Turbulent Mass Transfer in a Field of Developing Inhomogeneous Turbulence

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The problem of predicting mass transfer rates and mean compositions in turbulent flow has been the subject of considerable investigation (*7, 14*). It has long been recognized that the application of the statistical theory of turbulence to these problems requires a knowledge of velocity, density, and composition fluctuations and their correlations. With few exceptions (*12*) such data are unavailable, nor can they be predicted a priori from specification of the boundary conditions. In addition the mathematical complexity of problems involving anisotropic turbulence (which include the majority of practical cases) usually precludes the application of the statistical theory.

This paper describes an investigation of the possibility of treating turbulent mass transfer by means of a suitable eddy transfer coefficient, finding values of the eddy transfer coefficients as a function of position in a region of developing turbulence (as in mixing of coaxial jets), and finally relating the coefficient to the patterns of flow which must give rise to it. For the steady state case the definition chosen is given by writing the material balance equation for any nonreacting component j in the form

$$-\rho \vec{u} \cdot \nabla \chi_j + \nabla \cdot (\epsilon \nabla \chi_j) = 0 \quad (1)$$

In this equation only time mean values appear, and it has been assumed that the turbulent transfer coefficient is a scalar.* (In the situation treated below nearly all of the mass transfer is in one direction, and the tensor form is unnecessary.) This definition is not new, having been applied in essentially the same manner as used herein by other authors to systems involving fully developed turbulence (*9, 10*). Still others have obtained values of ϵ by assuming it to be a simple function of the flow parameters (usually a constant) at the point in question, integrating the equation, and matching the integrated result to experimental data (*1, 13*). This latter method, while useful, gives little information about the detailed behavior of the transfer coefficient. While the previous results can be applied to systems like those for which they were measured, they cannot be applied to problems with different boundary conditions or regions of developing flow which are cases of considerable practical interest. The obvious virtue of the proposed treatment is that it can lead to a prediction of turbulent mass transport without knowledge of the composition-velocity-fluctuation correlations needed in the statistical theory, while it retains the generality needed to describe systems involving different boundary conditions.

It should be pointed out that the proposed method of attack can yield only time mean (or overall) compositions. The extent to which the various species are mixed on a molecular scale cannot be determined by this approach.

* It is to be noted that the dimensions of the transfer coefficient are $M/L\theta$ rather than those of the usual eddy diffusivity L^2/θ. This is because the driving force is gradient of mass fraction rather than gradient of concentration. This definition precludes a diffusive mass transfer due to density variation.

ANALYSIS OF DATA OF FORSTALL AND SHAPIRO

Values of the turbulent transfer coefficient in the mixing regions of a system of coaxial jets were obtained from the original data of Forstall and Shapiro (*5*) made available through the cooperation of Professor Shapiro. The data were obtained from a system consisting of a primary jet of helium-air mixture fed into a co-axial secondary stream of air, both streams having initially low turbulence intensity. The data consist of measurements of mean composition and impact velocity as a function of position in the mixing region. System geometry and operating conditions are shown in Table 1. Details of the apparatus are presented in (*4, 5*).

The internal consistency of the original data was evaluated by calculation of the total mass flux through each axial cross section and of the total flux of helium through each axial cross section. Variations were less than 3%. Within the accuracy of the measurements axial symmetry was verified. Static pressure variations were insignificant for mass transfer considerations. It is recognized that the criteria above are necessary but not sufficient conditions, but they are the only criteria which can be applied. The condition that the composition and velocity be smooth, continuous functions of space coordinates is very important because the data were necessarily differentiated. On this basis the runs G 1-4 are omitted.

