COMMUNICATIONS TO THE EDITOR

Turbulent Diffusion and the Multicomponent Reynolds Analogy

H. L. TOOR
Carnegie Institute of Technology

An understanding of mass transfer in a turbulent multicomponent fluid necessitates some knowledge of turbulent diffusion in such a fluid. In this paper equations for turbulent diffusion in a multicomponent fluid will be developed and used to derive the multicomponent Reynolds analogy.

TURBULENT DIFFUSION

Consider transfer in a highly turbulent fluid where the flux due to molecular diffusion is negligible compared with the turbulent flux. The flux with respect to a fixed coordinate system may be written as the flux with respect to a coordinate system moving at the mean molar velocity plus the convective flux due to the moving coordinate. The flux with respect to the moving coordinate is caused by the turbulent mixing and, as in a binary system, may be assumed to be minus the product of a turbulent diffusivity, which is a function of the local conditions of turbulence, and the local gradient of the time mean concentration (*1*). For one dimension

$$N_i = Ny_i - \epsilon_i C \frac{dy_i}{dx}, \quad i = 1,2..n \qquad (1)$$

One of the above equations is not independent, and all the quantities used here are the usual time-averaged ones.

If Equation (1) is summed over all components, one obtains

$$\sum_{i=1}^{n} \epsilon_i C \frac{dy_i}{dx} = 0 \qquad (2)$$

so the two turbulent diffusivities in a binary system are always equal and any interaction between the molecular and turbulent diffusivities [of the type considered by Jenkins (*4*) for example] merely makes the single turbulent diffusivity a function of the single molecular diffusivity. In a multicomponent system in which the molecular diffusivities are unequal, however, any interaction between the molecular and turbulent diffusivities would not only make the turbulent diffusivities functions of molecular diffusivities but would make the turbulent diffusivities unequal.

If transfer takes place entirely by turbulent mixing, as assumed for example in mixing-length theories, then the ϵ_i are independent of molecular diffusivity and are all equal. Since this condition is approximately realized in binary systems where experiment and theory indicate that the effect of molecular Schmidt or Prandtl number on turbulent diffusivity is not large (*7*), it seems reasonable, at least as a first approximation, to assume that the same condition holds in multicomponent systems, $\epsilon_i = \epsilon$, for all *i*.

If it is further assumed that the presence of the flux *N* does not affect ϵ (this is equivalent to assuming that the velocity N/C is small enough so that its effect on the turbulence is negligible), then ϵ is fixed by the ordinary hydrodynamics and is independent of the number of species.

With $\epsilon_i = \epsilon$, Equation (1) becomes

$$N_i = Ny_i - \epsilon C \frac{dy_i}{dx}, \quad i = 1,2..n-1 \qquad (3)$$

There are of course other ways in which the eddy diffusivity could be defined, in terms of the mean mass velocity instead of mean molar velocity for example (*1*). However in dilute systems all definitions are essentially equivalent, and in concentrated systems the differences among the conclusions obtained by use of different definitions are probably less than the unknown coupling of the diffusion equations with the equations of motion. The present choice of a molar frame of reference leads to the simplest equations for gases, and the assumption that ϵ in Equation (3) has the same values as in a dilute binary fluid should give, at the worst, the correct magnitude of the transfer. In dilute mixtures the assumption should be very reliable.

Equation (3) is the same as the Maxwell-Stefan diffusion equation (*5*) for a system in which all the binary diffusion coefficients are equal to ϵ; therefore the solutions to Equation (3) also apply to the nonturbulent system in which all the diffusion coefficients are equal.

In equimolal transfer *N* is zero and from Equation (3) the flux of each component in an *n* component mixture follows the same equation as in binary equimolal transfer, so none of the multicomponent molecular diffusion phenomena which have been described earlier occur here (*10*).

