

# Simultaneous Heat and Mass Transfer in Free Convection Boundary Layers

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Expressions are derived for the heat and mass transfer coefficients for laminar free convection driven by simultaneous differences in temperature and composition for the asymptotic cases of equal Schmidt and Prandtl numbers approaching zero, equal Schmidt and Prandtl numbers approaching infinity, Schmidt number approaching infinity and Prandtl number approaching zero, and Schmidt number larger than Prandtl number and Prandtl number approaching infinity.

The results are applicable for horizontal cylinders or vertical axisymmetric bodies with arbitrary body contours insofar as the approximations of boundary-layer theory are valid. The results compare favorably with existing solutions and experimental results for particular conditions.

In either forced or free convection, the phenomena of heat and mass transfer are analogous, providing, of course, that effects due to variations in physical properties or interfacial velocity are negligible. For heat and mass transfer in forced convection, the flow field is known a priori. By contrast, in free convection the flow field is generated by and hence coupled with both the temperature and concentration fields. Nevertheless, analytical expressions describing the rates of transport with coupling can be developed in several instances.

The analyses herein are restricted to binary mixtures and laminar boundary-layer flows around submerged objects whose outer surfaces are maintained at a uniform temperature and composition. Physical properties are considered to be constant, except for the density term that is associated with the body force. Secondary phenomena, such as thermal diffusion (13), interfacial velocity (1), and diffusing heat capacities, that is, the Ackerman effect as described by Merk (7), are not considered. Hence the results describe only the gross features of the consequences of interaction. Notwithstanding these idealizations and restrictions, results of considerable generality and applicability are obtained.

The relatively simple situation which occurs when the Schmidt and Prandtl numbers are equal (an approximation for gaseous mixtures) is first treated in some detail. Next, the case of a small Prandtl number but a large Schmidt number (corresponding to liquid metals) is considered. After this, a model for large Prandtl and Schmidt numbers (representing the behavior of ordinary liquids) is analyzed.

The analyses are based primarily on asymptotic methods such as those described by Meksyn (6) and van Dyke (15). The results are, as a consequence, applicable to a wider variety of fluids than the results of Somers (12). In his study, the von Karman integral method was employed, and the results are restricted to the case of equal or almost equal Schmidt and Prandtl numbers. Asymptotic methods are used herein for a number of reasons: first, they are relatively easy to apply and the resulting analytic forms are readily interpreted; second, they can usually be improved in a rational manner; third, their accuracy is frequently adequate; and fourth, intermediate cases can often be approximated by interpolation.

The analysis starts with the generalized description of free convection previously presented by Saville and Churchill (10) in which the dependent variables are represented by rapidly converging series which are universal with respect to body contours within a specified class of body shapes. Only the highlights of the subsequent derivations are presented herein. Further details are given by Saville (9).

## BASIC EQUATIONS AND RELATIONSHIPS

The objective is to relate the rates of transfer of momentum, heat and mass to the transport properties of the fluid, the body force, and the geometrical form of the submerged object. These relations express the Nusselt, Sherwood, and shear stress numbers as functions of the Prandtl, Schmidt, and Grashof numbers and a ratio of driving forces.

The starting point is the familiar boundary-layer description. The dependent variables are then expanded according to the procedure described by Saville and Churchill (10). This technique results in rapidly converging series in many instances. For horizontal cylinders or spheres (objects which do not admit similarity transformations) one-term approximations were found to be adequate for most purposes, even for low Prandtl numbers. For objects which do admit similarity transformations, the technique yields the proper transformation directly.

The one-term approximations for the local shear stress, Nusselt, and Sherwood numbers, as given by Saville and Churchill (10), are:

$$N_T N_{Gr}^{-1/4} \sim \phi_1(x) [\xi(x)]^{1/4} F''(0) \quad (1)$$

$$N_{Nu} N_{Gr}^{-1/4} \sim \phi_2(x) [\xi(x)]^{-1/4} [-T'(0)] \quad (2)$$

$$N_{Sh} N_{Gr}^{-1/4} \sim \phi_2(x) [\xi(x)]^{-1/4} [-M'(0)] \quad (3)$$

where

$$\phi_1(x) = \left[ \frac{4}{3} \right]^{1/4} [r(x)]^\delta \left( \frac{d\xi}{dx} \right)^2 \quad (4)$$