To treat the axially-symmetric data Equation (1) is expanded in cylindrical coordinates:

$$-\rho u_z \frac{\partial \chi_j}{\partial Z} - \rho u_R \frac{\partial \chi_j}{\partial R} + \epsilon \left(\frac{\partial^2 \chi_j}{\partial Z^2} + \frac{1}{R}\frac{\partial \chi_j}{\partial R} + \frac{\partial^2 \chi_j}{\partial R^2} \right) + \left(\frac{\partial \epsilon}{\partial R} \right) \left(\frac{\partial \chi_j}{\partial R} \right) + \left(\frac{\partial \epsilon}{\partial Z} \right) \left(\frac{\partial \chi_j}{\partial Z} \right) = 0 \quad (2)$$

Application of this equation to the data requires knowledge of the axial and radial velocity components, density, and the appropriate derivatives of the mass fraction. The velocity components were obtained from the impact velocities reported in the data by computing the streamlines (based on mean flow) and finding the angle between the streamlines and the axis of the system. The streamlines were obtained by integration of the mass flux radially from the axis at each axial position and connection of radii bounding equal mass fluxes. Figure 1 is a plot of some of the streamlines so obtained. The appropriate derivatives of the composition were obtained by numerical differentiation of the composition data either by the Douglass-Avakian method (*11*) or by fitting polynomials to the data to a least-squared error and finding the derivative of the polynomial at the desired point.

Values of the turbulent transfer coefficient were found by numerical inte-

TABLE 1

Run no.	F 1-8	F 9-14	G 1-4	G 5-10
d_N (in.)	1/4	1	1	1/4
Primary velocity (ft./sec.) U_p	90	90	225	225
Secondary velocity (ft./sec.) U_s	45	45	45	45
% He in Primary (%$_v$) Feed	9.92	8.85	8.60	9.80
Duct diameter (in.)	4	4	4	4

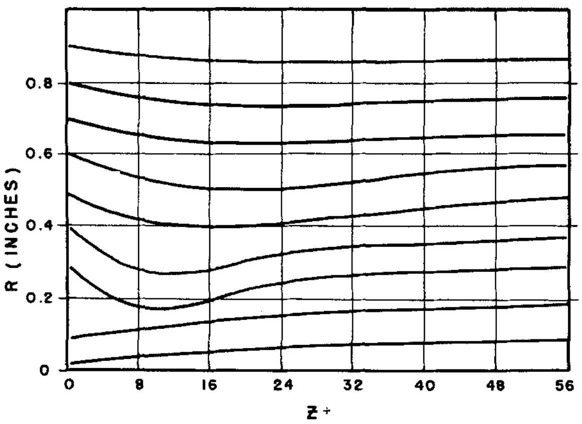

Fig. 1. Lines of mean flow runs G 5-10.

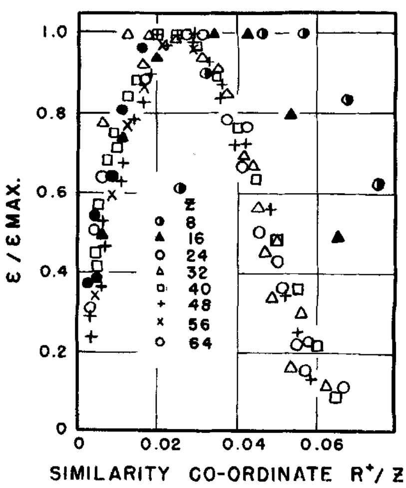

Fig. 2. Profiles of transfer coefficient runs G 5-10.

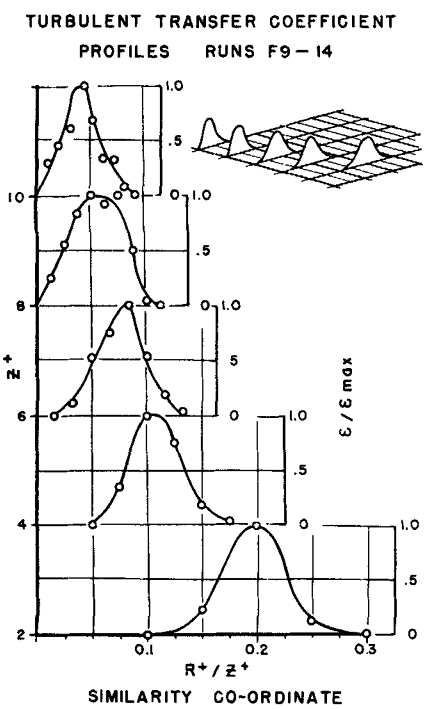

Fig. 3. Turbulent transfer coefficient profiles, runs F 9-14.

