Mass Transfer in Dilute Turbulent and Nonturbulent Systems with Rapid Irreversible Reactions and Equal Diffusivities

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Studies of mass transfer with rapid, homogeneous, irreversible reactions between dilute species with equal diffusivities are carried out with a device which reduces the system of mass transfer-reaction equations to the equations for mass transfer in the absence of reaction.

A relationship which gives the rate of transfer into stagnant, laminar, or turbulent reacting systems is developed. Under some conditions the reaction increases the transfer rates by a constant factor, while under other conditions the reaction increases the individual or over-all mass transfer coefficients by a constant factor.

Calculations of concentration profiles, conversion, etc. are carried out for the Graetz problem with a homogeneous reaction between a dilute species in the fluid and a dilute species diffusing from the tube wall. Reaction completion times for various stagnant elements in infinite media are given.

Although the flux into a turbulent reacting fluid is related to the time average behavior in the absence of reaction, the time average concentrations depend also upon the relative intensity of the concentration fluctuations in the absence of reaction. This latter dependence is obtained from a relationship of Hawthorne, et al.

Surfaces on which the time average composition is stoichiometric and the reactant concentrations are proportional to the relative intensity of the concentration fluctuations in the absence of reaction are obtained for turbulent flow in a tube with a homogeneous reaction between the fluid and wall. Distances for almost complete reaction are estimated.

The time average conversion is related to the accomplished mixing for the case in which two dilute reactants are turbulently mixed. In a uniform, stoichiometric mixture the fractional conversion equals the accomplished mixing. Calculations are carried out for uniform, nonstoichiometric mixtures.

It was shown in an earlier paper (*18*) that under certain conditions the differential equations which describe simultaneous diffusion, convection (or briefly, mass transfer), and homogeneous chemical reaction can be reduced to the equations for mass transfer without reaction. This allows the solution to the reaction mass transfer problem to be written down immediately if the solution to the corresponding pure mass transfer problem is known and also allows deductions concerning the interaction between the reaction and the mass transfer when the mass transfer problem cannot be solved. Thus information is obtained concerning certain limited types of reactions for rather general hydrodynamic and geometrical conditions, which compliments the usual approach which yields information concerning a broader class of reactions for limited hydrodynamic models. References to the various solutions based on film, penetration, and boundary-layer models are available elsewhere (*14, 15, 16*).

In this paper the earlier results (*18*) concerning the effect of a homogeneous, infinitely rapid, irreversible reaction on mass transfer are generalized, and some nonturbulent and turbulent problems are studied.

The mass transfer and chemical reaction in an isothermal system in which the D_i are constant, $\nabla \cdot \underline{V} = 0$, and cross-diffusion effects are negligible (essentially dilute systems) are described by

$$\frac{\partial C_i}{\partial \theta} + \underline{V} \cdot \nabla C_i = \frac{DC_i}{D\theta} = D_i \nabla^2 C_i + r_i \quad (1)$$

Consider the homogeneous irreversible reaction considered earlier (*18*) in which the stoichiometric equation is

$$a\,(1) + b\,(2) \rightarrow \text{products} \quad (2)$$

so

$$br_1 - ar_2 = 0 \quad (3)$$

If $D_1 = D_2$, Equations (1) and (3) together yield (*18*)

$$\frac{DC_t}{D\theta} = D_1 \nabla^2 C_t \quad (4)$$

with

$$C_t = bC_1 - aC_2 \quad (5)$$

(C_t above is C'_t in reference *18*.)

Equation (4) shows that the quantity C_t is conserved by the reaction. Its value in a differential element can only be changed by diffusion or convection through the boundaries of the element, so C_t must also satisfy all the usual macroscopic material balances. The fluxes are easily shown to satisfy the equation

$$\underline{N}_t = b\underline{N}_1 - a\underline{N}_2 = -D_1 \nabla C_t + \underline{V} C_t \quad (6)$$

Equations (4) and (6) are identical to the equations which describe mass transfer of a species in the absence of reaction if C_t is replaced by C°. Thus if the boundary conditions on C_t are of the same form as on C°, the solution to the reaction problem is obtained from the solution to the equivalent mass transfer problem by the transformations

$$\begin{aligned} C^\circ &\rightarrow C_t \\ \underline{N}^\circ &\rightarrow \underline{N}_t \\ \overline{D}^\circ &\rightarrow \overline{D}_1 \end{aligned} \quad (7)$$

The second transformation is more general than the one given earlier (*18*) and can be applied to a surface across which convection is taking place. The equivalent mass transfer or nonreacting problem is the one in which mass transfer takes place in the absence of reaction with the velocity field, geometry, diffusivity, and form of the boundary conditions the same as in the reaction problem.

The above results are valid for fast or slow reactions in both laminar and turbulent flow. In order to proceed any further it is necessary to identify the quantities C_t and N_t which will appear in the solution to a specific problem, and this identification is only possible in the case of reactions which are so fast with respect to the diffusion that a surface (or surfaces) of reaction exists.

Although the authors of (*18*) were not aware of it earlier, Burke and Schumann (*2*) and others (*9, 19*) used a technique in principle similar to the above to analyze combustion in a laminar flame.

NONTURBULENT SYSTEMS

The reaction surface (or surfaces) must be continuous, and since species 1 and 2 cannot exist together at the same point, C_t can be seen to be zero at, and only at, this surface. This condition allows C_t to be evaluated.

Concentration Profiles

In this section concentration profiles will be obtained for the class of nonturbulent problems in which the boundary conditions contain two constant concentrations. In particular problems in which the solutions to the equivalent mass transfer problem are of the form

$$\frac{C^\circ - C^\circ_0}{C^\circ_I - C^\circ_0} = f\,(Z_1, .\,.Z_n) \qquad (8)$$

where the function f depends upon geometry and the velocity field. In the most general case there is one reduced time coordinate and three reduced space coordinates. This would be the solution for example for a fluid element with internal circulation which has no symmetry and with initial constant concentration C°_0 which is immersed in an infinite medium of constant concentration C°_1, or whose surface is held at C°_I. With n equal to 3 (three space coordinates) Equation (8) is the solution for steady flow in a conduit where the inlet concentration is C°_0 and the wall concentration C°_I. For a symmetrical problem such as (steady) unidirectional flow in a tube n is 2.

Consider now a reacting system in which the concentration of species 1 is C_{1I} at the boundary at which the concentration is C°_I in the nonreacting system (for example the wall of a conduit or surface of a fluid element), and the concentration of species 2 is C_{20} at the boundary at which the concentration is C°_0 in the nonreacting system (for example the value at the inlet to a conduit or at time zero in a fluid element). It follows from Equations (7) and (8) that in the reacting system

$$\frac{C_t - C_{t0}}{C_{tI} - C_{t0}} = f(Z_1, .\,.\,Z_n) \qquad (9)$$

Since a reaction surface exists, there is no species 2 at boundary I and no species 1 at boundary 0; therefore

$$C_{tI} = b\,C_{1I}$$
$$C_{t0} = -\,aC_{20} \qquad (10)$$

and Equation (9) becomes

$$\frac{bC_1 - aC_2 + aC_{20}}{bC_{1I} + aC_{20}} = f(Z_1, .\,.\,Z_n) \quad (11)$$

Since $C_t = 0$ defines the reaction surface, Equation (11) yields

$$\frac{\beta}{\beta + 1} = f(Z_{1s}, .\,.\,Z_{ns}) \qquad (12)$$

The reaction surface defined by Equation (12) is the locus of points in the nonreacting system of Equation (8) at which the accomplished concentration change has the value $\beta/(\beta + 1)$.