$$\phi_2(x) = \left[ \frac{4}{3} \right]^{-1/4} \frac{d\xi}{dx} \quad (5)$$

$$\xi(x) = \int_0^x [S(z)]^{1/3} [r(z)]^{-2\delta/3} dz \quad (6)$$

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with  $\delta = 0$  for planar flows and  $\delta = 1$  for axisymmetric flows. The numbers  $F''(0)$ ,  $T'(0)$ , and  $M'(0)$  are found by solving the following equations, derived from the laws governing the conservation of momentum, energy, and mass (9):

$$F''' + FF'' - K_0 F'F' + T + \sigma M = 0 \quad (7)$$

$$T'' + N_{Pr} F T' = 0 \quad (8)$$

and

$$M'' + N_{Sc} F M' = 0 \quad (9)$$

with

$$F(0) = F'(0) = F'(\infty) = T(\infty) = M(\infty) = 0, \\ T(0) = M(0) = 1 \quad (10)$$

Here  $K_0$  is the coefficient of the first term in an expansion of the principal function  $K$  in terms of  $\xi$ , where

$$K(\xi) = \frac{1}{2} + \frac{1}{3} \frac{\xi r^{2\delta}}{S} \frac{d}{d\xi} \left( \frac{S}{r^{2\delta}} \right) \quad (11)$$

The procedure is first to determine  $\xi(x)$  and  $K_0$  from the body contour. Then the differential equations are solved. Since the primary intent is to develop functional relations rather than detailed numerical solutions, only the asymptotic forms of the differential equations for large and small values of the Schmidt and Prandtl numbers are investigated.

#### ASYMPTOTIC SOLUTIONS FOR EQUAL SCHMIDT AND PRANDTL NUMBERS

The interaction between heat and mass transfer is very simple for equal Schmidt and Prandtl numbers (the dimensionless concentration and temperature fields are equal), but the analysis serves to clarify more complex situations. Furthermore, the results are a reasonable approximation for many gaseous mixtures.

##### Case A: $N_{Sc}$ and $N_{Pr} \rightarrow 0$ , with $N_{Sc} = N_{Pr}$

The form of the differential equations and boundary conditions for Prandtl (and Schmidt) numbers of zero indicates that a singular perturbation analysis can be used. Hence, according to established procedures (15), two sets of asymptotic expansions are introduced. In an outer region

$$F(\eta) \sim (1 + \sigma)^{1/4} N_{Pr}^{-1/2} [f_o(y) + O(N_{Pr}^{1/2})] \quad (12)$$

and

$$T(\eta) \sim t_o(y) + O(N_{Pr}^{1/2}) \quad (13)$$

with

$$y = (1 + \sigma)^{1/4} N_{Pr}^{1/2} \eta \quad (14)$$

In an inner region

$$F(\eta) \sim (1 + \sigma)^{1/4} K_0^{-1/4} [f_i(\bar{y}) + O(N_{Pr}^{1/2})] \quad (15)$$

and

$$T(\eta) \sim t_i(\bar{y}) + O(N_{Pr}^{1/2}) \quad (16)$$

with

$$\bar{y} = (1 + \sigma)^{1/4} K_0^{-1/4} \eta \quad (17)$$

Substituting these expansions into the equations and letting the Prandtl number tend to zero, we obtain the following approximations for the inner region:

$$f_i''' + f_i f_i'' + K_0(1 - f_i'f_i') = 0 \quad (18)$$

and

$$f_i(0) = f_i'(0) = 0, \quad f_i'(\infty) = 1 \quad (19)$$

The temperature function  $t_i$  is constant and equal to unity. The corresponding equations for the outer region are

$$f_o f_o'' - K_0 f_o f_o' + t_o = 0, \quad (20)$$

$$t_o'' + f_o t_o' = 0 \quad (21)$$

and

$$f_o(0) = f_o'(\infty) = t_o(\infty) = 0, \quad t_o(0) = 1 \quad (22)$$

The boundary conditions have been determined by matching the inner and outer expansions to the same order in the Prandtl number. The result is that the inner stream function is determined by a differential equation and boundary conditions which are identical to those describing forced convection past wedge nosed bodies, that is, the Falkner-Skan equation (11). As a consequence, the solution for  $f_i$  can be regarded as known. The gradient of the temperature field is

$$T'(0) \sim (1 + \sigma)^{1/4} N_{Pr}^{1/2} t_o'(0) \quad (23)$$

Equations (20) and (21) can be solved by various methods. For  $K_0 = 2/3$ , corresponding to planar, sharp nosed bodies, the numerical solution of Lefevre (4) is  $t_o'(0) = -0.645$ . Use of a two-term approximation for  $f_o(y)$ , namely

$$f_o(y) = K_0^{-1/2} y + K_0^{1/2} (2K_0 - 1)^{-1} t_o'(0) y^2/2 + \dots \quad (24)$$

and Meksyn's method (6) yields  $t_o'(0) = -0.66$ .