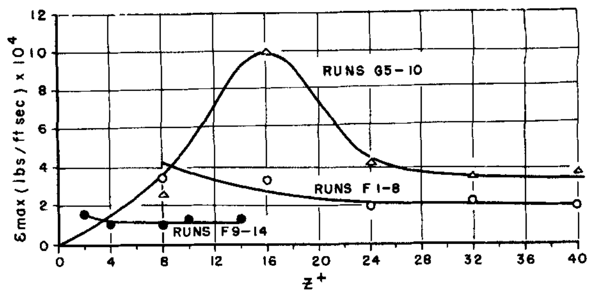

Fig. 4. Maximum turbulent transfer coefficient.

gration of Equation (2). An iterative method was used to carry out the integration, which gave substantially the same results as a more tedious finite difference method. A spot check showed that the original composition profile could be reproduced by integration of Equation (2) by the use of smoothed values of the turbulent transfer coefficient obtained and the original velocity data with the necessary compositions from the original data as boundary values. (The profile was obtained by an integral method. It was found that a relaxation technique required too fine a mesh to be practical.)

The values of the velocities, compositions, and computed values of turbulent transfer coefficient are given in Table 2.* The run number designations are those of Dr. Forstall.

DISCUSSION

Because of the numerical treatment of the data some scattering of the results is to be expected. Some of the profiles of turbulent transfer coefficient are shown in Figures 2 and 3. The difference in the form of the plots is a result of the fact that the data of runs F 9-14 (Figure 3) are sensibly confined to a region where the mixing field is developing, while those of runs G 5-10 (Figure 2) are for the region sufficiently far from the nozzle that the flow is fairly well developed. The ordinates of both plots are the normalized transfer coefficients. The relatively wide range of maxima of the transfer coefficients is illustrated in Figure 4.

For axial positions greater than 16 nozzle diam. downstream, where the similarity correlation (Figure 2) holds reasonably well, the ratio of the maximum values of the turbulent transfer coefficient to the maximum value of the rate of strain is nearly proportional to the downstream distance. However such a simple relation fails completely in correlating the data of runs F 9-14, and a more general method of relating the turbulent transfer coefficient to the patterns of flow producing it was sought.

* Tabular material has been deposited as document 7356 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or for 35-mm. microfilm.

To develop a more reasonable approach the following qualitative ideas were considered:

1. The character of the turbulence (and of the turbulent diffusion coefficient) at any point and time (P, t) is influenced not only by events at (P, t) but also by events upstream of (P, t) at points (P', t').
2. Turbulence is produced by shear, and therefore the coefficient of turbulent diffusion at (P, t) is dependent upon the shear at points influencing it, (P', t').
3. Turbulence produced in a parcel of fluid at some time and position will decay as the parcel moves away from the initial point unless further generation takes place. This decay, or the rate of it, is probably dependent on the viscosity of the fluid and the extent to which the parcel is agitated.

In order to give quantitative expression to the above, consider the turbulent diffusion coefficient at point $P(x, y, z)$ and at t denoted by $\epsilon(P, t)$. This will be a consequence of events at many points (P') upstream of P at times (t'). As an approximation one may write

$$\epsilon(P, t) = \int_{t'} \int_{V'} \xi(P', t', P, t) f(P', t') dV' dt' \quad (3)$$

Clearly it has been assumed that the fractional influence at P, t is independent of events between P', t' and P, t.