When Z_{2s} takes on a certain value, the reaction is complete in a symmetrical system with $n = 2$, for species 2 is then completely reacted, and with a proper choice of coordinates this value is zero. Thus Z_{1f}, the time or distance coordinate at which the reaction is complete in this two dimensional system, is given by

$$\frac{\beta}{\beta + 1} = f(Z_{1f}, 0) \qquad (13)$$

or the reaction is complete when the accomplished concentration change at the center line of the equivalent mass transfer problem has reached $\beta/(\beta + 1)$.

When one returns to the general case, the individual concentration profiles are obtained from Equation (11). On one side of the reaction surface

$$\frac{C_1}{C_{1I}} = (1 + \beta)\,f(Z_1, .\,.\,Z_n) - \beta \quad (14)$$

and $C_2 = 0$. On the other side

$$\frac{C_2}{C_{20}} = 1 - \frac{\beta + 1}{\beta} f(Z_1, .\,.\,Z_n) \quad (15)$$

and $C_1 = 0$.

The bulk mean concentration in the nonreacting system is obtained by integrating Equation (8) over n—1 space coordinates with a weighting function which depends upon geometry and velocity

$$\frac{\bar{C}^\circ - C^\circ_0}{C^\circ_I - C^\circ_0} = \int_{z_2} \cdot\cdot \int_{z_n} f(Z_1, .\,.\,Z_n)$$
$$w(Z_2, .\,.\,Z_n)\,dZ_2\,.\,.\,dZ_n = \bar{f}(Z_1) \quad (16)$$

and by integration of Equations (9) and (11)

$$\frac{\bar{C}_t - C_{t0}}{C_{tI} - C_{t0}} =$$
$$\frac{(\bar{C}_1/C_{1I}) - \beta(\bar{C}_2/C_{20}) + \beta}{\beta + 1} = \bar{f}(Z_1) \qquad (17)$$

Similarly from Equation (14)

$$\frac{\bar{C}_1}{C_{1I}} = \int_{z_2} \cdot\cdot \int_{z_n} [(1 + \beta)f(Z_1, .\,.Z_n) -$$
$$\beta]\,w(Z_2\,.\,.\,Z_n)\,dZ_2\,.\,.\,dZ_n \quad (18)$$

Equations (12), (17), and (18) allow determination of the mean concentrations of species 1 and 2 when f and w are known, and Equations (16) and (17) also show that in addition to Equation (7) there is the auxiliary transformation

$$\bar{C}^\circ \to \bar{C}_t \qquad (19)$$

But, since a bulk mean concentration is always some type of space average of the point concentration, the first transformation of (7) shows that Equation (19) is genera and is not restricted to the type of boundary conditions considered above.

Rate of Transfer

Consider the nonreacting system where species 1 is being transferred and the local rate of mass transfer is given by

$$N^\circ_1 = k^\circ\,(C^\circ_{1I} - \bar{C}^\circ_1) \qquad (20)$$

One now inquires as to what rate of transfer of species 1 will be across the same boundary if species 2 is present in the fluid. If the transformations (7) and (19) are applied to Equation (20) and a material balance is written on the conserved quantity $\bar{C}_t$

$$N_t = k^\circ\,(C_{tI} - \bar{C}_t) = S\,\frac{d(\bar{C}_t\,Q)}{dZ_1} \quad (21)$$

With Equation (10) this becomes

$$k^\circ\,(bC_{1I} - \bar{C}_t) = S\,\frac{d(\bar{C}_t\,Q)}{dZ_1} \quad (22)$$

and at some given value of Z_1, $\bar{C}_t = -\,aC_{20}$. Furthermore since there is no flux of species 2 at the boundary, Equations (6), (21), and (22) yield

$$N_1 = k^\circ\,(C_{1I} - \bar{C}_t/b) \qquad (23)$$

When C_{1I} and C_{20}, the geometry and velocity field, are given, the equivalent mass transfer problem is defined. (C_{1I} need not be constant, and, for that matter, the inlet concentration of species 2 does not have to be constant here either.) Therefore if the local mass transfer coefficient in this nonreacting problem is known, Equation (22) may be integrated. The resulting equation gives $\bar{C}_t$ as a function of Z_1, and Equation (23) then gives the flux of species 1 across the boundary. In the special case in which C_{tI} is a constant (and Q is treated as constant) the integration of Equation (22) or (21) yields Equation (17) with $\bar{f}(Z_1)$ in terms of k°. The integration can also be carried out if an external resistance is present as indicated in the next section.

Although Equations (22) and (23) give the flux of species 1 into the reacting system, it is desirable to determine the relationship between this flux and the flux which would be obtained in the absence of reaction. To this end, by using the definition of $\bar{C}_t$, Equation (23) is put in the form

$$N_1 = \left(1 + \beta\,\frac{\bar{C}_2/C_{20}}{1 - \bar{C}_1/C_{1I}}\right)k^\circ(C_{1I} - \bar{C}_1) \qquad (24)$$

which can be compared with the flux which would exist at the same point or instant of time if species 2 were not present and the bulk concentration were $\bar{C}_1$:

$$N_1^{\circ\prime} = k^\circ(C_{1I} - \bar{C}_1) \qquad (25)$$

Equation (25) shows that only when the reaction surface is so close to the boundary I that $\bar{C}_1$ can be taken as zero and $\bar{C}_2$ as C_{20} does the constant reaction factor $(1 + \beta)$ exist. In general the first factor on the right in Equation (24) varies from $(1 + \beta)$ at the inlet to a conduit or at time zero to 1.0 at the point or instant that the reaction is complete. k°, as well as $\bar{C}_1$ and $\bar{C}_2$, generally depends upon position or time, and although the former term does not depend upon the reaction, the latter two do. Consequently when the reaction surface is not close to the boundary, the effect of the reaction on the flux depends upon the form of the boundary conditions. [The film, penetration, and boundary-layer models inherently assume that the reaction surface is close to the I boundary which is why they all yield the constant reaction factor of $(1 + \beta)$ for the conditions of this study $(D_1 = D_2)$ (*7, 5, 6*).]

When C_{1I} and C_{20} are constants, Equation (17) may be combined with Equation (24):

$$N_1 = (1 + \beta)\, k^\circ C_{1I}(1 - \bar{f}(Z_1)) \quad (26)$$

But the flux which would exist at the point or instant under consideration if there were no reaction is given by Equation (20), and with Equation (16), since there is no species 1 present at the inlet (or at time zero) in this nonreacting system

$$N^\circ_1 = k^\circ C_{1I}\,(1 - \bar{f}(Z_1)) \quad (27)$$

so

$$N_1 = (1 + \beta)\, N^\circ_1 \quad (28)$$

and by integration over the boundary of the system

$$\bar{N}_1 = (1 + \beta)\, \bar{N}^\circ_1 \quad (29)$$

so both the point and mean rate of transfer in the presence of the reaction is $(1 + \beta)$ times the rate which would be obtained at the same point or over the same area in the absence of a reaction. This increased rate will persist even after the reaction is completed. Note that Equations (28) and (29) are valid whether or not the reaction surface is close to the boundary and that N°_1 is not the rate which would be obtained if the concentration at the position were $\bar{C}_1$; it is the rate which would be obtained at that position if species 2 were not present in the inlet stream, that is no reaction took place, and all other conditions were the same as in the reaction problem. In this latter case the bulk concentration of species 1 is not $\bar{C}_1$ but $\bar{C}^\circ_1$, and $\bar{C}^\circ_1$ is obtained by integrating Equation (20) with the boundary conditions $C^\circ_{10} = 0$ and $C^\circ_{1I} = C_{1I}$.

When the reaction surface is close to the boundary I, Equation (28) is then equivalent to the earlier statement that the reaction increases the mass transfer coefficient by a factor $(1 + \beta)$. Under these conditions Equations (28) and (29) are valid irrespective of the hydrodynamics or concentration boundary conditions, but when the reaction surface is not close to the boundary, the proof of Equations (28) and (29) depends upon the validity of Equation (9).