In the limit, as the Prandtl number tends toward zero, the flow near the surface is described by differential equations which are independent of the Prandtl number; hence

$$N_T N_{Gr}^{-3/4} \sim \phi_1(x) [\xi(x)]^{3/4} (1 + \sigma)^{3/4} K_0^{-3/4} f_i''(0) \quad (25)$$

On the other hand, the Nusselt number varies as the square root of the Prandtl number

$$N_{Nu} N_{Gr}^{-1/4} \sim \phi_2(x) [\xi(x)]^{-1/4} (1 + \sigma) N_{Pr}^{1/2} [-t_o'(0)] \quad (26)$$

Here  $f_i''(0)$  and  $t_o'(0)$  must be determined for each body shape from solutions of the appropriate equations. It is evident that the effects of temperature and concentration differences are additive in this situation.

##### Case B: $N_{Sc}$ and $N_{Pr} \rightarrow \infty$ , with $N_{Sc} = N_{Pr}$

In this case, the inertial terms may be neglected near the surface of the object (8). Hence, the appropriate expansions for the inner region are

$$F(\eta) \sim (1 + \sigma)^{1/4} N_{Pr}^{-3/4} [f_i(y) + O(N_{Pr}^{-1/4})] \quad (27)$$

and

$$T(\eta) \sim t_i(y) + O(N_{Pr}^{-1/4}) \quad (28)$$

Here

$$y = (1 + \sigma)^{1/4} N_{Pr}^{3/4} \eta \quad (29)$$

When these expansions are substituted into the equations, the limiting forms for large Prandtl numbers are

$$f_i''' + t_i = 0 \quad (30)$$

and

$$t_i'' + f_i t_i' = 0 \quad (31)$$

with

$$f_i(0) = f_i'(0) = 0, \quad t_i(0) = 1 \quad (32)$$

The solution is

$$f_i(y) = f_i''(0) \frac{y^2}{2} - \int_0^y \int_0^{z_2} \int_0^{z_1} t_i(z_1) dz_1 dz_2 dz_3 \quad (33)$$

and

$$t_i(y) = 1 + t_o'(0) \int_0^y \exp \left( - \int_0^{z_2} f_i(z_1) dz_1 \right) dz_2 \quad (34)$$

In the previous case, the undetermined constants in the inner and outer expansions of the solutions to the boundary-layer equations were determined by matching. A different procedure is used now. Allowing the Prandtl number to increase without bound in the complete set of conservation equations indicates that in the outer region, the temperature function is identically zero. Hence, the flow can be considered irrotational to this approximation; that is, the Laplacian of the stream function for a planar flow is zero. As a consequence, the appropriate boundary conditions for the inner problem are

$$t_i(\infty) = 0, \quad f_i'(\infty) < \infty \quad (35)$$

That is, the velocity in the direction tangent to the surface is bounded. Integration by parts leads to

$$f_i'(y) = t_i'(0) \frac{y^2}{2} \int_y^\infty g(z) dz + \left( f_i''(0) + t_i'(0) \int_0^y z g(z) dz \right) y - t_i'(0) \int_0^y \frac{z^2}{2} g(z) dz \quad (36)$$

where

$$g(z_1) = \exp \left( - \int_0^{z_1} f_i(z) dz \right) \quad (37)$$

It follows, then, that the appropriate boundary condition for the stream function is

$$f_i''(\infty) = 0 \quad (38)$$

Although the differential equations have been solved numerically by Lefevre (4), who found  $f_i''(0) = 1.08$  and  $t_i'(0) = -0.54$ , analytical methods can be used to obtain accurate approximations. The problem is to evaluate the integrals

$$- f_i''(0) = t_i'(0) \int_0^\infty z g(z) dz \quad (39)$$

and

$$- [t_i'(0)]^{-1} = \int_0^\infty g(z) dz \quad (40)$$

The Laplace method described by Erdelyi (3) is used, recognizing that the series which are obtained may be semiconvergent. If only the first two terms in the series

$$f_i(y) = f_i''(0) \frac{y^2}{2} - \frac{y^3}{6} + \dots \quad (41)$$

are introduced, then

$$f_i''(0) = - t_i'(0) \left[ 2 \cdot 6^{-1/3} \Gamma \left( \frac{2}{3} \right) [f_i''(0)]^{-2/3} + 2^{-1} [f_i''(0)]^{-2} \right] \quad (42)$$

and

$$- [t_i'(0)]^{-1} = 2 \cdot 6^{-2/3} \Gamma \left( \frac{1}{3} \right) [f_i''(0)]^{-1/3} + 2^{-1} \cdot 6^{-1/3} \Gamma \left( \frac{5}{3} \right) [f_i''(0)]^{-5/3} \quad (43)$$

