ergy Technical Center. We wish to also acknowledge the comments of R. I. McNeil and A. Bruce King of Gulf Research & Development Co.

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Manuscript received January 12, 1983; revision received May 26, and accepted June 10, 1983.

Coupled Boundary Layer Transport Involving Particles

Particle deposition by diffusiophoresis and thermophoresis is described using the linearized Fickian multicomponent diffusion equations. For these cases, the diagonal component of the diffusivity matrix for the particle is expected to be much smaller than the other diagonal components. Intuitive arguments, dimensional analyses and the standard interfacial transport theories in multicomponent form suggest that the larger boundary layer due to the larger diffusivity component describes the transition between bulk and surface values for both components.

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SCOPE

Coupled transport is characterized by a flux caused by a driving force not normally associated with the flux in question. The formalism for describing coupled transport processes involves a matrix of diffusivities, yielding straightforward but somewhat more complex computational procedures. Two examples of coupled transport are multicomponent diffusion and thermal diffusion. It has been shown that diffusiophoresis, the migration of a particle in a solute gradient, is a coupled transport process, describable as a multicomponent diffusion phenomenon. When particles are involved, thermal diffusion is often called thermophoresis; and this too has been modeled as a multicomponent diffusion phenomenon. These effects appear as a "cross term" in the coupled transport model. Both diffusiophoresis and thermophoresis have been suggested as methods for enhancing particle capture from gas and liquid streams. The description of coupled transport in the boundary layer provides a straightforward method for modeling particle collection by these mechanisms.

CONCLUSIONS AND SIGNIFICANCE

A general formalism for coupled boundary layer transport for low flux situations is presented. Even though in the particle migration situations of interest the uncoupled boundary layer thicknesses differ by about two orders of magnitude, it is shown

intuitively and by dimensional analysis that for strongly coupled transport one boundary layer thickness uniquely characterizes the system. However, the rigorous matrix analysis is still necessary for calculation of the mass transfer fluxes. Solutions for the film theory, penetration theory and boundary layer theory in coupled form substantiate this concept.

The model presented, while valid for any coupled transport situation, is especially useful for diffusiophoresis and thermophoresis. In diffusiophoresis, the "main term" diffusivity of the particle (due to its own gradient) is usually small. However, the present model illustrates that this does not simplify to an uncoupled equation because of the importance of the solute "main term" diffusivity. For thermophoresis, the same statement holds true regarding the relative importance of the coefficient relating heat flux and temperature gradient. The coupled transport model is not only simpler than some previous models for thermophoresis of particles in boundary layers, it is more rigorous.

INTRODUCTION

Diffusiophoresis involves the migration of a particle in a solute gradient. It has been studied in connection with acceleration of latex particle deposition (Smith and Prieve, 1982) and enhancement of particle capture. (Prieve et al., 1979). Rigorous theories for the particle velocity in electrolyte (Anderson, 1980; Prieve et al., 1984) nonelectrolyte (Anderson et al., 1982) gradients have also been presented. Recently, it has been shown that diffusiophoresis is describable as a multicomponent diffusion phenomenon (Shaeiwitz and Lechnick, 1984). This arose based on the rules normally associated with multicomponent diffusion, was derived from irreversible thermodynamics and was compared to the theories of diffusiophoresis by the ease of conversion between velocities and fluxes.

Thermophoresis of particles involves migration in a temperature gradient, usually toward the lower temperature. It has also been studied extensively as a mechanism for enhancing particle capture (Whitmore and Meisen, 1977; Rosner, 1980), as well as being a mechanism for fouling heat exchangers. Several models for particle capture in a boundary layer by this mechanism have been proposed, but they always involved addition of a term to the conservation equation of the particle, with no regard for the energy conservation equation (Walker et al., 1979; Rosner, 1980). This led to a conceptual picture of thermophoretic deposition involving a thermal boundary layer much larger than the particle mass transfer boundary layer (Rosner, 1980).

Numerous multicomponent transport models have appeared since the original work demonstrating that application of matrix methods allowed straightforward solution of the coupled equations of change both in Stefan-Maxwell form (Stewart and Prober, 1964) and in Fickian form (Toor, 1964). A necessary assumption was that the equations were "linearized;" that is, the transport coefficients were assumed constant. Some examples of subsequent applications include numerous involving mass transfer with chemical reaction (Toor, 1965; Tavlarides and Gal-or, 1969; Othmer and Scriven, 1969), multicomponent gas absorption including heat effects (DeLancey and Chiang, 1970; DeLancey, 1972), and multicomponent convective transport (Gal-or and Padmanabhan, 1968). Recently, it has also been shown that the Stefan-Maxwell formulation often provides a more convenient method of analysis since these diffusivities (mass or heat) may be predicted from the component binary diffusivities, at least in gas systems (Krishna and Standart, 1979).

With the relevance of diffusiophoresis and thermophoresis in enhancing particle migration in particle capture situations, it is important to have conceptually valid models for the capture process. The purpose of this work is to develop simple cases of these models in order to draw conceptual conclusions. The two major assumptions involved are the zero flux assumption and a constant diffusivity matrix. The former simplifies the mathematics, and the latter is a reasonably standard assumption. The diffusivities may be evaluated at average conditions; otherwise, a specific expression for the concentration and/or temperature dependence is required. The Fickian multicomponent diffusion formalism will be used to describe both mechanisms of particle migration, and the standard interface transport theories will be examined in multicomponent form. The Fickian form is used in place of the Stefan-Maxwell form because diffusiophoresis in the presence of electrolytes, the situation for which a significant amount of work has been done, takes place in the liquid phase. The binary diffusivities in the Stefan-Maxwell equations do not have the same meaning in liquids, and are in general not predicable from any theory. Secondly, since the solution in Fickian form has features of the binary case, it is easier to generalize concepts based on binary solutions when the equations are in this form.