When one gas is stagnant, species *n* for example in an *n* component mixture, it follows by summing Equation (3) over the nonstagnant species that the total flux is given by

$$N = \frac{\epsilon C}{y_n} \frac{dy_n}{dx} \qquad (4)$$

which is a generalization of the equivalent binary equation. In the binary case *N* is the flux of the single nonstagnant species, but in an *n* component system *N* is the sum of *n*—1 fluxes.

Combining Equation (4) with Equation (3) yields an explicit equation for the separate fluxes:

$$N_i = -\epsilon C\, y_n \frac{d(y_i/y_n)}{dx}, \quad i = 1,2..n-1 \qquad (5)$$

In the general case y_n is of course the sum of the mole fractions of all the nonstagnant gases.

INTEGRATED FORMS FOR STEADY STATE TRANSFER

In the following development the molar density is taken as constant. For one-dimensional steady state transfer, when there is no flow normal to *x*, all the fluxes are independent of *x* and Equation (3) can be directly integrated. A more general integration can be carried out if one considers the case where the fluxes all vary with *x* in the same manner:

$$\frac{N_i}{N_{iw}} = f(x) \qquad (6)$$

Cases in which Equation (6) may hold will be discussed later. Combining Equation (6) with Equation (3) and integrating from 0 to L, one obtains

$$N_w = \frac{\overline{\epsilon C}}{L} \ln\left(\frac{N_w y_{iL} - N_{iw}}{N_w y_{io} - N_{iw}}\right), i=1,2\ldots n \qquad (7)$$

where

$$\frac{L}{\overline{\epsilon}} = \int_o^L \frac{f(x)dx}{\epsilon} \qquad (8)$$

Equation (7) is indeterminate, and one more condition is required to allow solution for N_{iw}. When there is equimolal transfer, $N_w = 0$ and the solution is

$$N_{iw} = \frac{\overline{\epsilon C}}{L} (y_{io} - y_{iL}), i = 1,2\ldots n \qquad (9)$$

which is the same equation as obtained in a binary mixture.

When one or more components are stagnant, the total flux integral obtained from Equation (4) is

$$N_w = \frac{\overline{\epsilon C}}{L} \ln \frac{y_{nL}}{y_{no}} = \frac{\overline{\epsilon C}}{L} \frac{(y_{nL} - y_{no})}{(y_n)_m} \qquad (10)$$

and if this is combined with Equation (7), the separate fluxes are obtained:

$$N_{iw} = \frac{\overline{\epsilon C}}{L} \frac{y_{no}}{(y_n)_m} (\delta_T\, y_{io} - y_{iL}),$$
$$i=1,2\ldots n-1 \qquad (11)$$

where

$$\delta_T = \frac{y_{nL}}{y_{no}} \qquad (12)$$

The above results indicate that reverse diffusion and related phenomena which occur in multicomponent gaseous diffusion (*10*) can occur in turbulent multicomponent diffusion when there is a stagnant species but not in equimolal transfer. For a binary mixture Equation (11) reduces to the well-known form, and the effect of the bulk merely introduces the $(y_2)_m$ term (*8*).

It is important to note that the predicted interaction effects in turbulent diffusion are generally smaller than in molecular diffusion and are due to the bulk flow caused by the diffusion; therefore they disappear in equimolal transfer. In molecular diffusion the interactions, when one gas is stagnant, are a combination of interactions caused by differences in diffusion coefficients and flow interactions, and in this case, even in equimolal transfer, the interactions remain (*10*).