The solution is  $f_i''(0) = 1.04$  and  $t_i'(0) = -0.546$ , which is in excellent agreement with the numerical solution. Thus, for large Prandtl numbers, the asymptotic forms are (by using the exact solution)

$$N_T N_{Gr}^{-3/4} \sim 1.08 \phi_1(x) [\xi(x)]^{+1/4} (1 + \sigma)^{3/4} N_{Pr}^{-1/4} \quad (44)$$

and

$$N_{Nu} N_{Gr}^{-1/4} \sim 0.54 \phi_2(x) [\xi(x)]^{-1/4} (1 + \sigma)^{1/4} N_{Pr}^{1/4} \quad (45)$$

Once again, the effects due to temperature and concentration differences are additive.

The functional relations

$$N_{Nu} \propto N_{Pr}^{1/4}, \quad N_{Pr} \rightarrow \infty \quad (46)$$

$$N_{Nu} \propto N_{Pr}^{1/2}, \quad N_{Pr} \rightarrow 0 \quad (47)$$

have been known for some time. According to Ede (2), the former relation was derived by Lorenz in 1881, while the latter was suggested by Davis in 1921 and independently by Lefevre in 1956. These relations have been rederived here by using formal perturbation methods in order to provide a rationale for the assumptions which have heretofore been largely based on intuitive arguments. Furthermore, this development serves as a basis for the systematic construction of higher order terms in the expansions.

#### ASYMPTOTIC SOLUTIONS FOR SMALL PRANDTL NUMBERS AND LARGE SCHMIDT NUMBERS

Large Schmidt numbers and small Prandtl numbers are characteristic of liquid metals. Inner and outer expansions are developed in a manner similar to the previous analysis for small Prandtl numbers. For the inner region

$$f_i''' + f_i f_i'' + K_0(1 + \sigma m_i - f_i' f_i') = 0 \quad (48)$$

$$m_i'' + N_{Sc} f_i m_i' = 0 \quad (49)$$

with

$$f_i(0) = f_i'(0) = m_i(\infty) = 0, \quad f_i'(\infty) = m_i(0) = 1 \quad (50)$$

In the outer region

$$f_o f_o'' - K_0 f_o' f_o' + t_o = 0 \quad (51)$$

$$t_o'' + f_o t_o' = 0 \quad (52)$$

with

$$f_o(0) = f_o'(\infty) = t_o(\infty) = 0, \quad t_o(0) = 1 \quad (53)$$

The problem for the outer stream and temperature functions is the same as that treated previously, while the inner problem has the form of the equations for combined free and forced convection. The technique described by Meksyn (6) is applied to the inner problem to describe the effects of the Schmidt number and the ratio of driving forces,  $\sigma$ .

First, the expansions for small  $y$  are

$$f_i(y) = \frac{a}{2} y^2 - \frac{K_0}{6} (1 + \sigma) y^3 + \dots \quad (54)$$

and

$$m_i(y) = 1 + by + \dots \quad (55)$$

Then, following Meksyn

$$f_i'(\infty) \sim 2(a/6)^{2/3} \Gamma \left( \frac{1}{3} \right) - \frac{5}{16} K_0(1 + \sigma) (6/a)^{2/3} \Gamma \left( \frac{2}{3} \right) + \dots \quad (56)$$

so that retaining the first two terms gives

$$(a/6)^{2/3} = \left[ 4 \Gamma \left( \frac{1}{3} \right) \right]^{-1} \left[ 1 + \left[ 1 + 10 K_0(1 + \sigma) \Gamma \left( \frac{4}{3} \right) \Gamma \left( \frac{5}{3} \right) \right]^{1/2} \right] \quad (57)$$

For  $K_0 = 1$  (circular cylinders),  $\sigma = 0$  (forced convection), and a Schmidt number of 10, the result is  $m_i'(0) = -1.46$ . The exact value, as obtained by Squire (14), is