The novelty of this work is that the specific case of "main term" (diagonal) diffusivities differing by at least two orders of magnitude will be examined closely. Most previous investigations involved molecular gas systems, in which binary, and hence "main term" diffusivities are normally expected to be of similar magnitude. In the binary case involving particles migrating due to a solute or a temperature gradient, the particle diffusivity is expected to be several orders of magnitude smaller than either the solute or the thermal diffusivity. Hence, the individual boundary layer thicknesses would also be expected to differ by several orders of magnitude. As we will see, this is not necessarily the case for a coupled transport situation.

MULTICOMPONENT TRANSPORT

The macropscopic equations for the coupled fluxes, derivable from irreversible thermodynamics (Miller, 1960), are

$$-J_1 = D_{11}\nabla C_1 + D_{12}\nabla C_2 \tag{1a}$$

$$-J_2 = D_{21}\nabla C_1 + D_{22}\nabla C_2 \tag{1b}$$

for coupled diffusion of two species, and are

$$-J_1 = D_{11}\nabla C_1 + D_{1T}\nabla T, \tag{2a}$$

$$-J_T = D_{T1}\nabla C_1 + D_{TT}\nabla T, \tag{2b}$$

for coupled heat and mass transfer. Any factors arising from expressing the temperature driving force as ∇T instead of $\nabla \ln T$ are included in the diffusivity. In matrix and vector form these may be written as

$$-J = D\nabla C, \tag{3}$$

where the vector and matrix notation refer to the types of fluxes and forces, and have nothing to do with spatial vectors or tensors. In Eq. 3, if coupled heat and mass transport is involved, the second component involves temperature. The notation of Eq. 3 will be used in all further developments.

For coupled transport in a boundary layer on a flat plate, the equation in vector form is

$$v_x \frac{\partial C}{\partial x} + v_y \frac{\partial C}{\partial y} = D \frac{\partial^2 C}{\partial y^2}, \tag{4}$$

which represents two differential equations describing the boundary layer transport. Of course, the velocity equation still applies to the hydrodynamic boundary layer; however, no coupling

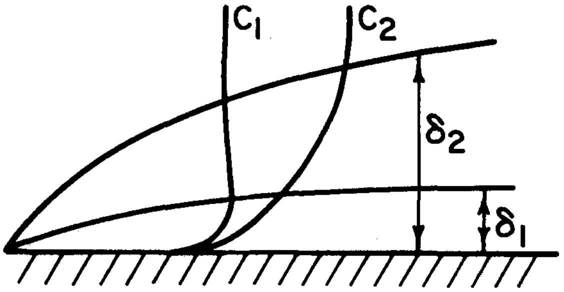

Figure 1. Boundary layer ordering for uncoupled transport. Species 1 is the particle and species 2 is either the small solute or temperature.

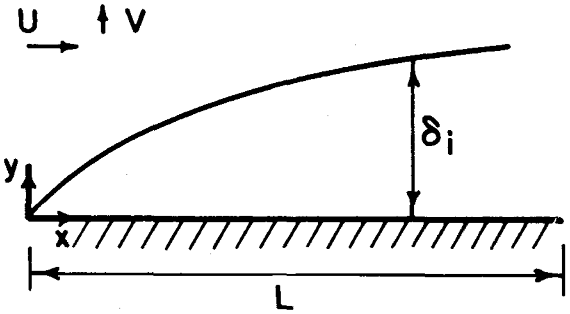

Figure 2. One single boundary layer for coupled transport with scaling factors indicated.

between momentum transport and either heat or mass transport is considered here, although it would certainly be possible to do so.

Since the coupling in Eq. 4 is linear, it has been shown (Toor, 1964; Cussler, 1976) that by introducing the transformation $\psi = t^{-1}C$, where t is the eigenvector (modal) matrix of D, reduces Eq. 4 into

$$v_x \frac{\partial \psi}{\partial x} + v_y \frac{\partial \psi}{\partial y} = \sigma \frac{\partial^2 \psi}{\partial y^2}, \tag{5}$$

which represents two uncoupled equations in ψ since σ is the eigenvalue matrix of D, and is, of course, diagonal. These eigenvalues are

$$\sigma = \frac{1}{2}[(D_{11} + D_{22}) \pm [(D_{11} + D_{22})^2 - 4(D_{11}D_{22} - D_{12}D_{21})]^{1/2}], \tag{6}$$

or in vector form

$$\sigma = \frac{1}{2}[tr\,\boldsymbol{D} \pm [(tr\,\boldsymbol{D})^2 - 4|\mathbf{D}|]^{1/2}]. \tag{7}$$