As a first approximation it was assumed that $\xi(P', t', P, t)$ has a value different from zero only for points $P' = P_s'$, where the set of points P_s' lies on the streamline passing through P, t; for $t' = t_s'$ where t_s' is the time at which the parcel of fluid at P, t passed through P_s', $(t - t_s')$ is nonnegative. These approximations permit rewriting of Equation (3):

$$\epsilon(P, t) = \int_{S'=-\infty}^{S'=S} \xi(P_s', t_s', P, t) g(P_s') dS' \quad (4)$$

This ignores the obvious fact that turbulence in one region alters the patterns of flow in adjacent regions.

Because the analysis is to be applied to a steady state situation ϵ and ξ are independent of t, and the choice of P, t, and P_s' will specify t_s'. Equation (4) may therefore be written

$$\epsilon(P) = \int_{S'=-\infty}^{S'=S} \xi_1(P_s', P) g(P_s') dS' \quad (5)$$

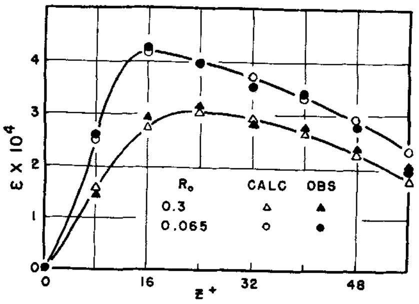

Fig. 5. Development of ϵ along flow lines, runs G 5-10.

The simplest expressions were adopted for the factors ξ_1 and g. If decay of eddy transfer coefficient is proportional to itself [see assumption (3) above]

$$\frac{d\epsilon}{dt_s'} = -\beta\epsilon \qquad (6)$$

This equation may be integrated immediately to give

$$\frac{\epsilon(P)}{\epsilon(P')} = \exp[-\beta(t-t_s')] = \xi_1(P_s', P) \qquad (7)$$

As noted earlier this treatment assumes that each bit of turbulence produced decays independently of subsequent events as the parcel of fluid in which it is produced moves downstream. For $g(P_s')$ a simple proportionality to the absolute value of the rate of strain was chosen. Because there is significant rate of strain in the radial direction only one obtains

$$g(P_s') = \alpha \left| \frac{\partial u_z}{\partial R} \right|_{P_s'} \qquad (8)$$

Substitution of Equations (7) and (8) into Equation (5) then yields

$$\epsilon(P) = \int_{S'=-\infty}^{S'=S} \alpha \left| \frac{\partial u_z}{\partial R} \right|_{P_s'} \exp[-\beta(t-t_s')]dS' \qquad (9)$$

A cursory examination of the dimensions of $\alpha(M/L^2)$ and of $\beta(1/\theta)$ as well as the assumptions involved will indicate that this formulation cannot be complete. It was desirable to see if it has the correct qualitative behavior.

The velocity data were used to evaluate the integral in Equation (9) along several streamlines for each run, the value of β being chosen to give the correct shape of the curve (by comparison with the turbulent transfer coefficient), and the value of α being chosen to make the curve pass reasonably well through the observed points. Because interpolation of the numerical results was necessary, the phrase observed points and the notation ϵ_{obs} should be interpreted as points read from smooth curves based on the values of ϵ obtained from Equation (2). Typical comparisons are shown in Figures 5, 6, and 7 where two streamlines (one originating in the primary stream and passing through the potential cone and one originating in the secondary stream) are shown in each plot. The coordinates of the plots are ϵ and $Z^+ = Z/d_N$. In view of the differences in the shapes of the curves the agreement is surprisingly good.

It is interesting that a single value of β could be used for all streamlines and all runs and that it was necessary to adjust α only for different runs, not for different streamlines in the same run. The values of α and β are shown in Table 3.

It is obvious that values of the coefficient of turbulent diffusion could not be obtained in regions where the composition is constant, for example the inlet streams. Therefore any unknown turbulence in the feed streams due to the boundary layers on the inner nozzle would be reflected in a necessary alteration of the constants.