Rate of Transfer with External Resistance

Consider a two phase nonreacting system where the transfer of species 1 in the external phase is given by

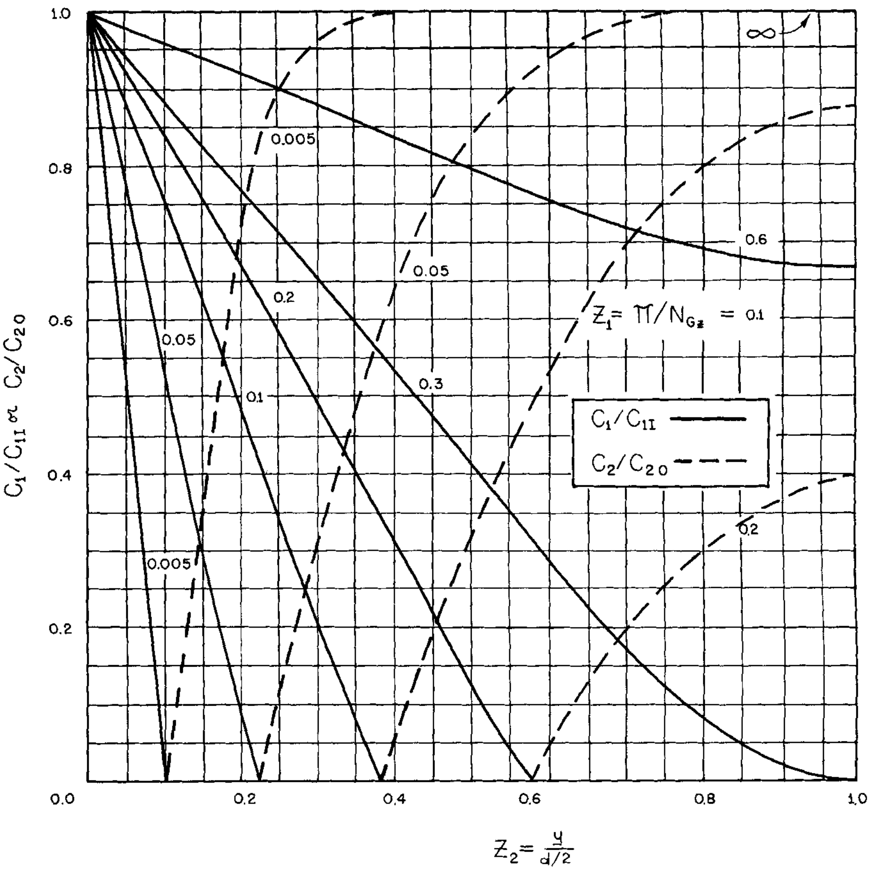

Fig. 1. Concentration profiles for laminar flow in a tube with reaction $\beta = 1$.

$$N^\circ_1 = k^\circ\,(\bar{\eta}^\circ_1 - \eta^\circ_{1I}) \quad (30)$$

With the equilibrium relationship

$$\eta^\circ_1 = mC^\circ_1 \quad (31)$$

the rate of transfer across the interface can also be written as

$$N^\circ_1 = k^\circ m(C_1^{*\circ} - C^\circ_{1I}) \quad (32)$$

If attention is focused on the internal phase, Equation (32) is a boundary condition for this internal phase.

Now let the earlier reaction with species 2 take place in the internal phase. Since there can be no species 2 in the external phase, Equation (32) becomes

$$N_t = k^\circ m(C_t^* - C_{tI}) \quad (33)$$

and this is the boundary condition for Equation (4). Consequently the boundary conditions on C_t in the presence of an external resistance are of the same form as on C°_1; therefore the earlier transformations apply to systems of this type also, and Equation (21) can in general be integrated by use of Equation (33) and a material balance.

In the same manner as before one obtains for the reaction problem

$$N_1 = \left(1 + \beta^* \frac{\bar{C}_2/C_{20}}{1 - \bar{C}_1/C_1^*}\right) K^\circ (C_1^* - \bar{C}_1) \quad (34)$$

and here if the reaction surface is close to the interface, the effective over-all mass transfer coefficient for species 1 is $(1 + \beta^*)$ times the value in the absence of reaction

$$N_1 = [(1 + \beta^*)\, K^\circ]\,(C_1^* - \bar{C}_1) \quad (35)$$

If C_1^* is a constant, Equation (28) follows as before with β^* in place of β. If C_1^* varies but the solution to the equivalent mass transfer problem is of the form

$$\frac{C_1^{\circ *} - \bar{C}_1^\circ}{C_{10}^{\circ *} - C_{10}^\circ} = G(Z_1) \quad (36)$$

which is frequently the case when the two phases are contacted, it can be shown that

$$N_1 = (1 + \beta_0^*)\, N^\circ_1 \quad (37)$$

where the rate of transfer if species 2 were not present would be

$$N_1^\circ = K^\circ (C_1^{\circ *} - \bar{C}_1^\circ) \quad (38)$$

with $C^\circ_{10} = 0$ and $C_1^{\circ *} = C_1^*$.

In many cases of interest the complete solution to the problem of mass transfer without reaction is not known (a sphere with circulation or a liquid film in a packed column are some examples) usually because the hydrodynamics are not known, and all that is available is a knowledge of the mass transfer coefficient. Since the mass transfer coefficient does not uniquely define the flow patterns, and thus the concentration profiles, a complete solution to the reaction problem is not possible.

Equations (28) or (37) will usually be valid in situations of this type however. Thus although the amount of species 1 which enters the system can be obtained, the amount reacted cannot be, and it can only be obtained when a complete solution to the equivalent mass transfer problem is available.

The minimum time or distance needed to complete the reaction occurs when stoichiometric amounts of species 1 and 2 are present in the fluid, and this corresponds to $\bar{C}_t = 0$. Thus for example when Equation (17) is valid, this minimum value of Z_{1f} is given by

$$\frac{\beta}{\beta + 1} = \bar{f}(Z_{1f}) \tag{39}$$

and $\bar{f}$ is known when the mass transfer coefficient in the absence of reaction is known.

Reaction Between A Laminar Fluid and A Species Dissolving from A Tube Wall

As an example of the application of the above equations consider steady laminar flow in a tube. The fluid enters the tube with species 2 at a constant concentration C_{20}, and the tube walls are held at a concentration C_{1I}. Species 2 reacts rapidly and irreversibly with species 1, the system is dilute enough for the earlier assumptions to hold, and $D_1 = D_2$. If the fluid is Newtonian and the parabolic profile is fully developed at the tube entrance, $f(Z_1, Z_2)$ is the solution to the Graetz problem, Z_1 is the reduced axial distance $= \pi (N_{Gz})^{-1}$, and Z_2 the reduced radial distance. The solution to the analogous irreversible first-order reaction problem is given by Lauwerier (*12*).

The concentration profiles for $\beta = 1$ were calculated with Equations (12 to 16) in conjunction with the Graetz and Leveque solutions (*10*) for $f(Z_1, Z_2)$. The profiles are presented in Figure 1 for various values of Z_1. The solid lines represent species 1 and the dotted lines species 2; their intersection is the reaction surface which is seen to move inward very rapidly at first, reaching 10% of the tube radius by $Z_1 = 0.005$, although a Z_1 of 0.3 is required for the reaction surface to reach the center of the tube, at which point species 2 is completely reacted.

The mean concentrations obtained from Equations (17) and (18) are shown for $\beta = 1$ in Figure 2. $\bar{C}_2$ becomes zero when the reaction surface reaches the tube center at $Z_1 = 0.3$. At this same point the reaction factor, which was $(1 + \beta) = 2$ at the tube entrance, has fallen to the limiting value of 1.0.