TABLE I. FUNCTIONAL DEPENDENCE OF THE SHEAR STRESS, NUSSELT, AND SHERWOOD NUMBERS ON THE GRASHOF, PRANDTL, AND SCHMIDT NUMBERS AND THE BUOYANCY FORCE RATIO

	$N_T$	$N_{Nu}$	$N_{Sh}$
$N_{Pr}, N_{Sc} \rightarrow 0$ ( $N_{Pr} = N_{Sc}$ )	$N_{Gr}^{3/4} (1 + \sigma)^{3/4}$	$N_{Gr}^{1/4} (1 + \sigma)^{1/4} N_{Pr}^{1/2}$	$N_{Gr}^{1/4} (1 + \sigma)^{1/4} N_{Sc}^{1/2}$
$N_{Pr}, N_{Sc} \rightarrow \infty$ ( $N_{Pr} = N_{Sc}$ )	$N_{Gr}^{3/4} (1 + \sigma)^{3/4} N_{Pr}^{-1/4}$	$N_{Gr}^{1/4} (1 + \sigma)^{1/4} N_{Pr}^{1/4}$	$N_{Gr}^{1/4} (1 + \sigma)^{1/4} N_{Sc}^{1/4}$
$N_{Pr} \rightarrow 0, N_{Sc} \rightarrow \infty$	$N_{Gr}^{3/4} (1 + \sigma)^{3/4}$	$N_{Gr}^{1/4} (1 + \sigma)^{1/4} N_{Pr}^{1/2}$	$N_{Gr}^{1/4} N_{Sc}^{1/3}$
$N_{Pr}, N_{Sc} \rightarrow \infty$ ( $N_{Sc} \gg N_{Pr}$ )†	$N_{Gr}^{3/4} N_{Pr}^{-1/4}$	$N_{Gr}^{1/4} N_{Pr}^{1/4}$	$N_{Gr}^{1/4} N_{Pr}^{-1/12} N_{Sc}^{+1/3}$

\* The dependence on  $\sigma$  is described by Equation (57).  
 † The dependence on  $\sigma$  is described by Equation (77).

-1.34, indicating that the accuracy of the method is adequate.\* Then

$$m_i'(0) \sim - \frac{(a/6)^{1/3}}{\Gamma\left(\frac{4}{3}\right)} N_{Sc}^{1/3} \quad (58)$$

Finally, for large Schmidt and small Prandtl numbers

$$N_{Sh} N_{Gr}^{-1/4} \sim \phi_2(x) \xi(x)^{-1/4} K_0^{-1/4} (a/6)^{1/3} \left[ \Gamma\left(\frac{4}{3}\right) \right]^{-1} N_{Sc}^{1/3} \quad (59)$$

The functional dependence on the one-third power of the Schmidt number is the expected result, since the flow near the surface is analogous to forced convection driven by the temperature field. The relations for the Nusselt number and the shear stress are still given to the present order of approximation by Equations (25) and (26).

#### ASYMPTOTIC SOLUTIONS FOR LARGE PRANDTL AND SCHMIDT NUMBERS

The Schmidt and Prandtl numbers characteristic of many viscous liquids are large compared with unity, with the Schmidt number larger (usually) than the Prandtl number. Adopting expressions similar to those used previously in the region near the object, namely

$$F(\eta) \sim N_{Pr}^{-3/4} [f_i(\eta) + 0(N_{Pr})^{-1/4}] \quad (60)$$

$$T(\eta) \sim t_i(\eta) + 0(N_{Pr})^{-1/4} \quad (61)$$

$$M(\eta) \sim m_i(\eta) + 0(N_{Pr})^{-1/4} \quad (62)$$

with

$$y = N_{Pr}^{1/4} \eta \quad (63)$$

we get

$$f_i''' + t_i + \sigma m_i = 0 \quad (64)$$

$$t_i'' + f_i t_i' = 0 \quad (65)$$

$$m_i'' + \frac{1}{\alpha} f_i m_i' = 0 \quad (66)$$

where  $\alpha = N_{Pr}/N_{Sc}$  is constant. The boundary conditions are

$$f_i(0) = f_i'(0) = f_i''(\infty) = t_i(\infty) = m_i(\infty) = 0, \quad t_i(0) = m_i(0) = 1 \quad (67)$$

