Turbulent Reactive Mixing of Reversible Reactions

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The convective diffusion equations were used to obtain mean concentrations and concentration fluctuation covariances during turbulent reactive mixing of fast reversible reactions. The mean concentrations are directly applicable to reactions that are much faster than the mixing, while the covariances can serve as a first approximation closure for slower reactions. The covariances should also be useful for testing more general closure models. Numerical examples are presented for the simple mass-action rate law with stoichiometric coefficients of unity.

Introduction

Although there are no solutions to the *unaveraged turbulent* convective diffusion equations, it *is* possible to relate the solution with certain chemical reactions to the solution without reaction. These *mapping* solutions can be obtained only for some kinds of reactions that are either much faster or much slower than the mixing, but because of the wealth of information that is available for nonreacting systems, they are quite useful. Mapping solutions have been obtained for a single irreversible bimolecular reaction (Toor, 1962) and later extended to the reversible reaction of H^+ with OH^- (Li and Toor, 1986). This article further extends the method to a wider class of reversible reactions, relating conversion to mixing for instantaneous, reversible reactions, but probably of more value, maps the limits of the covariance terms—terms that are needed to close the convective diffusion equations for finite-speed reactions. These limiting values can be used as a first-approximation closure, as has been done with the irreversible solution (Singh, 1973; Bilger et al., 1991), but since they represent an example of complex chemistry, they can also be used to test closure models that may handle complex chemistry but have not yet been appropriately tested.

Mapping Reactants to Tracer

The following development can handle the reversible reaction

$$aA + bB = cC + dD \tag{1}$$

with the equilibrium relationship

$$K = \frac{A_e^a B_e^b}{C_e^c D_e^d}. \tag{2}$$

For simplicity we have assumed ideality and will carry out the development with all coefficients set equal to unity.

The basic solution that relates instantaneous reactant concentrations to an instantaneous tracer concentration depends *only upon the equilibrium relationship and stoichiometric coefficients*, but a rate expression is needed to obtain the covariances. It can be of the general form

$$r_A = r_A(A,B,C,D). \tag{3}$$

(The earlier reversible reaction, which Li and Toor (1986) solved (without obtaining the covariance terms), is the $H^+ + OH^-$ reaction—in which case all the coefficients are unity, except d, which is zero, and component C (water) is in large excess.)

Physical properties are taken as constant and, like the irreversible case, it is necessary to assume that the molecular diffusivities are all equal. There is no reason to expect this to be any more restrictive here than in the earlier case, where it appears to be of little significance (Mao and Toor, 1971; Bilger et al., 1991). With this assumption the present reactive mixing problem is described by

$$K = \frac{A_e B_e}{C_e D_e} \tag{4}$$

$$L(A) = L(B) = -L(C) = -L(D) = r_A \qquad (5)$$

$$L = \frac{\partial}{\partial t} + v \cdot \nabla - D\nabla^2. \qquad (6)$$

Three linear combinations of concentrations satisfy the same equation as a nonreacting tracer (A without the reaction)

$$L(A-B) = L(A+C) = L(A+D) = L(f) = 0, \qquad (7)$$

so these "conserved scalars" mix without reacting. Explicit relationships between the reacting and pure mixing problem can now be obtained for each conserved scalar by matching boundary conditions in much the same way as in the irreversible case where there is only one conserved scalar:

Mix feed streams 1 and 2 and observe that during the mixing the following nondimensional conserved scalar concentrations satisfy the same boundary conditions and differential equation, $L() = 0$. Hence they are everywhere equal to the concentration of the nonreacting tracer that enters in stream 1 with a concentration of unity, so

$$\frac{(A-B)-(A-B)_2}{(A-B)_1-(A-B)_2} = \frac{(A+C)-(A+C)_2}{(A+C)_1-(A+C)_2} = \frac{(A+D)-(A+D)_2}{(A+D)_1-(A+D)_2} = f. \qquad (8)$$

Since the two feed streams can contain various combinations of reactants, we again choose the simplest, mix stream 1 containing only A at concentration A_0 with stream 2 containing only B at B_0. Then Eq. 8 gives

$$\frac{A-B+B_0}{A_0+B_0} = \frac{A+C}{A_0} = \frac{A+D}{A_0} = f. \qquad (9)$$

The C and D concentrations are equal here because they are produced at the same rate and neither is present in the feed. With this simplification the nondimensional versions of Eq. 9 are

$$X_A - \beta X_B = (1+\beta)f - \beta \qquad (10)$$

$$X_A + X_C = f. \qquad (11)$$

Considering f as known, there are two equations with three unknowns. The third relationship is obtained by assuming the reaction to be fast enough compared to the mixing to be effectively at equilibrium. Then the third equation, the nondimensional form of Eq. 4, is

$$\frac{X_A X_B}{X_C^2} = \frac{K}{\beta}. \qquad (12)$$

(The subscripts indicating equilibrium have been dropped in the preceding and subsequent equations.)