BOUNDARY LAYER ORDERING

A major question in coupled transport situations such as these is whether two distinct boundary layer thicknesses are necessary to fully characterize the system. This is especially important in the application to diffusiophoresis and thermophoresis, because, in the uncoupled case, the boundary layers would differ by at least one order of magnitude. In diffusiophoresis, the bulk of the work thus far has involved particles in liquids migrating in electrolyte gradients. Considering that a 1,000 Å particle will have a Stokes-Einstein diffusivity of 2.2×10^{-8} cm^2/s in water at room temperature, and that a typical electrolyte diffusivity is on the order of 10^{-5} cm^2/s, the particle's boundary layer might be expected to be 1.5 orders-of-magnitude smaller than that of the electrolyte. Similarly, for thermophoresis of particles, since $\alpha_{\text{air}} = 7.4 \times 10^{-1}$ cm^2/s, $\alpha_{H_2O} = 5.6 \times 10^{-3}$ cm^2/s, and the Stokes-Einstein diffusivity of a 1,000 Å particle in air at room temperature is 1.1×10^{-6} cm^2/s, the thermal boundary layer would be expected to be about two orders of magnitude larger than the diffusion boundary layer in either case. In these calculations the square root relationship between the ratio of boundary layer thicknesses and the ratio of diffusivities was used.

The results of this uncoupled boundary layer ordering are illustrated in Figure 1. However, if this figure and the "long" version of Eq. 4,

$$v_x \frac{\partial C_1}{\partial x} + v_y \frac{\partial C_1}{\partial y} = D_{11} \frac{\partial^2 C_1}{\partial y^2} + D_{12} \frac{\partial^2 C_2}{\partial y^2}, \tag{8a}$$

$$v_x \frac{\partial C_2}{\partial x} + v_y \frac{\partial C_2}{\partial y} = D_{21} \frac{\partial^2 C_1}{\partial y^2} + D_{22} \frac{\partial^2 C_2}{\partial y^2}, \tag{8b}$$

are examined, an intuitive inconsistency arises. Within δ_1, the profile of 2 (either concentration or temperature) will be approximately linear, since $\delta_1 \ll \delta_2$. This means that $\delta C_2/\partial y$ is constant in this region and $\partial^2 C_2/\partial y^2 = 0$ in this region. This *uncouples the equation for C_1 from that for C_2*. Outside δ_1, but within δ_2, the profile of 1 has reached its bulk value and is flat, while the profile of 2 is still changing. This means that $\partial^2 C_1/\partial y^2$ will be zero in this region and the equation for C_2 is uncoupled from that for C_1. This is, of course, impossible if Eqs. 8 are to be describing migration of species 1 in a gradient of 2.

Another intuitive argument may also be made to support this idea. Outside δ_1, if the transport is coupled, 1 will migrate due to the gradient of 2. However, this migration must then cause a gradient of 1 in this region, which actually would then cause a diffusive migration of 1 in its own gradient; although, the argument could be made that as species 1 migrates due to the gradient of 2, it is immediately replenished by some 1 from the bulk, maintaining the flat profile. Therefore, it appears that allowing the boundary layer to differ in magnitude is intuitively incorrect.

Now, let us proceed to nondimensionalize Eqs. 8 (Figure 2). The velocity scaling factors will be U for v_x and V for v_y, and the length scales will be L for x and δ_1 or δ_2 for y. The dimensionless concentration will be $\theta = (C - C_\infty)/(C_w - C_\infty)$, and the result from ordering the continuity equation, $V = U\delta_H/L$ (Denn, 1980), will be used. Equation 8a becomes

$$\left(\frac{\Delta C_1 U}{L}\right) v_x^* \frac{\partial \theta_1}{\partial x^*} + \left(\frac{\Delta C_1 U \delta_H}{L \delta_1}\right) v_y^* \frac{\partial \theta_1}{\partial y^*} = \left(\frac{D_{11}\Delta C_1}{\delta_1^2}\right) \frac{\partial^2 \theta_1}{\partial y^{*2}} + \left(\frac{D_{12}\Delta C_2}{\delta_2^2}\right) \frac{\partial^2 \theta_2}{\partial y^{*2}}, \tag{9}$$

which rearranges into

$$v_x^* \frac{\partial \theta_1}{\partial x^*} + \left(\frac{\delta_H}{\delta_1}\right) v_y^* \frac{\partial \theta_1}{\partial y^*} = \left(\frac{LD_{11}}{U\delta_1^2}\right) \left[\frac{\partial^2 \theta_1}{\partial y^{*2}} + \left(\frac{D_{12}\Delta C_2 \delta_1^2}{D_{11}\Delta C_1 \delta_2^2}\right) \frac{\partial^2 \theta_2}{\partial y^{*2}}\right]. \tag{10}$$

The lead term on the righthand side of Eq. 10 is just $(L^2/\delta_1{}^2)(1/ReSc_{11})$, where the Schmidt number is defined based on D_{11}. Similar nondimensionalization of Eq. 8b yields

$$v_x^* \frac{\partial \theta_2}{\partial x^*} + \left(\frac{\delta_H}{\delta_2}\right) v_y^* \frac{\partial \theta_2}{\partial y^*} = \left(\frac{L^2}{\delta_2^2 ReSc_{22}}\right) \left[\left(\frac{D_{21}\Delta C_1 \delta_2^2}{D_{22}\Delta C_2 \delta_1^2}\right) \frac{\partial^2 \theta_1}{\partial y^{*2}} + \frac{\partial^2 \theta_2}{\partial y^{*2}}\right]. \tag{11}$$

Therefore, for a physical situation to be coupled in such a way as to require this type of description,

$$\frac{D_{12}\Delta C_2 \delta_1^2}{D_{11}\Delta C_1 \delta_2^2} \sim 1 \quad \text{and} \quad \frac{D_{21}\Delta C_1 \delta_2^2}{D_{22}\Delta C_2 \delta_1^2} \sim 1. \tag{12}$$

If these are less than order unity, then the coupling vanishes, and if these are greater than order unity, the "main" term transport may be negligible, but, as will be discussed later, this type of description is still needed to completely characterize the system. Rearrangement of Eq. 12 suggests that

$$\frac{\delta_1}{\delta_2} = \left(\frac{D_{11}\Delta C_1}{D_{12}\Delta C_2}\right)^{1/2} = \left(\frac{D_{21}\Delta C_1}{D_{22}\Delta C_2}\right)^{1/2}, \tag{13}$$

which means that $D_{11}D_{22} = D_{12}D_{21}$, or at least that these two products are of the same order of magnitude.