The boundary conditions at infinity have been adopted for the reasons cited previously. Once again, the Laplace method is used to calculate the integrals

$$f_i''(0) = - t_i'(0) \int_0^\infty z g(z) dz - \sigma m_i(0) \int_0^\infty z h(z) dz \quad (68)$$

\* If two terms are used in the asymptotic expansion of the integral associated with  $m_i'(0)$ , the result is -1.40 at  $N_{Sc} = 10$ .

$$- [t_i'(0)]^{-1} = \int_0^\infty g(z) dz \quad (69)$$

and

$$- [m_i'(0)]^{-1} = \int_0^\infty h(z) dz \quad (70)$$

where

$$g(z_1) = \exp\left(-\int_0^{z_1} f_i(z) dz\right) \quad (71)$$

and

$$h(z_1) = \exp\left(-\int_0^{z_1} \alpha f_i(z) dz\right) \quad (72)$$

Then, by using the first two terms in the expansion for  $f_i$  about the origin

$$f_i''(0) = - t_i'(0) \left[ \left(\frac{4}{3}\right)^{1/3} \Gamma\left(\frac{2}{3}\right) [f_i''(0)]^{-2/3} + (1 + \sigma) 2^{-1} [f_i''(0)]^{-2} \right] - m_i'(0) \left(\frac{4}{3}\right)^{1/3} \Gamma\left(\frac{2}{3}\right) [f_i''(0)]^{-2/3} \alpha^{2/3} + (1 + \sigma) 2^{-1} [f_i''(0)]^{-2} \alpha^2 \quad (73)$$

$$- [t_i'(0)]^{-1} = 2 \cdot 6^{-2/3} \Gamma\left(\frac{1}{3}\right) [f_i''(0)]^{-1/3} + (1 + \sigma) 48^{-1/3} \Gamma\left(\frac{5}{3}\right) [f_i''(0)]^{-5/3} \quad (74)$$

and

$$- [m_i'(0)]^{-1} = 2 \cdot 6^{-2/3} \Gamma\left(\frac{1}{3}\right) [f_i''(0)]^{-1/3} \alpha^{1/3} + (1 + \sigma) 48^{-1/3} \Gamma\left(\frac{5}{3}\right) [f_i''(0)]^{-5/3} \alpha^{5/3} \quad (75)$$

These three equations can be reduced to a single cubic equation. In terms of

$$X = [f_i''(0)]^{4/3}, \quad a_1 = \left(\frac{4}{3}\right)^{1/3} \Gamma\left(\frac{1}{3}\right), \quad a_2 = (1 + \sigma)/2, \quad \alpha = N_{Pr}/N_{Sc}, \quad b_1 = 2 \cdot 6^{-2/3} \Gamma\left(\frac{2}{3}\right), \quad b_2 = (1 + \sigma) (48)^{-1/3} \Gamma\left(\frac{5}{3}\right) \quad (76)$$

this cubic is

$$b_1^2 X^3 + [b_1 b_2 (1 + \alpha^{4/3}) - a_1 b_1 (1 + \sigma \alpha^{1/3})] X^2 + [b_2^2 \alpha^{4/3} - a_1 b_2 (\alpha^{4/3} + \sigma \alpha^{1/3}) - a_2 b_1 (1 + \sigma \alpha^{3/5})] X - a_2 b_2 \alpha^{3/4} (1 + \sigma \alpha^{1/3}) = 0 \quad (77)$$

Equation (77) can be solved to obtain approximate values of  $f_i''(0)$ ,  $m_i'(0)$ , and  $t_i'(0)$  for specified values of  $\alpha$  and  $\sigma$ . The asymptotic behavior for small  $\alpha$ , that is,  $N_{Pr}/N_{Sc} \rightarrow 0$ , is

$$f_i''(0) = 0(1) \quad (78)$$

$$t_i'(0) = 0(1) \quad (79)$$

and

$$m_i'(0) = 0(\alpha^{-1/3}) = 0(N_{Sc}^{1/3}/N_{Pr}^{1/3}) \quad (80)$$

Near the object, the functional relationship between the mass transfer rate and the transport properties is similar to that for forced convection. The numerical results calculated from Equation (77) differ only slightly from results obtained by Mathers, Madden, and Piret (5), who used an analogue computed to obtain a few solutions to the same equations, namely (64) to (67). The asymptotic results, for  $N_{Pr}$  and  $N_{Sc} \rightarrow \infty$ , are

$$N_t N_{Gr}^{-3/4} \sim \phi_1(x) [\xi(x)]^{1/4} N_{Pr}^{-1/4} [f_i''(0)] \quad (81)$$

$$N_{Nu} N_{Gr}^{-1/4} \sim \phi_2(x) [\xi(x)]^{-1/4} N_{Pr}^{1/4} [-t_i'(0)] \quad (82)$$

$$N_{Sh} N_{Gr}^{-1/4} \sim \phi_2(x) [\xi(x)]^{-1/4} N_{Pr}^{1/4} [-m_i'(0)] \quad (83)$$

and all of the derivatives depend on the ratio of the Prandtl number to the Schmidt number.