It is interesting that over most of its range the fractional conversion curve F_2 shown in Figure 2 is very close to that which would be obtained if the flow were rodlike, the radial mixing were infinitely rapid, and the reaction velocity constant were given by

$$k_e \cong 50 \frac{D_1}{a^2} \tag{40}$$

The reaction surfaces for various values of β are shown in Figure 3. The region inside a conical reaction surface contains species 2, and the region outside contains species 1. Values of β equal to zero and infinity correspond to no reaction. As the former limit is approached, the reaction surface approaches $Z_1 = 0$ since the trace of species 2 present in the liquid feed is reacted very rapidly, while in the latter limit the reaction surface approaches the tube wall since a negligible amount of species 2 is reacted by the trace of species 1 at the wall.

The number of diameters needed to complete the reaction is given by

$$\frac{x_f}{d} = \frac{1}{4} N_{Pe} Z_{1f}(\beta) \tag{41}$$

where Z_{1f} is obtained from Equation (13). With $\beta = 1$ and a Reynolds number of 1,000 about seventy diameters are needed to complete the reaction in gases ($N_{Sc} \cong 1$) and about 70,000 diameters in liquids ($N_{Sc} \cong 1{,}000$).

Stagnant Media

Consider a sphere, a long cylinder, or a wide slab which initially contains species 2 at a constant concentration of C_{20} and which at time zero is immersed in an infinite medium in which the concentration is C_{1I}, or whose surface is brought to the concentration C_{1I} at the time zero. [The cylinder in an infinite medium is the case which was treated by earlier authors (2).] If Z_1 is the reduced time $(4D\theta)/(h^2)$ and Z_2 the reduced distance from the center, Equations (9) to (29) apply directly, and since the $f(Z_1, Z_2)$ are all known, the solutions to the reaction problem may be written down immediately.

Figure 4 shows for example the reduced time required to complete the reaction with an infinite surrounding medium. A few values of θ_f are given in the figure for spheres.

TURBULENT SYSTEMS

Since Equations (1) to (6) apply to turbulent systems, the transformations (7) also apply, but the quantities are instantaneous values.

If the instantaneous quantities are divided into the usual time averaged and fluctuating quantities, the time average of Equation (4)

$$\frac{\partial \tilde{C}_t}{\partial \theta} + \nabla \cdot (\tilde{\underline{V}} \tilde{C}_t + \widetilde{\underline{v} c_t}) = D \nabla^2 \tilde{C}_t \tag{42}$$

is naturally of the same form as the time averaged transport equation in the absence of reaction.

Equation (7) yields the following transformations:

$$\begin{gathered} \tilde{C}^o \to \tilde{C}_t \\ c^o \to c_t \\ \widetilde{\underline{v}c}^o \to \widetilde{\underline{v}c}_t \\ \tilde{\underline{N}} \to \tilde{\underline{N}}_t \end{gathered} \tag{43}$$

and these implicitly require that the instantaneous boundary conditions as well as the geometry and turbulent fields be of the same form in both reacting and nonreacting systems.

As before the above results are valid whatever the reaction velocity, but only rapid reactions lead to useful solutions.

If a small element in a turbulent fluid is considered, it is seen that just as in nonturbulent systems, when the reaction is rapid, there must be a region in the element which contains species 1 alone which is separated from a region containing species 2 alone by a reaction surface defined by

$$C_t = \tilde{C}_t + c_t = 0 \tag{44}$$

Consequently $C_t = 0$ does not define the reaction surface, rather the reaction must take place on surfaces which are distributed throughout a volume of fluid which surrounds the surface at which $\tilde{C}_t$ is zero. In this case a point at which $\tilde{C}_t$ is zero is a point at which the time average composition is the stoichiometric composition. This differs from the comparable nonturbulent system, where the same point is a reaction surface where C_1 and C_2 are zero, but is similar to a nonturbulent system with a slow reaction.

Thus on a time average basis there is no continuous reaction surface but rather a reactive volume, and the surface $\tilde{C}_t = 0$ must lie inside this volume. Measurements of concentration profiles under these conditions are available (*8*).

It follows that the time average behavior of a nonreacting system does not allow a complete determination of the time average behavior of a reacting system, even for rapid reactions.

Time average Profiles and Transfer Rates

The first and last of the transformations (43) are equivalent to those obtained for nonturbulent systems [Equation (7)], and they give the time average behavior of a reacting system in terms of the time average behavior in the equivalent nonreacting system. Since Equations (7) and (43) are of the same form, it is clear that the transformations may be applied to any region of a fluid, whatever the turbulence level.

Consequently when turbulent systems with rapid reactions are considered on a time averaged basis, they differ from nonturbulent systems only in the fact that a reaction surface does not exist. Thus if an equation like (8) describes the time average concentration profiles in the turbulent mass transfer problem where C^o_{I} and C^o_0 are constants, and the geometry and turbulent field are fixed

$$\frac{\tilde{C}^o - C^o_0}{C^o_{\mathrm{I}} - C^o_0} = f(Z_1, \ldots Z_n) \qquad (45)$$

and f here is not the same function as in Equation (8).

The application of Equation (43) to the reaction problem with the same geometry and turbulent field in which the concentration of species 1 is a constant $C_{1\mathrm{I}}$ at boundary I and a constant C_{20} at boundary 0 then yields

$$\frac{\tilde{C}_t - C_{t0}}{C_{t\mathrm{I}} - C_{t0}} = \frac{b\tilde{C}_1 - a\tilde{C}_2 + aC_{20}}{bC_{1\mathrm{I}} + aC_{20}} = f(Z_1, \ldots Z_n) \qquad (46)$$

It is assumed in both of the above instances that the boundary concentrations are independent of time. It is not necessary but it avoids the difficulty which arises in matching instantaneous boundary conditions in the reacting and nonreacting problems.

Equation (46) is similar to Equation (11). Since a reaction surface does not exist here, the individual concentration profiles cannot be obtained, so the turbulent equivalents of Equation (12) to (16) and (18) cannot yet be written. However Equations (17) and (19) do apply here, and they need not be rewritten for $\bar{\tilde{C}} = C$ for any subscript or superscript on C.

It follows that all the results obtained earlier concerning the rate of transfer of species I across the I boundary in a nonturbulent system are equally valid in turbulent systems in which the boundary concentrations are not fluctuating. In particular, since N_1 in the rate equations is the flux at the boundary, $\bar{\tilde{N}}_1 = N_1$ and Equations (20) to (38) are valid in turbulent systems of the type considered.

Although the flux of species 1 can be determined in the present study even though the complete problem is unsolved, this is not the case when the molecular diffusivities are unequal. Sherwood and Ryan (*16*) for example studied a system in which there was a rapid irreversible reaction with unequal diffusivities, and in order to determine N_1 they were forced to assume (incorrectly as they realized) the existence of a reaction surface defined by $\tilde{C}_t = 0$.

At this point it can be seen that the turbulent reaction problem is similar to the nonturbulent problem in which only the mass transfer coefficient is known; in both cases the flux of species 1 into the system can be obtained, but the individual concentration profiles and the amount of reaction cannot be obtained because the hydrodynamics are undefined. Nevertheless some knowledge is to be gained at this point by considering the following problem in some detail.

Reaction Between a Turbulent Fluid and a Species Dissolving From a Tube Wall

A turbulent fluid containing only species 2 at a constant concentration C_{20} enters a tube whose wall is held at the constant concentration $C_{1\mathrm{I}}$. Species 1 and 2 react rapidly and irreversibly in accordance with Equation (2).