REYNOLDS ANALOGY

Mass transfer to a turbulent fluid is described by the combination of Equation (3) (suitably generalized) with a differential material balance, but most binary analogy calculations have avoided the necessity of solving the resulting equation by assuming the manner in which the flux varies with radial position. Since the absolute value of the radial flux must vary from a maximum at the wall of, say, a tube to zero at the center, a linear approximation to $f(x)$ in Equation (6) is correct in the limits,

$$\frac{N_i}{N_{iw}} = \left(1 - \frac{2x'}{d}\right) \quad i = 1,2 \qquad (13)$$

It can be shown for the binary case that for a constant flux at the wall this linear form is a good approximation (*3*). (Since binary equimolal and dilute binary transfer with the nondilute gas stagnant are both equivalent to heat transfer, results will be quoted without necessarily making any distinction.) Furthermore it has also been shown that away from the tube inlet and for all but the lowest Schmidt numbers (and consequently for all binary mass transfer) the wall concentration distribution has little effect on the mass transfer coefficient (*9*). Thus the multicomponent mass transfer problem will be attacked by considering mass transfer to a turbulent fluid moving in a tube under such conditions that there is a constant wall flux for all species.

The question immediately arises as to whether Equation (13) is as valid in a multicomponent system as in a binary one. In equimolal transfer, where there is no difference between multicomponent and binary transfer, the binary results are valid for each species, and so Equation (13) should be a good approximation for all components, This argument breaks down near the wall, where molecular diffusion is controlling, but can be replaced there by the argument that the flux cannot vary significantly in a thin film.

A more rigorous and more general argument, similar to that used by Churchill and Balzhiser (*3*) for heat transfer, is possible. For steady state transfer in a tube with radial symmetry, a material balance with axial diffusion neglected yields

$$C \frac{\partial(u(x'')y_i)}{\partial Z} + \frac{1}{x''} \frac{\partial}{\partial x''} (x''N_i)$$
$$= 0, i = 1,2\ldots n \qquad (14)$$

If the system is relatively dilute in diffusing species, then u can be taken as approximately constant with Z, and in the limit of a long tube with a constant wall flux for all species (the limit considered, usually implicitly, in binary systems) the concentration profiles change slowly; so $\partial y_i/\partial Z$ can be taken as independent of x'' and equal to $d\overline{y}_i/dZ$. With these assumptions Equation (14) is similar to the analogous heat transfer problem, and integration and substitution of a material balance which neglects variations in axial velocity yields

$$\frac{N_i}{N_{iw}} = \frac{4}{x''d} \int_o^{x''} x \;\frac{u(x)}{V}\, dx, i=1,2\ldots n \qquad (15)$$

which is essentially the same equation as obtained for heat transfer (*3*). Consequently for fluids which are not too concentrated the variation of N_i/N_{iw} with radius depends only upon the velocity profile, is the same for all species, and is the same as in heat transfer or in binary mass transfer. This variation is found to be close to linear (*3*) when a normal turbulent velocity profile is substituted into Equation (15).

By earlier assumptions the eddy diffusivity of mass has the same values in a multicomponent system as in a binary system when the hydrodynamics are the same in both cases. Since the eddy diffusivity of mass in a binary system is approximately equal to the eddy diffusivity of momentum, Equation (8) with the linear flux variation, the normal shear-stress relationship, and a force balance becomes

$$\frac{L}{\overline{\epsilon}} = \frac{\rho}{\tau_w} \int_{L1}^{L2} du \qquad (16)$$

and $L_2 - L_1 = L$.

The Reynolds analogy is obtained when the turbulence is assumed to extend to the wall so the velocity at L_1 is zero, when L_2 is taken as the radial position where the local velocity has the same value as the mean velocity, and when it is further assumed that at this latter point all the local concentrations have the same values as the bulk mean concentrations. Then

$$\frac{\overline{\epsilon}}{L} = \frac{\tau_w}{\rho V} = \frac{f}{2} V \qquad (17)$$

Subscript L now refers to the bulk concentration, subscript o becomes subscript w, and subscript w refers to the flux at the wall. Equation (7) becomes, for example,

$$N_w = \frac{f}{2} G_m \ln\left(\frac{N_w \overline{y}_i - N_{iw}}{N_w y_{iw} - N_{iw}}\right),$$
$$i = 1,2\ldots n \qquad (18)$$

and this is the multicomponent Reynolds analogy.

