$
 [12]

with

$$\epsilon_3 + \epsilon_4 + \epsilon_5 = \epsilon, w_{1,\infty} + w_{2,\infty} + \epsilon = 1.$$

Compatibility conditions:

(i) The interface is impermeable to air. Thus

$$\left(\sum_{n=1}^{4}\rho D_{mn}\frac{\partial w_n}{\partial y} - \rho_1 v\right)_i = 0.$$
 [13]

(ii) The energy balance equation which connects the two boundary layers is:

$$k_L \frac{\partial T_L}{\partial y} \bigg|_s = \dot{m}\lambda + k \frac{\partial T}{\partial y} \bigg|_i$$
[14]

where $\dot{m} = mass$ condensation rate

$$\dot{m} = -\rho v|_{i}.$$
[15]

Equations [2]-[5] subject to conditions [6] and [7]-[10] subject to surface conditions [11]-[15] must be solved.

SIMILARITY TRANSFORMATION

A similarity transformation can be effected through the use of:

$$\rho_L u_L = \frac{1}{r_0} \frac{\partial(\psi_L r_0)}{\partial y}, \rho_L v_L = -\frac{1}{r_0} \frac{\partial(\psi_L r_0)}{\partial y}, \eta_L = y\sqrt{(C_L)}/R,$$
$$\psi_L = \mu_{L\infty}\sqrt{(C_L)}xf_L/R, \theta_L = (T_L - T_i)/(T_\infty - T_i), \theta = \frac{T - T_i}{T_\infty - T_i},$$
$$\rho u = \frac{1}{r_0} \frac{\partial(\psi r_0)}{\partial y}, \rho v = -\frac{1}{r_0} \frac{\partial(\psi r_0)}{\partial x}, \eta = \frac{y\sqrt{C}}{R}, \psi = \mu_\infty \sqrt{(C)}x\frac{f}{R}$$

where

$$C = \operatorname{Re}_{\infty} = \frac{3}{2} \frac{U_{\infty}R}{\nu_{\infty}}$$
, and, $C_L = \operatorname{Re}_{L,\infty} = \frac{U_{\infty}R}{10\nu_{L^{\infty}}}$.

With variables transformed as above, the similarity transformation yields the ordinary differential equations for transport near the forward stagnation point.

Since we have assumed that all the components other than steam and air are present only in trace amounts, a regular perturbation analysis of the resultant ordinary, nonlinear, differential equations can be made in order to obtain a zeroth order and a first order set of equations. The zeroth order equation will then correspond to the case of laminar condensation of binary mixture of vapor and noncondensable air on a translating droplet. The first order solution will describe the transport of the other components. Therefore, we write

$$w_{1} = w_{10} + \epsilon w_{11} + \dots + w_{2} = w_{20} + \epsilon w_{21} + \dots + w_{3} = \epsilon w_{30} + \epsilon^{2} w_{31} + \dots$$

$$w_{4} = \epsilon w_{40} + \epsilon^{2} w_{41} + \dots + w_{5} = \epsilon w_{50} + \epsilon^{2} w_{51} + \dots$$

$$w_{5L} = \epsilon w_{5L0} + \epsilon^{2} w_{5L1} + \dots + f = f_{0} + \epsilon f_{1} + \dots$$

$$f_{L} = f_{L0} + \epsilon f_{L1} + \dots + \theta = \theta_{0} + \epsilon \theta_{1} + \dots + \theta_{L} = \theta_{L0} + \epsilon \theta_{L1} + \dots$$

The zeroth order equations are:

For the continuous phase boundary layer:

$$[\phi_{\mu}(f'_{0}/\phi_{\rho})']' + 2f_{0}[f'_{0}/\phi_{\rho}]' + [1 - \phi_{\rho}\left(\frac{f'_{0}}{\phi_{\rho}}\right)^{2} - \operatorname{Fr}(\phi_{\rho} - 1)] = 0(\epsilon),$$
[16]

$$\frac{1}{\phi_{C_p} \Pr_{\infty}} (\phi_k \theta'_0 + \phi_{\mu} / \text{Sc} \ \frac{C_{p1 \to 2}}{C_p} w'_{10} \theta'_0 + 2f_0 \theta'_0 = 0(\epsilon) , \qquad [17]$$

$$[\phi_{\mu}w_{10}'/\mathrm{Sc}] + 2f_0w_{10}' = 0(\epsilon), \qquad [18]$$

and for the liquid boundary layer

$$[\phi_{\mu L}(f_{L0}'\phi_{\rho L})']' + 2f_{L0}[f_{L0}'\phi_{\rho L}]' + [1 - \phi_{\rho L}\left(\frac{f_{L0}'}{\phi_{\rho L}}\right)^2 - \operatorname{Fr}_L(\phi_{\rho L^{-1}})] = 0(\epsilon), \quad [19]$$

$$\frac{1}{\phi_{C_{pL}} \Pr_L} (\phi_{kL} \theta'_{L0})' + 2f_{L0} \theta'_{L0})' = OCE).$$
[20]

The corresponding boundary conditions and compatibility conditions become:

e-surface

$$\frac{f'_0}{\phi_{\rho}} \to 1, \ w_{20} = w_{2,\infty}, \ w_{10} = 1 - w_{20}, \ \theta_0 = 1, \ \eta \to \infty \ .$$
[21]

i-surface

$$f_{L0} = 0, 15(\mu/\mu_L) \sqrt{\left(\frac{c}{c_L}\right) \left(\frac{f'_0}{\phi_\rho}\right)'} = \left(\frac{f'_{L0}}{\phi_{\rho L}}\right)', 15\left(\frac{f'_0}{\phi_\rho}\right) = \left(\frac{f'_{L0}}{\phi_{\rho L}}\right) = 10G_0$$

$$\theta_0 = \theta_{L0} = 0, w_{10} = w_{10} (p_{\infty}, T_{i0}) = \frac{1 - (p_{v,i}(T_{i0})/p_{\infty})}{\left(\frac{M_v}{M_g}\right) (p_{v,i}(T_{i0})/p_{\infty}) + 1}$$

$$G = G_0 + \epsilon G_1$$
, and $w_{20} = 1 - w_{10}$, as $\eta = 0$ [22]

$$(\phi_{\mu}/\mathrm{Sc})w_{10}' + 2f_{0,i}w_{10} = 0, f_{0,i} = \frac{\Delta T_i}{2\mu_{\infty}\lambda} \left\{ k_L \theta_{L0,i}' \sqrt{\left(\frac{C_L}{C}\right) - k\theta_{\nu g0,i}'} \right\}, \eta = 0, \qquad [23]$$

and

$$(f'_{L0}/\phi_{\rho L}) \rightarrow 1, \ \theta_{L0} = \theta_{L,0}, \ \eta_L \rightarrow -\infty.$$
 [24]

Next, the first order equations become:

$$(\phi_{\mu}(f_{1}'/\phi_{\rho})')' + 2f_{0}(f_{1}'/\phi_{\rho})' + 2f_{1}(f_{0}'/\phi_{\rho})' - 2\frac{f_{0}'f_{1}'}{\phi_{\rho}} = 0,$$
^[25]

$$\frac{1}{\phi_{C_p} \Pr_{\alpha}} (\phi_k \theta_1')' + \left(\frac{\phi_{\mu}}{C_p} \sum_{n=1}^4 \left(\sum_{m=1}^5 \left(\frac{C_{p_m}}{\mathrm{Sc}_{mn}}\right)\right) (w_{n0}' \theta_1' + w_{n1}' \theta_0') + 2(f_0 \theta_1' + f_1 \theta_0') = 0, \quad [26]$$

$$\sum_{n=1}^{2} \left(\frac{\phi_{\mu} w_{n1}'}{\text{Sc}_{mn}}\right)' + \sum_{n=3}^{4} \left(\frac{\phi_{\mu} w_{n0}' A_{n}}{\text{Sc}_{mn}}\right)' + 2(f_{0} w_{m1}' + f_{1} w_{m0}') = 0, \quad m = 1, 2$$
[27]