## CONCLUSIONS

Asymptotic expressions were developed for the shear stress, Nusselt, and Sherwood numbers for free convection driven by simultaneous differences in temperature and composition. The results apply for laminar boundary-layer flows past horizontal cylinders and vertical axisymmetric bodies with fairly general body contours. In all cases, simple formulas were developed and, in some instances, shown to give relatively accurate approximations to solutions obtained by numerical methods.

The functional relationships between the relevant dimensionless groups are shown in Table 1. The physically exceptional cases of ( $N_{Pr} \rightarrow \infty$ ,  $N_{Sc} \rightarrow 0$ ) and ( $N_{Pr} > N_{Sc} \rightarrow \infty$ ) have not been included. The relations for these situations can be derived from those already given by simply interchanging the roles of the Schmidt and Prandtl numbers. The results show that the effects of temperature and concentration differences are not always additive, that is, the effective Grashof number is not always the sum of the Grashof numbers for heat and mass transfer. The effective body force results from density gradients due to temperature and composition variations in the fluid. The formulas reported in this paper apply principally to cases where these variations reinforce one another. However, in some instances, for example, for equal Schmidt and Prandtl numbers, the results apply irrespective of whether or not the variations reinforce one another. The first-order results obtained here by systematic perturbation methods can be extended in a rational manner.

## NOTATION

- $a$  = coefficient in the series expansion of  $f_i$ , Equation (54)  
 $a_1$  = constant, Equation (76)  
 $a_2$  = constant, Equation (76)  
 $b$  = coefficient in the series expansion of  $m_i$ , Equation (55)  
 $b_1$  = constant, Equation (76)  
 $b_2$  = constant, Equation (76)  
 $D_{AB}$  = molecular diffusivity of component A in the fluid, length<sup>2</sup>/time  
 $F$  = dimensionless stream function, that is,

$$\psi(x_1, x_2) = \left(\frac{4}{3}\right)^{3/4} \xi^{3/4} F(\xi, \eta)$$

- $f_i$  = dimensionless stream function for the inner region, Equations (15), (27), (60)  
 $f_o$  = dimensionless stream function for the outer region, Equations (20) and (51)  
 $g$  = gravitational constant, length/(time)<sup>2</sup>

$$g(z) = \exp \left[ - \int_0^z f_i(z_1) dz_1 \right]$$

$$h(z) = \exp \left[ - \int_0^z \alpha f_i(z_1) dz_1 \right]$$

- $J_A$  = mass flux density at surface, mass/(time)(length)<sup>2</sup>  
 $k$  = thermal conductivity, energy/(time)(length)(temperature)

- $K$  = principal function, Equation (11)

- $K_0$  = first term in the series expansion of  $K$

- $l$  = characteristic length

- $M$  = scaled mass fraction,  $(\omega - \omega_s)/(\omega_S - \omega_s)$

- $m_i$  = scaled mass fraction in the inner region, Equations (49) and (62)

- $N_{Gr}$  = Grashof number,  $g\beta\Delta\theta l^3/\nu^2$

- $N_{Nu}$  = Nusselt number,  $ql/k\Delta\theta$

- $N_{Pr}$  = Prandtl number,  $\nu/\alpha_t$

- $N_{Sc}$  = Schmidt number,  $\nu/D_{AB}$

- $N_{Sh}$  = Sherwood number,  $J_A l/\rho D_{AB} \Delta\omega$

- $N_T$  = shear stress number,  $\tau l^2/\nu\mu$

- $O(\epsilon)$  = of the order of  $\epsilon$

- $q$  = heat-flux density at surface, energy/(time)(length)<sup>2</sup>

- $r(x)$  = dimensionless radius of revolution,  $r_1/l$

- $r_1(x)$  = radius of revolution of an axisymmetric body

- $S(X)$  = sine of the angle between the body force vector and a normal to the surface of the immersed object