Equation 12 suggests several components to the criterion for a system to be coupled. One is that D_{12}/D_{11} be of order unity or larger. This is a standard criterion for the importance of ternary diffusion effects (Cussler, 1976). Another criterion is that $\Delta C_2/\Delta C_1$ be of order unity or larger. Many gas systems and macromolecular-solute-solvent systems exhibit ternary diffusion effects for this reason (Cussler, 1976). However, the ratio of δ_1/δ_2 is not a criterion for the onset of ternary diffusion. One reason was explained previously, that of the intuitive inconsistency occurring outside the thin boundary layer and within the large boundary layer. A second reason comes from Eq. 12. In cases where the coupling is between small solutes or electrolytes, the boundary layer thicknesses based upon the binary diffusivities will be of the same order of magnitude, so these cases are not of interest in the present argument. However, the cases involving coupling between particles and small electrolytes, or between particles and temperature are the ones for which this argument is necessary. Since a particle's binary diffusivity will be much smaller than that of a small solute, then, based upon binary diffusivities, $\delta_1 \ll \delta_2$. As shown earlier, for a 1,000 Å particle $\delta_1^2 \sim 10^{-3}\,\delta_2^2$, in the presence of a small solute, and $\delta_1^2 \sim 10^{-4}\,\delta_2^2$, in the presence of a temperature gradient. Examination of Eq. 12 would then suggest that for $\Delta C_1 \sim \Delta C_2$ and $D_{11} \sim D_{12}$, which Eq. 13 shows also implies $D_{21} \sim D_{22}$ regardless of the relationship between ΔC_1 and ΔC_2, that the equation for species 1 would be uncoupled, but that the equation for species 2 would remain highly coupled, perhaps to the extent of making the diagonal term for this species negligible. On the other hand, if $\delta_1 \sim \delta_2$, then Eq. 12 suggests that the only criterion for the onset of coupling would involve the relative values of diffusivities and concentration (or temperature) gradients, which makes more sense. It is also true that while D_{11} and D_{22} are generally assumed to be very close to the binary diffusivities, this might not always be true, and if D_{11} and D_{22} approached the same order of magnitude, so would δ_1 and δ_2, since, in Eqs. 10 and 11, δ_1 and δ_2 are related to D_{11} and D_{22}, and not the binary diffusivities.

In Eqs. 9, 10 and 11, which were non-dimensionalized based upon the *a priori* assumption that there are two boundary layer thicknesses, one for each species, the length term was always scaled relative to the thickness associated with the dependent variables in the same derivative term. However, an argument could possibly be made to scale the length relative to the thickness associated with the prevalent dependent variable described by the entire equation; that is, δ_1 for the entirety of Eq. 10 and δ_2 for the entirety of Eq. 11. If this were true, then much of the argument following Eq. 12 would be unnecessary, and the second equality in Eq. 13 would follow immediately as a criterion for coupled transport. The overriding argument for one characteristic thickness would not change.

Equation 5 may be made dimensionless, just like the coupled equations. The result is

$$v_x^* \frac{\partial \phi_i}{\partial x^*} + \left(\frac{\delta_H}{\delta_i}\right) v_y^* \frac{\partial \phi_i}{\partial y^*} = \left(\frac{L^2}{\delta_i^2 Re Sc_i}\right) \frac{\partial^2 \phi_i}{\partial y^2}, \tag{14}$$

where ϕ_i is a dimensionless ψ_i, the subscript refers to the two uncoupled equations, and Sc_i is defined as the ratio of the kinematic viscosity to the ith eigenvalue of the diffusivity matrix. If D_{11} and D_{22} are of the same order of magnitude, so will the eigenvalues and there will be no argument about the relative boundary layer thicknesses. If $D_{11} \ll D_{22}$ as in the cases under discussion, close examination of Eq. 6, plus the criterion following Eq. 13 would suggest that $\sigma_1 \ll \sigma_2$. While this analysis would appear to contradict the previous argument in favor of equal boundary layer thicknesses, the fact that the concentration differences between bulk and surface, ΔC_1 and ΔC_2, are not present suggests that no conclusions should be drawn from dimensional analysis of the transformed equation. After all, ϕ_i and σ_i are present as a mathematical convenience, and have no real physical significance. Indeed, the same argument has been used in favor of the Stefan-Maxwell formulation (Taylor, 1982); although if the equations are inverse transformed after a solution is found, the physical significance returns.

Another possibility which must be acknowledged, and was suggested by Eq. 14 and the subsequent discussion, is that there are two distinct boundary layers which become equal in magnitude when coupling, as described by Eq. 12, becomes important. If the coupling were less important, the two boundary layers would differ by less than the uncoupled case, and a continuum of δ_2 values would exist describing the relative importance of the "cross" terms. However, the analysis which follows does not support this concept.