and

$$\sum_{n=1}^{2} \left(\frac{\phi_{\mu} w'_{n1}}{\text{Sc}_{mn}} \right)' + \sum_{n=3}^{4} \left(\frac{\phi_{\mu} w'_{n0} A_n}{\text{Sc}_{mn}} \right)' + 2f_0 w_{m0} A_m = 0, \, m = 3, 4$$
[28]

with

$$\left(\phi_{\mu L}\left(\frac{f_{L1}'}{\phi_{\rho L}}\right)'\right)' + 2f_{L0}\left(\frac{f_{L1}'}{\phi_{\rho L}}\right)' + 2f_{L1}\left(\frac{f_{L0}'}{\phi_{\rho L}}\right)' - 2\frac{f_{L0}'f_{L1}}{\phi_{\rho L}} = 0,$$
[29]

$$\frac{1}{\phi_{C_pL} \Pr_{\infty}} (\phi_{KL} \theta'_{L1})' + 2(f_{L0} \theta'_{L1} + f_{L1} \theta'_{L0}) = 0, \qquad [30]$$

and

$$\left(\frac{\phi_{\mu L}}{\mathrm{Sc}_{5L}}w'_{5L0}\right)' + 2f_{L0}w'_{5L0} - \bar{K}'w_{5L0} = 0, \qquad [31]$$

where

$$\bar{K'} = \bar{K}R^2\phi_{\rho L}/(\nu_{L\infty}\mathrm{Re}_L), A_1 = \frac{\epsilon_3}{\epsilon}, A_2 = \frac{\epsilon_4}{\epsilon}, A_3 = \frac{\epsilon_5}{\epsilon}$$

and

$$A_1 + A_2 + A_3 = 1.$$
 [32]

The corresponding boundary conditions and compatibility conditions are: e-surface

$$\frac{f_1'}{\phi_{\rho}} \rightarrow 0, \ \theta_1 = 0, \ w_{11} = -1, \ w_{21} = 0, \ w_{30} = A_1, \ w_{40} = A_2,$$

and

$$w_{50} = A_3, \text{ as } \eta \to \infty \,. \tag{33}$$

i-surface:

$$f_{L1} = 0, 15 \left(\frac{\mu}{\mu_L}\right) \sqrt{\left(\frac{C}{C_L}\right) \left(\frac{f_1'}{\phi_\rho}\right)'} = \left(\frac{f_{L1}}{\phi_{\rho L}}\right)', 15 \left(\frac{f_1'}{\phi_\rho}\right) = \left(\frac{f_{L1}}{\phi_{0L}}\right)' = 10G_1, \ \theta_1 = \theta_{L1} = 0,$$
$$HC_{50} = C_{5L0}, \ w_{11} = -A_3', \ w_{21} = 0, \ w_{30} = 0, \ w_{40} = 0, \ w_{50} = A_3',$$
$$\sum_{n=1}^2 \left(\frac{\phi_n}{Sc_{mn}} w_{n1}'\right) + \sum_{n=3}^4 \left(\frac{\phi_n}{Sc_{mn}} w_{n0}'\right) + 2(f_0 w_{11} + f_1 w_{10}) = 0, \ \eta = 0,$$
 [34]

and

$$2(w_{20}f_1 + w_{21}f_0) = \frac{\Delta Ti}{\mu_{\infty}\lambda} \left(k_L \theta'_{L1,i} \sqrt{\frac{C_L}{C}} - k \theta'_{vgl,i} \right) - \sum_{n=1}^2 \left(\frac{\phi_\mu}{Sc_{2n}} w'_{n1} \right) - \sum_{n=3}^4 \left(\frac{\phi_\mu}{Sc_{2n}} w'_{n0} \right), \quad [35]$$

L-surface:

$$\left(\frac{f_{L1}'}{\phi_{\rho L}}\right) = 0, \ \theta_{L1} = \theta_{L1,0}, \ w_{5L0} = w_{5L0,0}, \ \eta_L \to -\infty$$
[36]

In the above $A'_3 = (\epsilon'/\epsilon)$ where ϵ' is the concentration of methyl iodide at the interface, and this is not prescribed at the beginning but is obtained during solution matching. The above set of equations [16]-[20] subject to conditions [21]-[24], and [25]-[32] subject to conditions [33]-[36], were solved numerically by quasi-linearization method.