The minimum possible distance required for species 2 to be completely reacted is obtained when a stoichiometric quantity of species 1 has entered the fluid, and this, as before, corresponds to $\bar{\tilde{C}}_t = 0$. Equation (17) then gives this minimum distance. If the variation of k^o in the immediate vicinity of the tube entrance is neglected

$$\bar{f}(Z_1) = 1 - \exp\,(-4\,k^o x/Ud) \qquad (47)$$

and Equation (17) with $\bar{\tilde{C}}_t = 0$ gives

$$\left(\frac{x_f}{d}\right)_{l\,\mathrm{min}} = \frac{U}{4\,k^o} \ln\,(1 + \beta) \qquad (48)$$

[Note that with slight modifications Equation (48) applies to more complicated geometries, a tower packed with solid species 1 for example.]

Another value for the reaction distance can also be obtained by determining the distance required for $\tilde{C}_t$ at the center line to go to zero. This is equivalent to assuming that a time average reaction surface does exist, so Equation (12) can be used. For the constant wall concentration under consideration analogy calculations of the type carried out by Sleicher and Tribus (*17*) can be used to determine $f(Z_1, 0)$. For simplicity only distances away from the tube inlet are considered, and the variation of k^o near the inlet is again neglected. The result is

$$\left(\frac{x_f}{d}\right)_{\mathrm{min}} = \frac{U}{4\,k^o} \ln\,[(1 + \beta)\,\Phi^o] \qquad (49)$$

The length given by this equation is also a minimum, for when $\tilde{C}_t = 0$, $\tilde{C}_1$ and $\tilde{C}_2$ are not yet zero. $(x_f/d)_{\mathrm{min}}$ is only slightly greater than $(x_f/d)_{l\,\mathrm{min}}$, for Φ^o is not much greater than 1.

With the Colburn equation (*15*) for k^o Equation (49) becomes

$$\left(\frac{x_f}{d}\right)_{\mathrm{min}} = \frac{(N_{Sc})^{2/3}}{2f'} \ln\,[(1 + \beta)\,\Phi] \qquad (50)$$

so $(x_f/d)_{\mathrm{min}}$ is not a strong function of Reynolds number or β. In fact the largest variation is from gases to liquids, approximately 60 to 4,500, respectively for $\beta = 1.0$ and $N_{Re} = 10^4$. In terms of the apparent holdup time

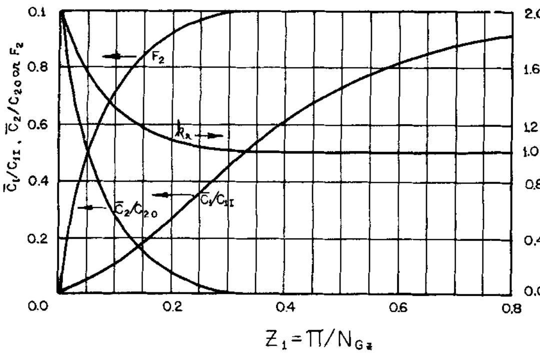

Fig. 2. Mean concentrations, conversion, and reaction factor. Laminar flow in a tube $\beta = 1$.

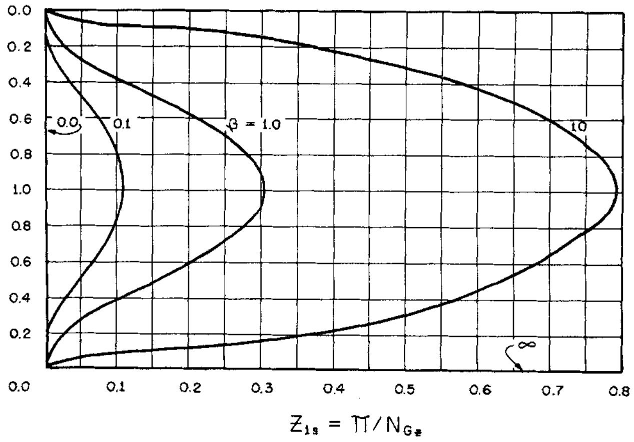

Fig. 3. Reaction surfaces for various values of β. Laminar flow in a tube.

$$\frac{\theta_f D_1}{d^2} = \frac{\ln[(1+\beta)\Phi^\circ]}{2f'(N_{Re})(N_{Sc})^{1/3}} \quad (51)$$

The holdup time required to carry out the reaction decreases markedly with increasing Reynolds numbers and is a strong function of tube diameter.

The surface at which $\widetilde{C}_t = 0$ may be calculated from Equation (12) with the above mentioned analogy calculation for $f(Z_1, Z_2)$. For a Schmidt Number of 1.0 the simpler Reynolds analogy may be used away from the inlet if one makes the approximation that the developed concentration profiles with a constant wall concentration are the same shape as the developed profiles with a constant wall flux. The result is

$$\frac{u}{U} = \frac{1}{\beta+1}\exp\left(2f'\frac{x_s}{d'}\right) \quad (52)$$

The curves in Figure 5 for various values of β were obtained for a Reynolds number of 40,000 with u^+, y^+ used to obtain u/U. The curves are much flatter in the center than the comparable laminar ones in Figure 3 and if put on a comparable scale are about 1/40 the length.

Turbulent Mixing and Rate of Reaction

It has been seen that even with rapid reactions the time average rate of reaction between two species in a turbulent fluid could not be deduced from the time average concentration profiles in the equivalent nonreacting system. However a relationship developed by Hawthorne, Weddell, and Hottel (8) to measure the unmixedness in turbulent combustion can be used to determine the reaction rate. Since the nomenclature and viewpoints are different, the derivation of this relationship is outlined.

Attention is centered on a fixed point in the turbulent reacting fluid where neither $\widetilde{C}_1$ nor $\widetilde{C}_2$ is zero and the time average conditions are steady. Reaction surfaces must exist on an instantaneous basis, and because of the gross turbulent mixing these surfaces move back and forth past the point. Consequently both species cannot be present at the same instant; the instantaneous concentration is either C_1 or C_2.

By definition C_t is bC_1 when species 1 is present and $-a\,C_2$ when species 2 is present. Thus when C_t is positive so that

$$\widetilde{C}_t + c_t > 0 \quad (53)$$

then

$$C_1 = \frac{C_t}{b} = \frac{\widetilde{C}_t + c_t}{b} \quad (54)$$

and when Equation (53) does not hold C_1 is zero. $\widetilde{C}_t$ can be positive or negative. Consider the former case.

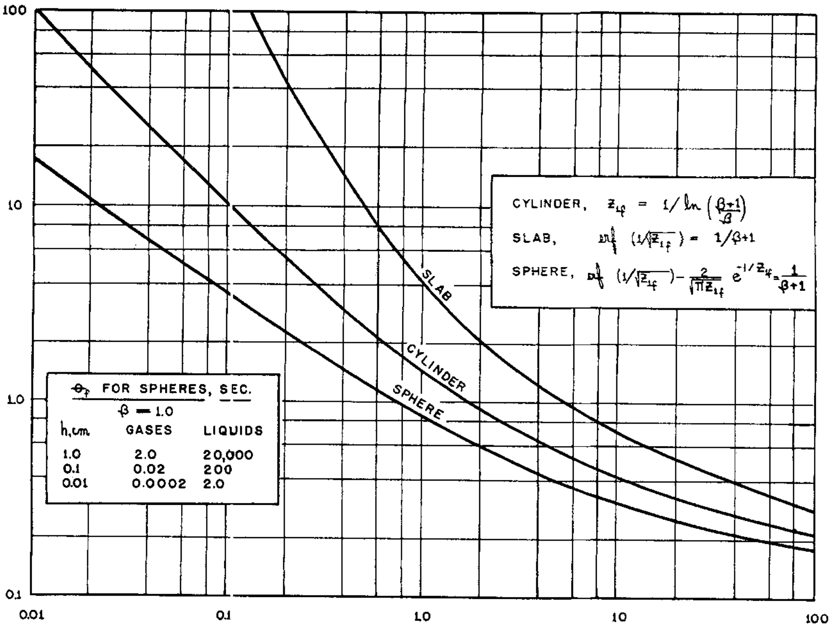

Fig. 4. Reaction completion times for stagnant elements in infinite media.