The previous analysis assumes that all of the diffusivities are constant. Even though this is not always true, the usual procedure is to evaluate the diffusivity at an average condition and use the result of the constant diffusivity solution. For purposes of conceptualization, it is necessary to include any variation of the diffusivities with concentration in the ordering analysis. For diffusiophoresis and thermophoresis, $D_{12} = KC_1$, since it is necessary to multiply the velocity by the particle concentration to get a flux. Equation 8a then becomes

$$v_x \frac{\partial C_1}{\partial x} + v_y \frac{\partial C_1}{\partial y} = D_{11} \frac{\partial^2 C_1}{\partial y^2} + K \frac{\partial C_1}{\partial y} \frac{\partial C_2}{\partial y} + D_{12} \frac{\partial^2 C_2}{\partial y^2}\,. \tag{15}$$

One of the intuitive arguments presented previously now fails because for $y < \delta_1, \partial C_2/\partial y$ is constant, the second term on the right hand side of Eq. 15 remains indicating that coupling does not vanish entirely. However, subsequent dimensional analysis yields the following equation in place of Eq. 10:

$$v_x^* \frac{\partial \theta_1}{\partial x^*} + \left(\frac{\delta_H}{\delta_1}\right) v_y^* \frac{\partial \theta_1}{\partial y^*} = \frac{LD_{11}}{U\delta_1^2}\left[\frac{\partial^2 \theta_1}{\partial y^{*2}} + \left(\frac{D_{12}\Delta C_2 \delta_1^2}{D_{11}\Delta C_1 \delta_2^2}\right)\frac{\partial^2 \theta_2}{\partial y^{*2}} + \left(\frac{D_{12}\Delta C_2 \delta_1}{D_{11} C_1 \delta_2}\right)\left(\frac{\partial \theta_1}{\partial y^{*2}}\right)\left(\frac{\partial \theta_2}{\partial y^{*2}}\right)\right]\,. \tag{16}$$

In order for the third term in Eq. 16 to be important

$$\frac{D_{12}\Delta C_2 \delta_1}{D_{11} C_1 \delta_2} \sim 1 \tag{17}$$

Comparison of Eqs. 13 and 17 yields

$$\frac{\delta_1}{\delta_2} = \left(\frac{D_{11}\Delta C_1}{D_{12}\Delta C_2}\right)^{1/2} = \frac{D_{11}C_1}{D_{12}\Delta C_2}\,. \tag{18}$$

Since the case of $\Delta C_1 \sim C_1$ cannot be ruled out *a priori*, as it is the important case of the surface at a steady state zero concentration, the only situation for which both criteria in Eq. 18 can be satisfied as $\delta_1 = \delta_2$. If the surface concentration is on the order of the bulk concentration $\Delta C_1 \sim C_1$, as long as the surface and bulk concentrations do not become close enough such that $\Delta C_1 \ll C_1$. Either way, the result with practical significance is the same as with no concentration dependence of D_{12}.

MULTICOMPONENT INTERFACIAL THEORIES

There are three theories most often cited to describe transport near interfaces: the penetration theory, the film theory and boundary layer theory. Although there are others such as the surface renewal theory and some combining features of several of these, these three theories will be sufficient to support the argument for one boundary layer thickness. Since analytical solutions are only available for the case of all constant diffusivities. only these results will be presented.

Film Theory

The film theory is the stagnant layer model for transport near an interface. The equation for the coupled case is

$$0 = D\frac{d^2 C}{dy^2}, \tag{19}$$

with boundary conditions

$$y = 0 \quad C = C_w, \tag{20a}$$

$$y = \delta \quad C = C_\infty. \tag{20b}$$

The solution for the concentration profile is

$$\left[\frac{C - C_w}{C_\infty - C_w}\right] = \left[\frac{y}{\delta}\right], \tag{21}$$

and for the flux is

$$-J = D\left[\frac{C_\infty - C_w}{\delta}\right]. \tag{22}$$

Equation 22 represents two equations

$$-J_1 = \frac{D_{11}}{\delta_1}(C_{1\infty} - C_{1w}) + \frac{D_{12}}{\delta_2}(C_{2\infty} - C_{2w}), \tag{23a}$$

$$-J_2 = \frac{D_{21}}{\delta_1}(C_{1\infty} - C_{1w}) + \frac{D_{22}}{\delta_2}(C_{2\infty} - C_{2w}). \tag{23b}$$

In the binary case, the film thickness, an empirical parameter, is automatically set by knowledge of the flux, the concentration difference and the diffusivity. In the coupled case, if the film thicknesses are treated as empirical parameters, Eqs. 23 represent two equations in two unknowns for these parameters. If the argument, made previously, that δ_1/δ_2 cannot be a criterion for coupled transport is valid, then Eq. 12 suggests that the δ values in Eqs. 23 will be similar in magnitude. However, extreme care must be used in interpreting the results of a coupled film theory, since the nature of the coupling is different from all other situations in that the concentration profiles do not involve the diffusivities and the eigenvalues of the diffusivity matrix. As will be shown, the film theory is quite useful when used in conjunction with boundary layer theory to estimate boundary layer thicknesses.