The calculation starts with an appropriate initial guess of the interface temperature $T_i(T_0 < T_i < T_\infty)$, and then the simultaneous zeroth order equations are solved. With the zeroth order solutions as the input, the first order solutions are numerically evaluated by the same quasi-linearization technique. The solution to the problem is obtained when the difference between the calculated $\theta_{L,0}$ and the actual $\theta_{L,0}$ meets a set convergence criterion. It is very important to note that G, the measure of the surface mobility, is an unknown. The entire set of equations have to be repeatedly iterated upon until the shear stress continuity conditions are identically satisfied and a unique G is determined for each set of parameter values. This

complicated portion of the numerical procedure was not required in Chung in view of the neglect of the internal viscosity there.

RESULTS AND DISCUSSION

Numerical solutions for material transport, associated with condensation on a translating droplet are shown in figures 2-5. The variation in the non-condensable gas mass fraction (air) explored includes the range $0.01 < w_{1,\infty} < 0.5$. The droplet radius, R, lies in the range 0.025 cm < R < 0.075 cm. The ambient temperature is fixed at 125°, and the atmosphere is taken to consist of saturated air and other components in trace amounts. The terminal velocities, U_{∞} , corresponding to various ambient conditions have been calculated using an equation recommended



Figure 2. Effect of droplet size on the material transport of iodine; $T_{\infty} = 125^{\circ}$ C; R = 0.075 cm, 411 cm/s $< U_{\infty} < 422$ cm/s; R = 0.05 cm, 322 cm/s $< U_{\infty} < 355$ cm/s; R = 0.025 cm, 146 cm/s $< U_{\infty} < 153$ cm/s.



Figure 3. Particulate removal due to condensation.



Figure 4. Effect of chemical reaction and distribution coefficient on methyl iodide removal, R = 0.05 cm, $w_{1,\infty} = 0.05$, k = 1 s⁻¹.----, Danckwerts solution (1951).



Figure 5. Effect of chemical reaction rate constant on the methyl iodide removal R = 0.05 cm, $w_{1,\infty} = 0.05$, $H = 10^3$.

by Lapple (1951):

$$U_{\infty} = 0.153 \, \xi \left\{ \frac{D\xi}{\nu_{\infty}} \right\}^{0.428}, \text{ where } \xi = \{ gD \, (\rho_L / \rho_{\infty} - 1) \}^{1/2}, 2 \le \text{Re}_{\infty} \le 10^3.$$
 [37]

It is recognized that the terminal velocity given by [37] does not reflect the effects due either to

interfacial condensation velocity or of the increase in droplet size. It does not also involve μ_L . However, for finite interfacial velocities and very small increases in droplet size, the correlation has been shown to demonstrate good comparison with experiment and will therefore be employed here.

Figure 2 shows the variation in \dot{m} , the absolute mass transport value for elemental iodine, as a function ΔT , for various droplet sizes and $w_{1,\infty}$, *m* is a dimensional quantity. The mass transport depends on the radius of the droplet in a rather complex manner as evident from this figure. To explain this feature, consider the following. It can be shown, from a simple dimensional analysis, that for a given ΔT and $w_{1,\infty}$, *m* for a translating droplet is proportional to $U_{\infty}^{1/2}/R^{1/2}$, where $U_{\infty} = U_{\infty}(R)$ only. Furthermore, the functional dependence of U_{∞} on R is nonlinear, and U_{x} increases with increasing R until a certain size is reached and then after the rate of velocity increase is lower compared to the rate of radius increase. The overall effect is such that, for a given ΔT and $w_{1,x}$, *m* could be higher or lower for various droplet sizes, the dependence itself being nonstraightforward. Thus, there appears to be an optimal droplet size for material transport under any given conditions. Also, from figure 2 it is clear that for the same droplet size and for a given ΔT , the absolute value of iodine transport decreases with increasing $w_{1,\infty}$. Comparison of figure 2 here with figure 4 of Chung shows that the mass transport results were overpredicted there by as much as a factor of (3/2) in several cases. With decreasing noncondensable gas in the bulk, the theory of Chung becomes increasingly inaccurate. The neglect of viscous effects in the dispersed phase contribute to the high values reported in Chung. A purely inviscid solution such as the one used for describing the velocity field inside the drop predicts convective velocity components which are about an order of magnitude higher than those realized in the present study and the transport results based on such a theory are likely to be exaggerated when dealing with liquid droplets in gas-mixture environment. With increasing mass fraction of the noncondensables in the bulk, the discrepancy between the present results and those of Chung are somewhat smaller. This is owing to the increasingly dominant presence of a new resistance in the continuous phase with increasing $w_{1,\infty}$.