Even though the Reynolds analogy neglects the molecular diffusion which takes place adjacent to the boundary between a turbulent fluid and another phase, it is a good approximation for mass transfer in a binary gas because the Schmidt number in gases is close to 1 and the molecular diffusion equation in a binary gas is of the same form as the turbulent-diffusion equation. In multicomponent gases the molecular-diffusion equations can differ considerably from the above turbulent-diffusion equations, and so if molecular diffusion is significant, Equation (18) cannot in general be nearly so good an approximation for mass transfer in a multicomponent gas as in a binary one. Further developments, which include the effect of molecular diffusion, will be presented in a later paper.

Few concrete generalizations are possible in the case of liquids because of the paucity of data concerning multicomponent liquid diffusion. However in a liquid in which there are negligible cross-diffusion effects, Equation (1) describes molecular diffusion, and if the molar density does not vary too greatly and the various molecular diffusion coefficients are approximately equal (all these conditions may be satisfied in many systems), then, as in a binary system (2), Equation (18) can be used for mass transfer if the righthand side is corrected for the effect of Schmidt number by multiplying by the average Schmidt number to the −2/3 power:

$$N_w = \frac{f}{2} G_m (N_{Sc})^{-2/3} \ln\left(\frac{N_w \bar{y}_i - N_{iw}}{N_w y_{iw} - N_{iw}}\right) \qquad i = 1,2\ldots n \tag{19}$$

NOTATION

C = molar density, moles/cu. ft.
d = diameter, ft.
f = Fanning friction factor
$f(x)$ = function of x
G_m = molar mass velocity, moles/(hr.)(sq. ft.)
L = length of diffusion path, ft.
n = number of species
N = total molar flux with respect to a fixed coordinate, moles/(hr.)(sq. ft.)
N_i = molar flux of species i, with respect to a fixed coordinate, moles/(hr.)(sq. ft.)
N_{Sc} = Schmidt number
u = axial velocity, ft./hr.
V = mean axial velocity, ft./hr.
x = distance coordinate, ft.
x' = distance from wall, ft.
x'' = radial distance from tube center, ft.
y = mole fraction
Z = axial distance, ft.
δ_T = interaction term
ϵ = turbulent diffusivity, sq. ft./hr.
$\bar{\epsilon}$ = mean diffusivity which reduces to ϵ when the diffusivity is constant
ρ = mass density, lb.-mass/cu. ft.

Subscripts

i = species i
m = log mean
n = species n
w = position w or wall
o,L = positions in x coordinates
L_1,L_2 = positions in x' coordinates

LITERATURE CITED

1. Bird, R. B., W. E. Stewart, and E. H. Lightfoot, "Notes on Transport Phenomena," Wiley, New York (1958).
2. Chilton, T. H., and A. P. Colburn, *Ind. Eng. Chem.*, **26**, 1183 (1934).
3. Churchill, S. W., and R. E. Balzhiser, *Chem. Eng. Progr. Symposium Ser. No. 29*, **55**, 127 (1959).
4. Jenkins, R., "Heat Transfer and Fluid Mechanics Institute," Stanford Univ. Press, Palo Alto, Calif. (1951).
5. Maxwell, J. C., "Scientific Papers," **2**, Dover, New York (1952).
6. Reichardt, H., *Natl. Advisory Comm. Aeronaut. Tech. Memo 1047* (1943).
7. Sherwood, T. K., *Chem. Eng. Progr. Symposium Ser. No. 25*, **55**, 71 (1959).
8. ———, and R. L. Pigford, "Absorption and Extraction," McGraw-Hill, New York (1952).
9. Sleicher, C. A., Jr., and Myron Tribus, *Trans. Am. Soc. Mech. Engrs.*, **79**, 789 (1957).
10. Toor, H. L., *A.I.Ch.E. Journal*, 3, 198 (1957).