Then from Equation (53) C_1 is zero except when $c_t > -\widetilde{C}_t$. Using this condition and Equation (54) one obtains

$$\widetilde{C}_1 \equiv \int_{-\infty}^{\infty} (\widetilde{C}_1 + c_1)\,\phi\,(c_1)\,dc_1 =$$

$$\frac{1}{b}\int_{-\widetilde{c}_t}^{\infty} (\widetilde{C}_t + c_t)\,\phi\,(c_t)\,dc_t \quad (55)$$

and the same result is obtained when $\widetilde{C}_t$ is negative.

Although Equation (55) may be directly transformed with Equation (43), it is more convenient to consider $\phi(c_t)$ first. Equation (43) shows that the distribution of c_t about $\widetilde{C}_t$ must be the same as the distribution of c° about $\widetilde{C}^\circ$, $\phi(c_t) \rightarrow \phi(c^\circ)$. If it is assumed that c° is normally distributed about C°, then c_t is normally distributed about $\widetilde{C}_t$, as assumed by Hawthorne, et al. (8).

When this normal distribution is substituted into Equation (55), the integration may be carried out (8) and the result can be reduced to

$$b\frac{\widetilde{C}_1}{\widetilde{C}_t} = 1 + \frac{\gamma_t}{\sqrt{2}}\,ierfc\left(\frac{1}{\sqrt{2}\,\gamma_t}\right) =$$

$$1 + g(\gamma_t) \quad (56)$$

and *ierfc* is the first integral of the complimentary error function for which tables are available (3).

It follows from Equations (56) and (5) that

$$a\,\widetilde{C}_2/\widetilde{C}_t = g(\gamma_t) \quad (57)$$

So no additional information is to be gained by consideration of the situation $c_t < -\widetilde{C}_t$.

Equations (56) and (57) show that the time average concentration profiles of the individual species, and thus the average rate of reaction, depend upon both $\widetilde{C}_t$ and γ_t.

Hawthorne, et al. (8) measured $\widetilde{C}_1$ and $\widetilde{C}_2$ in turbulent combustion and from the measurements computed what is essentially γ_t, which they termed the "unmixedness factor." In this study the object is to relate $\widetilde{C}_1$ and $\widetilde{C}_2$ to the behavior of the equivalent nonreacting system.

It has been seen that $\widetilde{C}_t$ can be determined from information concerning the time average behavior of nonreacting systems. It remains necessary to relate γ_t to the relative intensity of the concentration fluctuations in nonreacting systems in order to determine the time average concentrations of species 1 and 2.

Reaction Between a Turbulent Fluid and a Species Dissolving From a Tube Wall

It is worth considering first the problem treated earlier in which a turbulent fluid with concentration C_{20} enters a tube whose wall is held at C_{1I}. The instantaneous solution to the equivalent mass transfer problem for a given Reynolds number must be

$$\frac{\widetilde{C}^\circ + c^\circ - C^\circ_0}{C^\circ_I - C^\circ_0} = f_1(Z_1, \ldots Z_4) \quad (58)$$

and solving for c°, squaring, and using

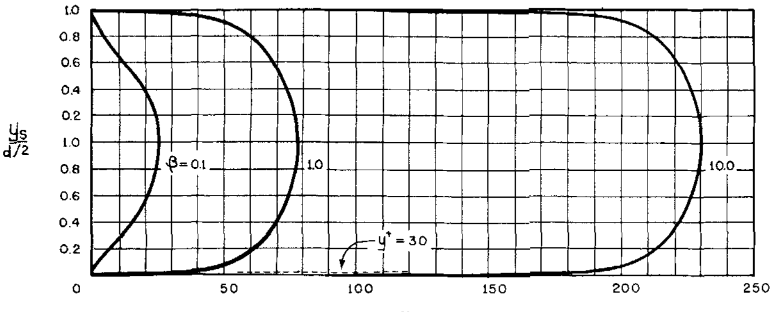

Fig. 5. Surfaces at which $\tilde{C}_t = 0$. Turbulent flow. $N_{Sc} = 1$, $N_{Re} = 40{,}000$.

the condition that $\tilde{f}_1 = f(Z_1, Z_2)$ in Equation (45) one gets

$$\frac{\Gamma^{o2}}{(C^o_{\mathrm{I}} - C^o_0)^2} = \tilde{f}_1^2 - f^2 = f_2^2\,(Z_1, Z_2) \tag{59}$$

When the reaction is present, the transform of Equation (59) divided into Equation (59) yields

$$\frac{\Gamma_t}{\Gamma^o} = \left(\frac{C_{t\mathrm{I}} - C_{t0}}{C^o_{\mathrm{I}} - C^o_0}\right) \tag{60}$$

If C^o_0 is taken as zero, C^o_{I} as $C_{1\mathrm{I}}$, and $C_{t\mathrm{I}}$ and C_{t0} are identified as before, Equation (60) becomes

$$\frac{\Gamma_t}{\Gamma^o} = b\,(1 + \beta) \tag{61}$$

and Γ^o becomes the root-mean-square fluctuation which would exist at the point of interest if no species 2 were present in the inlet stream, and all other conditions were the same as in the reaction problem.

From Equations (61), (45), and (46)

$$\gamma_t = \gamma^o \frac{f(Z_1, Z_2)}{f(Z_1, Z_2) - \beta/(1 + \beta)} \tag{62}$$

The time average concentration profiles of the individual species are now completely defined by Equations (56), (57), (62), and (46). In addition to a knowledge of $f(Z_1, Z_2)$, the time average behavior γ^o and the relative intensity of the fluctuations in the equivalent nonreacting system in which C^o_0 is zero and C^o_{I} is $C_{1\mathrm{I}}$ must be known.

The equations for $\tilde{C}_1$ and $\tilde{C}_2$ when $\tilde{C}_t$ is zero take on particularly simple forms, for then Equation (56) reduces to

$$\tilde{C}_1 = \frac{1}{\sqrt{2\pi}b}\Gamma_t \tag{63}$$

and with Equation (61) this becomes

$$\tilde{C}_1 = \frac{(1 + \beta)}{\sqrt{2\pi}}\gamma^o\,\tilde{C}^o \tag{64}$$

Using Equation (45) with $C^o_0 = 0$ and $C^o_{\mathrm{I}} = C_{1\mathrm{I}}$ and Equation (64) one obtains

$$\tilde{C}_1/C_{1\mathrm{I}} = \frac{(1 + \beta)}{\sqrt{2\pi}}\gamma^o f(Z_1, Z_2) \tag{65}$$

When $\tilde{C}_t = 0$, $f(Z_1, Z_2)$ is merely $\beta/(\beta + 1)$ from Equation (46), so Equation (65) becomes

$$\tilde{C}_1/C_{1\mathrm{I}} = \beta\,\gamma^o/\sqrt{2\pi} \tag{66}$$

and since $\tilde{C}_2 = \frac{b}{a}\tilde{C}_1$

$$\tilde{C}_2/C_{20} = \gamma^o/\sqrt{2\pi} \tag{67}$$

γ^o must approach zero at the wall and at large values of x/d, and it is probably small compared with one over most of the tube. As β gets large, the $\tilde{C}_t = 0$ surfaces move towards the wall and to large values of x/d (Figure 5), regions in which γ^o is approaching zero. Mixing length arguments indicate that the quantity $\beta\,\gamma^o$ on the $\tilde{C}_t = 0$ surface is generally less than the relative turbulence intensity. Consequently the surfaces drawn in Figure 5 are surfaces on which $\tilde{C}_1/C_{1\mathrm{I}}$ and $\tilde{C}_2/C_{20}$ are small numbers rather than zero as in the comparable laminar case.