Figure 3 is about particulate capture. The results depict the capturing of solid particles by a spray droplet moving in a condensation flow field. The extent of capture itself is presented in terms of a dimensionless quantity, (\dot{m}/\dot{m}_0) , that is based on a reference mass flux, \dot{m}_0 . The reference mass flux is derived from a pseudo-physical situation where a droplet is translating in an environment made of one component that has the same physical properties as steam but will not condense, while the others are noncondensable but absorbable substances that exist in trace amounts. The flow field in this latter case is described by the well known Homann's solution (Homann 1936). The Homann's solution itself, however, cannot be explicitly expressed in terms of a single equation but can be obtained by a straightforward numerical integration. The dimensionless manner of presentation explicitly exhibits the features introduced by the presence of noncondensables and forced flow. The diffusion coefficients of the particles in the continuous phase required for calculations are estimated through the use of Stokes-Einstein relationship (Friedlander 1967). The range of parameters investigated include, $0.025 \le R \le$ 0.075 cm, $0.01 < w_{1,\infty} < 0.05$ and the ratio of particle radius to that of droplet, I, in the interval $5 \times 10^{-6} < I < 10^{-5}$. For a given ΔT , the dimensionless mass flux which is a direct measure of the extent of particle capture, decreases with increasing $w_{1,\infty}$. This is due to the weakening of the condensation flow field due to the presence of the noncondensable. Owing to the smallness of the particle size, and the low particle density due to the presence in trace amounts that are considered in our analysis, it is not necessary to include the effect of particles themselves on the flow field (Friedlander 1967, Suneja 1973). For a given ΔT and $w_{1,x}$, the dimensionless mass flux increases with higher I. This can be understood when we realize that particle size modifies the diffusion coefficient. Comparison of figure 3 here with figure 5 of Chung shows that the extent of particulate capture was severely overpredicted in several cases there. With increasing droplet radius and the size of the particle captured, and a simultaneous decrease in the noncondensable gas concentration, the theory of Chung which ignores the effect of internal viscosity of the droplet yields increasingly inaccurate results.

Figure 4 shows the methyl iodide transport, \dot{m} , as a function of the mass transfer driving potential, $\Delta C = (C_{\infty} - C_0/H)$ and ΔT . Here C_{∞} is the molar concentration of methyl iodide in the free stream, and there its value has been normalized to unity. The droplet is assumed to contain the chemical additive hydrazine. The rate constant for this first order chemical reaction is known (Hasty 1969). The results presented are for various assumed values of H and ΔT . For given ΔC and ΔT , \dot{m} increases with H. This is as would be expected. However, the effect of varying H becomes significant at higher ΔT values. For given ΔC and H, an increase in ΔT increases *m*. This is directly due to the strengthening of the condensation flow field with higher ΔT . Figure 4 provides a comparison between our results with those of Danckwerts (1951) for $H = 10^3$, $\bar{K} = 1 \, \mathrm{s}^{-1}$. The Danckwerts solution corresponds to the absorption by a stagnant drop, and the results obtained from that model show favorable comparison with the experimental results for the removal rates of methyl iodide (Postma et al. 1975). The close agreement with the present results can be explained by the fact that the methyl iodide removal process, at small enough \bar{K} values, is essentially liquid-phase resistance controlled. The Danckwerts model will be invalid when the vapor phase resistance is comparable to the liquid-phase value. In this region, our model will still provide useful results. The agreement with the Danckwerts model, and hence with the experimental results, demonstrated by the present theory is very much better when compared with Chung. In both figures 4 and 5 the droplet radius and $w_{1,\infty}$ have been taken to be fixed at 0.05 cm and 0.05, respectively. Figure 5 illustrates the effect of the chemical reaction rate constant together with condensation on methyl iodide removal. By investigating the removal rates at different values of the rate constant, the effect of various chemical additives could be appreciated. It is readily evident that with increasing rate constant, hence enhanced chemical reaction, the removal rate of methyl iodide in the presence of a condensation flow field increases.