A comparison of the values of the turbulent transfer coefficient obtained via Equation (9) from the flow pattern (ϵ_{calc}) with the values calculated from the composition data (ϵ_{obs}) is shown in Figure 8. In this plot all the points are presented for all of the streamlines for which Equation (9) was evaluated. While there is considerable scatter, this is not surprising when the number of operations required to produce either coordinate is considered. Furthermore when one recognizes that the two constants α and β have been adjusted only to give the general level of the curves, and their dimensions indicate that some further theoretical work is necessary, the agreement is satisfactory.

The authors know of no previous effect to relate the turbulent transport coefficient to the properties of the flow causing it in a field of developing turbulence. Emmons has developed a probability theory of transition to turbulence (3) taking account of events upstream of the point under consideration. The effect of time for diffusion in a homogeneous field of turbulence has also been discussed (2, 6).

The results of this investigation indicate that it is possible to obtain numerical values of turbulent exchange coefficients with sufficient accuracy to permit interpretation. Furthermore the results show that the qualitative ideas leading to the relation between transfer coefficient and patterns of flow are correct. In principle enough information of this kind would permit prediction of the turbulent diffusion as a function of the boundary conditions and patterns of flow in the mixing field.

TABLE 3

Run no.	F 1-8	F 9-14	G 5-10
$\alpha \times 10^7$ (lb.$_m$/sq. ft.)	4.07	2.15	2.59
β (sec.$^{-1}$)	32.5	32.5	32.5

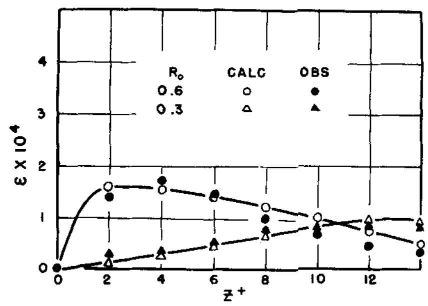

Fig. 6. Development of ϵ along flow lines, runs F 9-14.

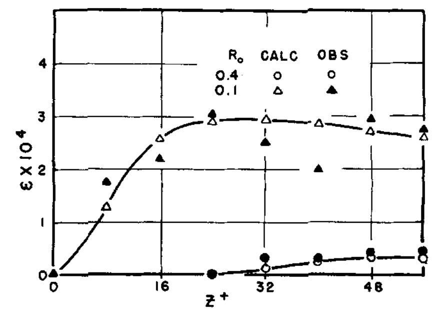

Fig. 7. Development of ϵ along flow lines, runs F 1-8.

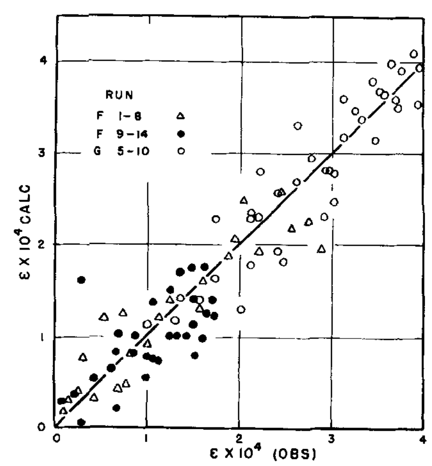

Fig. 8. Comparison of Equation (9) with experiment.

There are several aspects of the approach indicated which need further clarification by application to other data. It is necessary to generalize the quantities α and β so that they may be predicted. These two quantities must to some extent be dependent on the properties of the fluids involved and the boundary conditions on the mixing region. Furthermore Equation (9) does not admit a diffusive transfer on a streamline to which there has been no transfer of momentum. Since many experimental results show clearly a more rapid spread of mass than of momentum in jet mixing (for example 4), it appears that a modification may be necessary to include regions near the edge of the jet. (The present results are not accurate near the edge of the jet.)