- $T$  = dimensionless temperature  $(\theta - \theta_s)/(\theta_S - \theta_s)$

- $t_i$  = dimensionless temperature function for the inner region, Equations (16), (18), (61)

- $t_o$  = dimensionless temperature function for the outer region, Equations (13) and (52)

- $x$  = dimensionless distance along the surface,  $x_1/l$

- $x_1$  = distance along the surface of the immersed object, length

- $x_2$  = distance normal to the surface of the immersed object, length

- $X$  =  $[f_i''(0)]^{4/3}$

- $y$  = scaled distance normal to the surface, Equations (14) and (29)

- $\bar{y}$  = scaled distance normal to the surface, Equation (17)

- $z, z_1, z_2, z_3$  = dummy integration variables

## Greek Letters

- $\alpha$  =  $N_{Pr}/N_{Sc}$

- $\alpha_t$  = thermal diffusivity of the fluid, (length)<sup>2</sup>/time

- $\beta$  = coefficient of expansion for temperature changes, (temperature)<sup>-1</sup>

- $\gamma$  = coefficient of expansion for composition changes, dimensionless

- $\Gamma(\ )$  = gamma function

- $\delta$  = zero for planar flows, unity for axisymmetric flows

- $\Delta\theta$  = characteristic temperature difference,  $(\theta_S - \theta_s)$

- $\Delta\omega$  = characteristic fraction mass difference,  $(\omega_S - \omega_s)$

- $\theta, \theta_s, \theta_\omega$  = temperature, temperature at surface, temperature far from surface

- $\eta$  = scaled normal distance, =

$$(x_2 Gr^{1/2}/l) \left(\frac{3}{4}\right)^{3/4} \frac{[S(x)]^{1/3}}{\xi^{1/4} [r(x)]^{26/3}}$$

- $\mu$  = viscosity, mass/(length)(time)

$\nu$  = kinematic viscosity, (length)<sup>2</sup>/time  
 $\xi$  = scaled tangential distance, Equation (6)  
 $\rho$  = density, mass/(length)<sup>3</sup>  
 $\sigma$  =  $\gamma\Delta\omega/\beta\Delta\theta$   
 $\tau$  = shear stress at surface, force/(length)<sup>2</sup>  
 $\phi_1, \phi_2$  = variables defined by Equations (4) and (5)  
 $\omega, \omega_s, \omega_\infty$  = mass fraction of component undergoing transfer, mass fraction at surface, mass fraction far from surface

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Manuscript received April 15, 1968; revision received July 26, 1968; paper accepted July 29, 1968. Paper presented at AIChE Philadelphia meeting.

# Combined Reactors: Formulation of Criteria and Operation of a Mixed Tubular Semifluidized Reactor

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Combined reactors in which the mixed reactor is followed by a tubular reactor can be optimal for a large number of simple adiabatic exothermal reactions. In the present paper, optimality criteria defined earlier for simple reactions involving a single reactant species have been extended to reactions involving two reactant species and for a system of consecutive reactions.

The oxidation of benzene has been studied in an adiabatic semifluidized mixed tubular (MT) reactor. A definite improvement is possible when the oxidation is carried out in this reactor, as observed by a comparison of the experimental results obtained in tubular, mixed, and MT reactors under adiabatic conditions.

The methods of optimizing the performance of a chemical reactor by introducing a temperature sequence in the case of stirred tank reactors and imposing an external temperature gradient in the case of tubular reactors have been described by Denbigh (7) who has also summarized (8) other important contributions in this area. In addition, the behavior of mixed and tubular reactors can also form a valuable basis for optimizing reactor performance.

In an elaborate analysis of mixing, Cholette, Blanchet, and Cloutier (5, 6) considered the case of a reactor which is partially mixed. The system was analyzed in terms of fully mixed and plug flow zones, and short circuiting was omitted from the analysis. The performance of a partially mixed reactor would then be determined by

the location of the fully mixed zone in the reactor. Two cases were considered: the fully mixed zone is present in the first part of the reactor and the second part is in tubular flow, and the first part of the reactor is in tubular flow while the second part is fully mixed. These two combinations were called, respectively, MT and TM combinations. Equations were then proposed for simple chemical reactions of different kinetics to predict the performance of MT and TM combinations under isothermal conditions.

These investigators also considered adiabatic systems (again for simple reactions) and formulated equations for the optimum combination of the mixed and tubular portions of a combined reactor. The studies were then extended by Aris (1) and by Douglas (9) who presented