The axial distance given by Equation (50) corresponds to that distance at which the center line concentrations have the above values. Since the center line concentration of species 2 is the maximum value across the tube, the fractional conversion of species 2 at axial distances given by Equation (50) must, from Equation (67), satisfy the condition

$$F_2 > 1 - \gamma^o/\sqrt{2\pi} \tag{68}$$

The above problem has been considered in detail because of its relative simplicity, but the concepts and techniques which have been used apply equally well to the problem of a reaction between a solid and a turbulent fluid in more complicated geometries. In the next section it will be shown that they can also be applied to the reaction between two miscible fluids.

Reaction Between Two Miscible Fluids

Reactions are commonly carried out by mixing a stream of species 1 with a stream of species 2, either in a stirred tank or in a conduit with turbulent flow. The general problem is to determine the time average concentrations of the reactants as a function of position (or time), given the turbulent field and the kinetics of the reaction.

Beek and Miller (*1*) have discussed the general design problem from a mass transfer viewpoint. Kramers (*11*) has reported some measurements, and Corrsin (*4*) has carried out a preliminary analysis of a related problem.

In the present analysis rapid reactions are considered, so the kinetics drop out and the parameters left are the stoichiometry and the turbulent field. Although this analysis is limited essentially to the case in which both

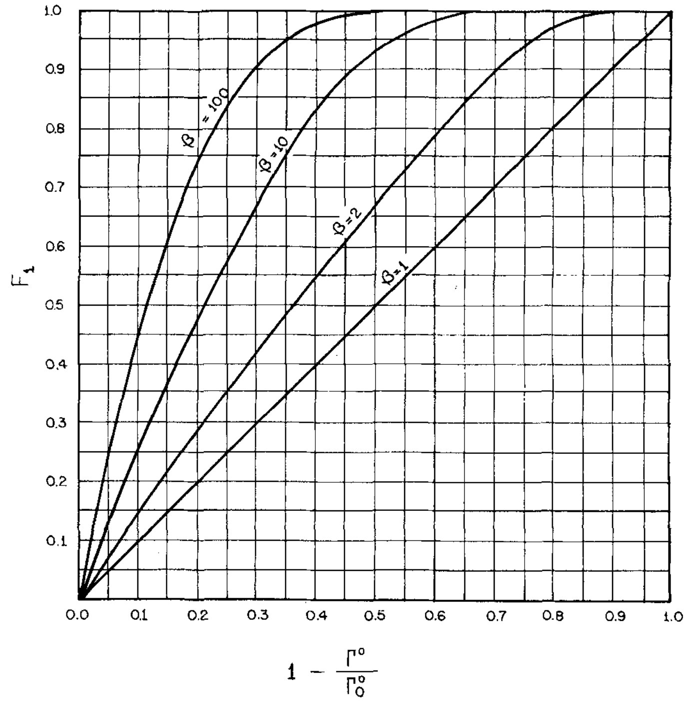

Fig. 6. Fractional conversion as a function of accomplished mixing.

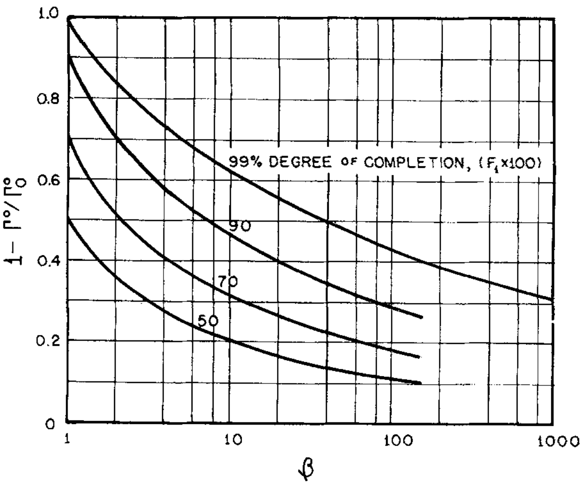

Fig. 7. Accomplished mixing vs. β for fixed degree of completion of reaction.

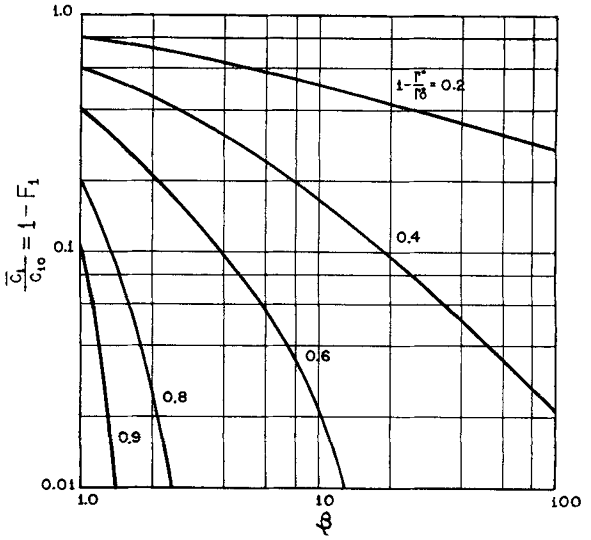

Fig. 8. Bulk composition vs. β for fixed values of accomplished mixing.

streams are the same inert solvent with one dilute reactant initially dissolved in one stream and one in the other, the results can also be considered to be an approximation to the case in which two miscible streams react with each other.

Consider first the equivalent mass transfer problem in which a fluid stream (or streams) containing a species at a concentration C°_0 is brought together in a mixing device with a stream of the same fluid which contains species 1 at a concentration C°_1; the mixing may take place in a tube or in a stirred tank. For convenience the following discussion will be in terms of a steady state tubular reactor. If Z_1 is the reduced distance measured downstream from the point of contact of the two streams and Z_2 is reduced radial distance, Equation (58) gives the instantaneous concentrations for fixed geometry and hydrodynamics (f_1 of course depends on the mixing device). The development is similar to the above, and given the root-mean-square concentration fluctuations in the nonreacting problem the time average concentration profiles and fractional conversion can be computed. In this case γ° can be much larger than in the earlier case.

The calculations are simplified if the mixture is uniform on a coarse scale (*11*), for then in the nonreacting problem

$$\tilde{C}^\circ = \bar{C}^\circ = \bar{C}^\circ_0 \qquad (69)$$

This type of uniformity is obtained by evenly dispersing one fluid in another. Molecular diffusion will eventually result in homogeneity on a molecular scale. If the turbulence itself is homogeneous, then Γ° depends only upon Z_1.

If Equation (59) is written at some reference plane Z_{10}, say the distance at which uniformity on a coarse scale is attained, and this equation is divided into Equation (59)

$$\Gamma^\circ/\Gamma^\circ_0 = f_2(Z_1)/f_2(Z_{10}) \qquad (70)$$

The subscript 0 in this and subsequent equations now refers to conditions at the Z_{10} plane. At this plane the concentrations of species 1 and 2 are C_{10} and C_{20}, respectively.

Beek and Miller calculated $\Gamma^\circ/\Gamma^\circ_0$ in a tubular reactor assuming homogeneous isotropic turbulence and showed how these mixing results can be used as a guide in designing reactors (*1*). The next step which is needed is to relate the conversion to $\Gamma^\circ/\Gamma^\circ_0$, and this step is carried out below for the limiting case of fast reactions in a dilute system with equal diffusivities.

When a reaction is present and the mixture is uniform on a coarse scale at Z_{10}, the transform of Equation (70) gives

$$\Gamma_t/\Gamma_{t0} = \Gamma^\circ/\Gamma^\circ_0 \qquad (71)$$

and

$$\tilde{C}_t = \bar{C}_t = \bar{C}_{t0} \qquad (72a)$$

$$\tilde{C}_1(Z_1) = \bar{C}_1(Z_1) \qquad (72b)$$

$$\tilde{C}_2(Z_1) = \bar{C}_2(Z_1) \qquad (72c)$$

Since C_t is conserved by the reacaction and neither species 1 nor 2 can leave through the tube walls, the last equality of Equation (72*a*) is valid even if the mixture is nonuniform.