It is clear finally that prediction of the turbulent transfer coefficient by

Equation (9) requires a knowledge of the flow pattern. The flow pattern must in turn be determined in conjunction with a knowledge of the turbulent momentum transport. It is suggested that methods similar to those employed herein be applied to the momentum transfer problem.

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NOTATION

d_N = diameter of primary jet nozzle
ϵ = turbulent transfer coefficient, ($lb._m$/ft. sec.)
f, g = generation functions
P = coordinates of the point P
R = radial coordinate, (in. where numerical values are given)
R^+ = dimensionless radial coordinate, R/d_N
R_o = radial coordinate at the plane of the nozzle, in.
S = distance along a streamline
t = time
$\vec{u}$ = velocity vector
u_z = axial velocity, (ft./sec. where numerical values are given)
u_R = radial velocity, (ft./sec. where numerical values are given)
V = volume
X = mole fraction of helium
Z = axial coordinate
Z^+ = dimensionless axial coordinate, Z/d_N

Greek Letters

α, β = constants (see text)
ξ, ξ_1 = influence functions
ρ = density, ($lb._m$/cu. ft where numerical values are given)
χ_j = mass fraction of species, j
∇ = vector operator (gradient)

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Axial Dispersion in a Packed Bed

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Until recently the transport of mass in the direction of flow by a diffusion mechanism has been neglected in the study of rate processes in packed beds. Beginning in 1953 a number of theoretical studies (*1, 2, 3, 4, 5, 6, 7, 8, 9*) and experimental investigations (*10, 11, 12, 13, 14, 15*) were made to determine the nature and magnitude of the axial diffusion mechanism. The perfect mixing cell model, in which each of the interstices of a packed bed acts as a mixing stage, was proposed by Kramers and Alberda (*4*) and further investigated by others (*2, 14*). The experimental measurements of McHenry and Wilhelm (*14*) verified the deductions of the mixing cell model for gas-flow systems.

The measurements of axial diffusion in liquid-flow systems made by Geankoplis and his co-workers (*13, 15*), Carberry and Bretton (*11*), Ebach and White (*12*), and others produced diffusion coefficients that were in sharp disagreement with the perfect mixing cell model. Figure 1 summarizes the results of several of these investigations.

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Carberry (*11*) inferred from his measurements that some kind of capacitive effect appeared to exist. Deans and Lapidus (*3*) suggested that stagnant fluid regions produced the capacitive effect and from order-of-magnitude considerations estimated that this could account for the discrepancy between the gas-flow and the liquid-flow diffusion coefficients. Turner (*8, 9*) and Aris (*1*) have investigated a mathematical model for the capacitive effect in terms of distributed pockets of stagnant fluid in a packed bed. Application to existing experimental data was not made however. In the present investigation an analysis somewhat similar to Turner's, although differing considerably in detail, is applied to reported experiments in axial dispersion.

THE FILM MODEL

A packed bed can be idealized by dividing each of its interstices into two regions. In one region turbulence produces complete mixing. In the other region the fluid is stagnant, and mixing occurs incompletely by molecular diffusion. The continuity equation for a tracer substance in the fluid is

$$D_L \frac{\partial^2 c}{\partial x^2} - \frac{U}{\epsilon_f} \frac{\partial c}{\partial x} - \frac{\partial c}{\partial t} - \frac{\epsilon - \epsilon_f}{\epsilon_f} \frac{\partial q}{\partial t} = 0 \qquad (1)$$

This equation differs from the one used by earlier investigators only in the appearance of a term that accounts for the holdup in the stagnant fluid and in the use of a so-called *effective porosity* which is the fraction of the packed bed volume occupied by the perfectly mixed regions. In most of the experiments reported it appears that no large-scale, transverse concentration gradients existed, thus the absence of such terms in Equation (1).

In addition an equation to describe mass transport in the stagnant fluid is required. It will be assumed that such transport occurs only by molecular diffusion and only in the direction transverse to the flow. Furthermore it will be assumed that the stagnant fluid forms a thin film of uniform thickness surrounding the particles in the packed