Boundary Layer Theory

The equation for this more rigorous theory of interface transport is Eq. 4. Typical boundary conditions are

$$x = 0 \quad C = C_\infty, \tag{24a}$$

$$y = \infty \quad C = C_\infty, \tag{24b}$$

$$y = 0 \quad C = C_w. \tag{24c}$$

For diffusion and convection in high Schmidt number in liquids, Lighthill (1950) has shown that a similarity solution exists to the uncoupled problem as

$$\frac{C - C_w}{C_\infty - C_w} = \frac{1}{\Gamma(4/3)}\int_0^\eta e^{-s^3}\,ds, \tag{25}$$

$$\eta = \frac{y\sqrt{B}}{\left[9D\int_o^x \sqrt{B}\,dx\right]^{1/3}}. \tag{26}$$

where B = $(\partial v_x/\partial y)_{y=0}$. The mass transfer coefficient is

$$k = \frac{-D\sqrt{B}}{\Gamma\left(\frac{4}{3}\right)\left[9D\int_o^x \sqrt{B}\,dx\right]^{1/3}}. \tag{27}$$

The solution for the flux in the coupled problem follows immediately (Cussler, 1976) as

$$\begin{aligned} J_1 = {} & \left(\frac{D_{11} - \sigma_2}{\sigma_1 - \sigma_2}\right)k(\sigma_1)(C_{1\infty} - C_{1w}) \\ & + \left(\frac{D_{11} - \sigma_1}{\sigma_2 - \sigma_1}\right)k(\sigma_2)(C_{1\infty} - C_{1w}) \\ & + \left(\frac{D_{12}}{\sigma_1 - \sigma_2}\right)k(\sigma_1)(C_{2\infty} - C_{2w}) \\ & + \left(\frac{D_{12}}{\sigma_2 - \sigma_1}\right)k(\sigma_2)(C_{2\infty} - C_{2w}), \end{aligned} \tag{28}$$

where $k(\sigma_i)$ is Eq. 27 with the indicated eigenvalue substituted for D. The similar equation for J_2 is

$$\begin{aligned} J_2 = {} & \left(\frac{D_{21}}{\sigma_1 - \sigma_2}\right)k(\sigma_1)(C_{1\infty} - C_{1w}) \\ & + \left(\frac{D_{21}}{\sigma_2 - \sigma_1}\right)k(\sigma_2)(C_{1\infty} - C_{1w}) \\ & + \left(\frac{D_{22} - \sigma_2}{\sigma_1 - \sigma_2}\right)k(\sigma_1)(C_{2\infty} - C_{2w}) \\ & + \left(\frac{D_{22} - \sigma_1}{\sigma_2 - \sigma_1}\right)k(\sigma_2)(C_{2\infty} - C_{2w}). \end{aligned} \tag{29}$$

Following the method suggested by Levich (1962), the boundary layer thickness may be estimated by equating the flux in Eqs. 28 and 29 to a film theory result, Eqs. 23. Equating terms multiplied by the same concentration difference, and realizing for most situations that all $k(\sigma_i)$ are the same constant (call it β) multiplied by $\sigma_i^{2/3}$, yields

$$\frac{D_{11}}{\delta_1} = \beta\left[\left(\frac{D_{11} - \sigma_2}{\sigma_1 - \sigma_2}\right)\sigma_1^{2/3} + \left(\frac{D_{11} - \sigma_1}{\sigma_2 - \sigma_1}\right)\sigma_2^{2/3}\right], \tag{30}$$

$$\frac{D_{12}}{\delta_2} = \beta D_{12}\left[\frac{\sigma_1^{2/3}}{\sigma_1 - \sigma_2} + \frac{\sigma_2^{2/3}}{\sigma_2 - \sigma_1}\right], \tag{31}$$

$$\frac{D_{21}}{\delta_1} = \beta D_{21}\left[\frac{\sigma_1^{2/3}}{\sigma_1 - \sigma_2} + \frac{\sigma_2^{2/3}}{\sigma_2 - \sigma_1}\right], \tag{32}$$

$$\frac{D_{22}}{\delta_2} = \beta\left[\left(\frac{D_{22} - \sigma_2}{\sigma_1 - \sigma_2}\right)\sigma_1^{2/3} + \left(\frac{D_{22} - \sigma_1}{\sigma_2 - \sigma_1}\right)\sigma_2^{2/3}\right]. \tag{33}$$

Recall that the case of interest is probably describable by D_{11} and D_{22} being of very different orders-of-magnitude. Examination of Eqs. 6 and 7 indicates that the two eigenvalues will be of different orders-of-magnitude. This can be confirmed by assigning $D_{11} = 1$, $D_{22} = 100$, and then choosing $D_{12}D_{21} = 90$. This latter choice insures a positive diffusivity determinant and positive eigenvalues since $D_{11}, D_{22} > 0$ always, and is consistent with the criterion following Eq. 13. The resulting eigenvalues are $\sigma_1 = 0.099$, $\sigma_2 = 100.9$. Equations 30 and 33 do not provide any conclusive information for the following two reasons. First of all, $D_{22} - \sigma_2$ can take on any value less than or equal to 100; and secondly, since only a very slight change in numbers could make $\sigma_1 \sim 1$, the same ambiguity would hold for $D_{11} - \sigma_1$. However, examination of Eqs. 31 and 32 demonstrate that regardless of the values of the parameters, δ_1 and δ_2 have identical expressions. Furthermore, $\sigma_1^{2/3} \sim 0.2$ and $\sigma_2^{2/3} \sim 20$, and since $\sigma_2 - \sigma_1 \sim \sigma_2$, then δ_1 and $\delta_2 \sim \sigma_2^{2/3}$, and it will be the larger eigenvalue, due to the component with the larger diffusivity, which again determines that the larger boundary layer thickness applies to both species.