In the present paper, we have performed a quasi-steady analysis. It has been shown by Chung (1963) that a quasi-steady analysis of the transient process involving heat removal in the presence of condensation yields sufficiently accurate results. The total removal rates, during the droplet's lifetime, can be ascertained by an integration of the various steady state transfer rates over the many, discrete and finite time intervals that comprise the total time. Every discrete steady state transfer rate is based on an updated bulk-liquid condition which is derived from the previous transfer rate.

CONCLUSIONS

Numerical solutions to the nonlinear, coupled boundary layer equations governing laminar condensation heat and mass transfer in the vicinity of the forward stagnation point of a freely falling spherical cold water droplet in a mixture of steam, air and other materials are presented. The environment surrounding the droplet is composed of a condensable (steam), a noncondensable and nonabsorbable (air), a noncondensable but absorbable (e.g. elemental iodine), a chemically reactive component (e.g. methyl iodide) and a particulate substance. The effect of chemical reaction between the reactive component in the continuous phase and an additive in the droplet liquid has thus been considered. The theoretical basis of this study is considerably more refined compared to an earlier one (Chung & Ayyaswamy 1978). Significantly, the droplet-inside has been carefully treated and effects due to the presence of hydrodynamic and thermal boundary layers inside the drop are included in evaluating transport rates. The effect of viscosity on the internal core motion and the consequent reduction in the vortex strength inside the drop has been accounted for. The neglect of the tangential convective component in the thermal and concentration boundary layer equations of the earlier study has been remedied by their inclusion. The relation between the continuous phase and the dispersed phase has been carefully defined. The interfacial shear stress continuity conditions have been identically satisfied by employing repeated iterative procedures.

With the inclusion of the viscosity effects for the droplet-inside, the convective velocity components are reduced by an order of magnitude compared to that of an inviscid Hill's spherical vortex solution. The associated heat and mass transfer predictions are significantly smaller in magnitude.

Acknowledgement—The authors are indebted to one of the referrees for many helpful comments, to Dr. E. B. Dussan V, Dr. S. S. Sadhal and Mr. T. Sundararajan for many useful suggestions and advice. Sponsorship of this work by the National Science Foundation under Grant ENG 77-23137 is gratefully acknowledged.

NOMENCLATURE

- A strength of the Hill's vortex as used in Chung
- C_p specific heat at constant pressure
- D diameter of the droplet
- \mathscr{D}_{12} binary diffusion coefficient
- D_{ij} multicomponent diffusion coefficients
- f dimensionless stream function
- g acceleration due to gravity
- k thermal conductivity of mixture
- m mass flux
- M molecular weight
- p pressure
- Pr Prandtl number = ν/κ
- Q heat flux
- r_0 radial distance from the axis of symmetry
- r r-direction in spherical coordinate
- R radius of droplet
- Re Reynolds number = $(3/2)(U_{\infty}R/\nu_{\infty})$
- Sc Schmidt number = ν/\mathcal{D}_{12}
- Sc_{ij} Schmidt number = ν/D_{ij}
- T temperature
- T_0 droplet initial bulk temperature; instantaneous bulk temperature subsequently
- *u* velocity in the *x* direction
- v velocity in the y direction
- U_x terminal velocity of the droplet, free stream velocity
- w mass fraction
- x coordinate measuring distance along circumference from the stagnation point
- y coordinate measuring radial distance outward from the interface