When the two streams enter in stoichiometric proportions, $\bar{C}_{t0} = 0$, and for $Z_1 > Z_{10}$, $\tilde{C}_t = 0$. Then Equation (63) is valid and with Equations (71) and (72*b*)

$$\bar{C}_1/\bar{C}_{10} = \Gamma^\circ/\Gamma^\circ_0 \qquad (73)$$

so the factional conversion measured from Z_{10} is equal to the accomplished mixing:

$$F_1 = F_2 = 1 - \Gamma^\circ/\Gamma^\circ_0 \qquad (74)$$

When the inlet streams are not in stoichiometric proportions, Equation (56) must be used in place of (63). With Equation (72) and the definition of $\bar{C}_{t0}$ Equation (56) gives for the fractional conversion

$$F_1 = 1 + (\beta - 1)(1 + g(\gamma_t)) \qquad (75)$$

Here β corresponds to the bulk average concentrations at Z_{10}. Also at Z_{10}, $F_1 = 0$, so

$$g(\gamma_{t0}) = \beta/(1-\beta) \qquad (76)$$

and

$$\gamma_{t0} = \Gamma_{t0}/\bar{C}_{t0} = \Gamma_{t0}/\bar{C}_t \qquad (77)$$

Then γ_t can be written as

$$\gamma_t = \gamma_{t0}\Gamma_t/\Gamma_{t0} = \gamma_{t0}\Gamma^\circ/\Gamma^\circ_0 \qquad (78)$$

so Equation (75) becomes

$$F_1 = 1 + (\beta - 1)\cdot[1 + g(\gamma_{t0}\Gamma^\circ/\Gamma^\circ_0)] \qquad (79)$$

Equations (76) and (79) together with Equation (74) were used to compute the fractional conversion curves shown in Figure 6. These curves give F_1 for $\beta \geqq 1$. Since there is no distinction between species 1 and 2, in using the graph species 1 is to be taken as the limiting reactant and $F_2(\beta) = F_1(\beta)/\beta$. F_1 is also the degree of completion of the reaction. The flow rates and composition of the unmixed streams do not appear explicitly in the above equations. They are of course implicit in the parameters.

The results relate the conversion to the accomplished mixing; to obtain the conversion as a function of distance the turbulence field must be specified and the mass transfer problem of relating the accomplished mixing to distance must be solved. The Reynolds and Schmidt numbers will appear only in this latter relationship. Beek and Miller's results (*1*) could be used for example to estimate conversion-distance curves for their conditions.

The degree of completion of the reaction has been seen to be equal to the accomplished mixing when the mixture is stoichiometric, $\beta = 1$. Figure 6 shows that an excess of one component increases the degree of completion of the reaction, as would be expected, and an increase in β when β is close to 1 is more effective than when β is larger.

The cross plot of the results in Figure 7 gives the accomplished mixing required to give various degrees of completion as a function of β. At a 99% completion level the accomplished mixing must be 0.99 for a β of 1, but only 0.83 for a β of 2. Since the distance required to change $(1 - \Gamma^{o}/\Gamma^{o}_{0})$ a given amount naturally increases as the quantity approaches 1, the fractional decrease in length obtained by increasing β would be expected to be significant. When one uses Beek and Miller's results for example, for both gases and liquids the change of β from 1 to 2 would approximately halve the length required for 99% completion of the reaction. A change of β from 1 to 10 however would only decrease the length a factor of 3.

Since in a given apparatus the accomplished mixing is a unique function of length for fixed Reynolds and Schmidt numbers, the effect of β on the reaction at a fixed length can be determined directly from Figure 6. Thus the lines of constant $(1 - \Gamma^{o}/\Gamma^{o}_{0})$ shown in Figure 8 are lines of constant position. At the position which corresponds to 20% accomplished mixing a change of β from 1 to 10 decreases $\overline{C}_1/C_{10}$ from 0.8 to 0.52; at the 60% mixing point this change in β drops $\overline{C}_1/C_{10}$ from 0.4 to 0.2, while at the 90% mixing point a change of β from 1 to only 1.4 changes this reduced concentration from 0.1 to 0.01. Consequently increasing the concentration of one species in order to increase the conversion of the other is most effective when a high conversion is required and the mixture is initially close to stoichiometric.

CONCLUSIONS

The problem of mass transfer with rapid, irreversible, homogeneous reactions between dilute reactants with equal diffusivities is solved if the equivalent mass transfer problem without reaction is solved.

The rate of transfer of a reacting species can be determined from the mass transfer coefficient in the equivalent nonreacting system without the necessity of assuming any hydrodynamic model.

The time average behavior of the reacting system can be deduced if the root-mean-square concentrations fluctuations as well as the time average behavior are known in the equivalent nonreacting system.

NOTATION

a,b = stoichiometric coefficients
c = concentration fluctuation, $C - \widetilde{C}$
c_t = $b\,c_1 - a\,c_2 = C_t - \widetilde{C}_t$
C = concentration
C_t = $b\,C_1 - a\,C_2$
C_t^* = $b\,C_1^*$
d = diameter
D = diffusivity
f, f_1, f_2, G = functional symbols
f' = Fanning friction factor
g = function defined by Equation (56)
F = fractional conversion
h = distance from midpoint to surface
$ierfc$ = first integral of complimentary error function
k = individual mass transfer coefficient
k_e = effective reaction velocity constant
k_r = reaction factor
K = over-all mass transfer coefficient
m = equilibrium constant
N = molar flux at boundary
$\underline{N}$ = molar flux vector with respect to a fixed coordinate system
N_t = $b\,N_1 - a\,N_2$
$\underline{N}_t$ = $b\,\underline{N}_1 - a\,\underline{N}_2$
$\overline{N}_1^{o\prime}$ = molar flux defined by Equation (25)
N_{Gz} = Graetz number
N_{Pe} = Peclet number
N_{Re} = Reynolds number
Q = volumetric flow rate
r = rate of reaction
S = scale factor
u = velocity in x direction
U = mean velocity
$\underline{V}$ = velocity vector
$\underline{v}$ = velocity fluctuation vector
w = weighting function
x = axial distance
y = distance from wall
Z = dimensionless distance or time coordinate

Greek Letters

β = stoichiometric parameter, $a\,C_{20}/b\,C_{1I}$ or $a\,\overline{C}_{20}/b\,\overline{C}_{10}$
β^* = $a\,C_{20}/b\,C_1^*$
β_o^* = $a\,C_{20}/b\,C^*_{10}$
γ = relative intensity of concentration fluctuation, $\Gamma/\widetilde{C}$
γ_t = $\Gamma_t/\widetilde{\overline{C}}_t$
Γ = root-mean-square concentration fluctuation, $\sqrt{\widetilde{c^2}}$
Γ_t = $\sqrt{\widetilde{c_t^2}}$
η = external concentration
θ = time
ϕ = distribution function
Φ = reduced center line concentration $= \dfrac{C^{o}_{cL} - C^{o}_{I}}{\overline{C}^{o} - C^{o}_{I}}$

Subscripts

$1, \ldots 4$ = species or coordinates
f = complete reaction
i,j = indices referring to species or coordinates
n = number of coordinates
s = reaction surface
I, 0 = boundary I or 0
cL = center line

Superscripts

o = absence of reaction
$*$ = equilibrium value
— = bulk or space average
∼ = time average
$D/D\theta$ = substantial derivative
∇ = gradient
∇^2 = Laplacian operator

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