A similar argument would apply to thermophoresis with one important exception. If only a diffusion equation with an extra term for the thermal diffusion effect were used, it would be tempting to apply the same high Schmidt number analysis in liquid systems. However, since all the analyses presented indicate that the larger boundary layer thickness controls, then in thermal diffusion it will be the thermal boundary layer in control, which is not much smaller than the hydrodynamic boundary layer. This is no problem since straightforward boundary layer solutions do exist for the case of a thermal and hydrodynamic boundary layer of similar magnitude; but, great care must be taken in deciding what type of approximation applies to a coupled transport situation.

Penetration Theory

The penetration theory is essentially an unsteady state model for the development of a boundary layer. Although more often associated with situations involving short exposure times, it should yield the same conceptual result as boundary layer theory given that the concept is correct. The equation for the coupled case is

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial y^2}, \tag{34}$$

with boundary conditions

$$C = C_\infty \quad t \le 0 \quad \forall y, \tag{35a}$$

$$C = C_\infty \quad \forall t > 0 \quad y \to \infty, \tag{35b}$$

$$C = C_w \quad \forall t > 0 \quad y = 0. \tag{35c}$$

The results have been given elsewhere (Cussler, 1976) as

$$\frac{C_1 - C_{1w}}{C_{1\infty} - C_{1w}} = \left[\frac{(D_{11} - \sigma_2) + D_{12}\left(\dfrac{C_{2\infty} - C_{2w}}{C_{1\infty} - C_{1w}}\right)}{\sigma_1 - \sigma_2}\right] \operatorname{erf}\left(\frac{y}{(4\sigma_1 t)^{1/2}}\right) + \left[\frac{(D_{11} - \sigma_1) + D_{12}\left(\dfrac{C_{2\infty} - C_{2w}}{C_{1\infty} - C_{1w}}\right)}{\sigma_2 - \sigma_1}\right] \operatorname{erf}\left(\frac{y}{(4\sigma_2 t)^{1/2}}\right), \tag{36}$$

$$\frac{C_2 - C_{2w}}{C_{2\infty} - C_{2w}} = \left[\frac{D_{21}\left(\dfrac{C_{1\infty} - C_{1w}}{C_{2\infty} - C_{2w}}\right) + (D_{22} - \sigma_2)}{\sigma_1 - \sigma_2}\right] \operatorname{erf}\left(\frac{y}{(4\sigma_1 t)^{1/2}}\right) + \left[\frac{D_{21}\left(\dfrac{C_{1\infty} - C_{1w}}{C_{2\infty} - C_{2w}}\right) + (D_{22} - \sigma_1)}{\sigma_2 - \sigma_1}\right] \operatorname{erf}\left(\frac{y}{(4\sigma_2 t)^{1/2}}\right). \tag{37}$$

In the binary case, it is a simple matter to define the penetration thickness, $y = \delta$, at $(C - C_w)/(C_\infty - C_w) = 0.99$, and obtain the proportionality $\delta = 4(Dt)^{1/2}$ (since erf $2 \sim 0.99$). In the coupled case, defining such a relationship is just as simple. In Eqs. 36 and 37, the arguments of both erf terms must be large enough for the right hand side to go to one. This can only occur, for a given time, when y is large enough. Since one of the eigenvalues will be much larger than the other, both dimensionless concentrations do not approach unity until y is large enough relative to the larger eigenvalue. Therefore,the concentrations of both species reach the "bulk" value at the same penetration thickness, which scales with the larger eigenvalue. This conclusions supports the intuitive and boundary layer arguments presented previously that the larger of the uncoupled boundary layer thicknesses control the coupled case.

IDEMPOTENT METHOD

The entire analysis presented previously employs the modal matrix method of solving the linearized multicomponent transport equations. Stewart and Prober (1964) have presented an alternative analysis involving the idempotent elements of their matrix $\dot{A}$, which is the inverse of the Fickian diffusivity matrix D. Their Eq. 42 gives the expression for the concentration profiles, which rewritten in the present nomenclature is

$$[C - C_w] = \sum_{r=1}^{s} \phi_r E_r [C_\infty - C_w], \tag{38}$$

where

$$E_r = \prod_{\substack{\lambda_j=\lambda_1 \\ \lambda_j \ne \lambda_r}}^{\lambda_j=\lambda_s} \frac{[A - \lambda_j I]}{\lambda_r - \lambda_j}, \; r = 1, \ldots, s; \quad s > 1. \tag{39}$$

The values of λ are the eigenvalues of A, s is the number of distinct eigenvalues of A, and if $s = 1$, $E_1 = I$. For ternary diffusion, $s = 2$, and the right hand side of Eq. 39 only involves one matrix. Therefore, none of the elements in the vectors $E_r[C_\infty - C_w]$ can ever be zero (or negligible) in Eq. 38. However, if $s > 2$, the righthand side of Eq. 39 involves products of matrices, and, depending on the values of the elements of A, some elements of $E_r[C_\infty - C_w]$ could be zero (or negligible). If this were true for the terms representing the smallest eigenvalues of A, which correspond to the largest eigenvalues of D and hence the largest penetration distance, these elements would be associated with thinner boundary layers. For ternary diffusion; that is, $s = 2$, the result is universal and is in agreement with the previous analysis presented in this paper. For $s > 2$, the result is not universal, and depends upon the specific values of the diffusivities involved. The exact physical situation for which all of the boundary layers will not be of the same magnitude can only be found by specific numerical calculations.