Greek symbols

- $\Delta T = T_{\infty} T_0$
- $\Delta T_i = T_\infty T_i$
 - ϵ mass fraction of fission products in the bulk
 - η similarity variable = $y\sqrt{c}/R$
 - θ angle in spherical coordinates
 - θ dimensionless temperature
 - $\theta_L (T_L T_i)/(T_\infty T_i)$
 - κ thermal diffusivity = $k/\rho C_p$
 - λ latent heat of vaporization

- μ dynamic viscosity
- ν kinematic viscosity = μ/ρ
- ρ density
- σ surface tension coefficient
- ϕ_{C_p} ratio (C_p/C_{p_m})
- ϕ_k ratio (k/k_{∞})
- ϕ_{μ} ratio (μ/μ_{∞})
- ϕ_{ν} ratio $(2\nu_{\infty}/\nu_L)$
- ϕ_{ρ} ratio (ρ/ρ_{∞})
- Ψ stream function

Subscripts

- e at the outside edge of the continuous phase boundary layer
- g gas
- *i* at the dispersed-continuous phase interface
- L liquid
- l, i in the liquid at the interface
- 0 bulk condition in droplet
- s at the s-surface
- v condensable in vapor phase
- v, i of the condensable vapor at the interface
- vg, i of the continuous phase at the interface
 - 1 air
 - 2 vapor or steam
 - 3 elemental iodine
 - 4 particulate
 - 5 methyl iodide
 - ∞ in the bulk phase, far away from the droplet

REFERENCES

- CHAO, B. T. 1969 Transient heat and mass transfer to a translating droplet. J. Heat Transfer 91, 273-281.
- CHUNG, P. M. 1963 Unsteady laminar film condensation on a vertical plate. J. Heat Transfer 85, 63-70.
- CHUNG, J. N. & AYYASWAMY, P. S. 1978 Material transport with steam condensation on a moving spray droplet including the effect of internal chemical reaction. *Topics in Two-Phase Heat Transfer and Flow* (Edited by S. G. Bankoff), pp. 153-164. ASME, New York.
- CHUNG, J. N. & AYYASWAMY, P. S. 1981 Laminar condensation heat and mass transfer to a moving drop. AIChE J. To be published.

DANCKWERTS, P. V. 1951 Absorption by simultaneous diffusion and chemical reaction into particles of various shapes, and into falling drops. *Trans. Faraday Soc.* 47, 1014–1023.

FRIEDLANDER, S. K. 1967 Particle diffusion in low-speed flows. J. Colloid and Interface Sci. 23, 157-164.

- HASTY, R. A. 1969 The rate of reaction of methyl iodide and hydrazine in aqueous solutions. J. Phys. Chem. 73.
- HOMANN, F. 1963 Der Einfluß Großber Zahigkeit bei der stromung un den Zylinder und um die Kugel. ZAMM 16, 153-164; Forsch. Ing-Wes. 7, 1-10.
- LAPPLE, C. E. 1951 Fluid and Particle Mechanics. Univ. Press, Delaware.
- PARSLY, L. F. 1971 Spray prpgram at the nuclear safety pilot plant. Nucl. Tech., J. Am. Nucl. Soc. 10, 472-485.

- POSTMAN, A. K., RITZMAN, R. L., GIESECKE, J. A. & SCHMIDT, E. W. 1975 Models for predicting the removal of airborne contaminants by reactor containment sprays. Battelle Pacific Northwest Laboratories Rep. BNWL-B-417, UC-78.
- PRAKASH, S. & SIRIGNANO 1978 liquid fuel droplet heating with internal circulation Int. J. Heat Mass Transfer 21, 885-895.
- SUNEJA, S. K. 1973 Capture of particles by drops falling at high speeds. J. Colloid Interface Sci. 44, 509-516.
- TAITEL, Y. & TAMIR, A. 1974 Film condensation of multicomponent mixtures. Int. J. Multiphase Flow 1, 697-713.