DISCUSSION

The analysis presented in this paper is rigorously limited to situations involving constant diffusivities, although, as has been mentioned, excellent results have been obtained in the past by evaluating these at average values and assuming these to be constant. However, as evidenced by the result of the dimensional analysis, the qualitiative conclusion appears to be universal, and it is therefore reasonable to discuss implications in terms of diffusiophoresis and thermophoresis.

In a recent report detailing diffusiophoretic particle deposition on a membrane from which electrolyte was diffusing (Lin and Prieve, 1983) it was noted that the electrolyte film thickness was an important factor. Since the present argument suggests that the same film thickness applies to the particle, this is no longer a surprising result. Since diffusiophoretic deposition, although recognized for many years as a significant phenomenon, is a relatively new idea with respect to coating by latex paints and enhanced particle capture in granular media filtration, a major significance of the multicomponent diffusion model is the ease in describing any deposition geometry; only the solution to the binary case and the diffusivity matrix need be known (Cussler, 1976), the latter having been shown to be measurable by standard ternary diffusion techniques (Shaeiwitz and Lechnick, 1984). Diffusiophoresis has also been recognized as a significant mechanism in wet scrubbers, Pilat and Prem (1976; 1977) and the present model should allow more rigorous treatment of this physical situation.

Many previous descriptions of both diffusiophoresis and thermophoresis of particles have only involved consideration of the migration velocity of the particle (Rosner, 1980; Whitmore and Meisen, 1977; McNab and Meisen, 1973; Walker et al., 1979). However, examination of Eqs. 28, 29, 36 and 37 shows that even though D_{12} may be much greater than D_{11}, as would probably be true in the case of a very large ($>1\mu$m) particle, that all four diffusivities still contribute to the solution. Of special importance is D_{22}, which is major factor in determining σ_2, which has been shown to be the most important parameter in specifying the system. Therefore, the models for thermal diffusion which only consider the particle continuity equation should not be considered rigorously correct, although their solution may yield valuable qualitative and possibly quantitative information. It is also true that the present solution technique is much simpler than one such model (Rosner, 1980), assuming all diffusivities are known, and this model of thermophoresis also neglects the diffusion boundary layer of the particle, which has been shown here to be fundamentally incorrect. Again, these models need not be discarded totally, since they may be valuable empiricisms. Another author has pointed out that there are few models of thermophoretic deposition in useful flow geometries (Goren, 1977). Again, an advantage of the present formulation is that given that all the diffusivities are known, and this solution exists for any physical situation for which a binary mass or heat transfer solution exists.

Based on previous work using the Stefan-Maxwell equations to model thermal diffusion (Taylor, 1982; Standart et al., 1979), it is not clear that the Fickian description is any better (or worse). In gas systems especially, a Stefan-Maxwell treatment may be easier since it relies upon knowledge of the individual binary diffusivities. However, it is also true that binary particle diffusivities are not easily obtainable, especially in concentrated systems where particle-particle hydrodynamic and possibly electrostatic interactions are involved. This is definitely true in liquid systems, where, as has already been stated, the binary diffusivities in the Stefan-Maxwell equation do not really have the same significance as in gas systems. The advantage of the linearized Fickian treatment is the existence of an immediate solution from the binary result. Of course, this advantage would vanish if the diffusivities were too difficult to evaluate or measure.

Finally, it is important to reiterate one major significance of the fact that the larger boundary layer characterizes the entire coupled system. This is that useful approximate solutions based on very large or very small Prandtl or Schmidt numbers must be used with care. For example, particle thermophoresis in liquids may not be modeled by the large Schmidt number approximation commonly applied to diffusion in liquids.

ACKNOWLEDGMENT

The author gratefully acknowledges helpful discussions with J. J. L. Higdon as well as financial support from grant NSF CPE 80-12431. W. E. Stewart suggested the analysis involving the idempotent matrices.

NOTATION

A = elements of inverse of diffusivity matrix
B = $(\partial v_x/\partial y)_{y=0}$
C = concentration
D,D_{ij} = diffusion coefficient, if subscripted, i = species, j = gradient of species causing diffusion
E = principal idempotent matrices of A
J = flux
K = proportionality constant between D_{12} and C_1
k = mass transfer coefficient
L = length scale in direction of flow for boundary layer ordering
Re = Reynolds number
Sc = Schmidt number
t = if a matrix, modal matrix of diffusivity matrix if a scalar, time
T = temperature
v_x,v_y = velocity components
U,V = scaling factors for x and y velocity components
x = direction with flow
y = direction normal to flow
β = proportionality factor between mass transfer coefficient and diffusivity to the ⅔ power
λ = eigenvalues of A matrix
σ = boundary layer or film thickness
η = defined in Eq. 22
σ = eigenvalue of diffusivity matrix
ϕ = dimensionless ψ
ψ = transformed concentration $\psi = t^{-1}C$
θ = dimensionless concentration

Subscripts

1 = species 1, the particle
2 = species 2, the electrolyte or small solute
H = refers to hydrodynamic boundary layer
w = refers to wall or surface concentration
T = thermal, refers to a diffusivity
∞ = refers to bulk value of concentration

Superscript

* = dimensionless quantity

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Manuscript received January 20, 1983; revision received July 11, and accepted July 14, 1983.