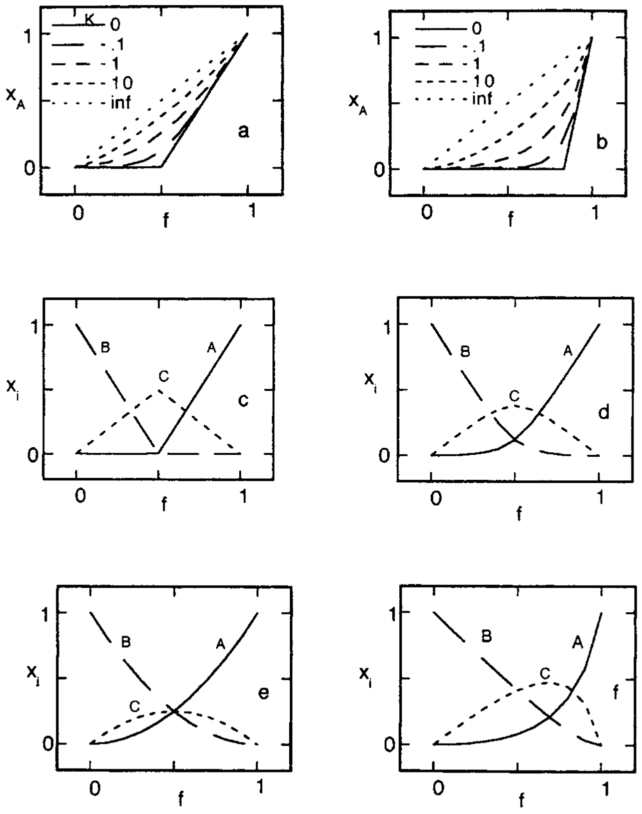

Figure 1. Reactant concentrations vs. tracer concentrations.

(a) $\beta = 1$; (b) $\beta = 5$; (c) $\beta = 1$, $K = 0$; (d) $\beta = 1$, $K = 0.1$; (e) $\beta = 1$, $K = 1$; (f) $\beta = 5$, $K = 1$.

Equations 10, 11 and 12 are the starting points for the remainder of this article. The solution to these equations is

$$X_A = \frac{Z \pm \sqrt{Z^2 + 4K(1-K)f^2}}{2(1-K)} \qquad (13a)$$

$$Z = (1+\beta-2K)f - \beta, \qquad (13b)$$

which together with Eqs. 10 and 11 relate the reactant concentrations to the tracer concentration.

Equations 10 through 13 also result from mixing a volume f of stream 1 with a volume $1-f$ of stream 2 and allowing the resulting mixture to come to both chemical and physical equilibrium. The viewpoint of Danckwerts (1957) that f is the local instantaneous volume fraction of the feedstream 1 is ambiguous, but leads to the correct result in a system with equal diffusivities, if f is understood to be the concentration of a passive tracer entering in stream 1 (Shenoy and Toor, 1988).

Equation 13 is plotted in Figure 1a and 1b for two values of β and various values of K, while the remainder of the figure maps the three reactant concentrations for selected values of β and K.

$K = 0$ is the irreversible limit (Toor, 1962; Bilger et al., 1991), where the reactants are totally segregated by the reaction

$$X_A = (1+\beta)f - \beta, \qquad f \geq \frac{\beta}{1+B} \tag{14a}$$

$$X_A = 0, \qquad f \leq \frac{\beta}{1+B}, \tag{14b}$$

and $X_A = f$ at the large K limit where no reaction occurs.

For $K = 1$ the reactant equations take on the simple forms,

$$X_A = \frac{f^2}{\beta - (\beta - 1)f} \tag{15}$$

$$X_B = \frac{\beta(1-f)^2}{\beta - (\beta - 1)f} \tag{16}$$

$$X_C = \frac{\beta f(1-f)}{\beta - (\beta - 1)f}. \tag{17}$$

Mean Reactant Concentrations

The mean concentrations also depend upon the mixing information that is contained in the PDF, since

$$\bar{X}_i = \int_0^1 X_i(f)\phi(f)\,df, \qquad i = A, \ldots, D. \tag{18}$$

In most cases Eq. 18 must be integrated numerically, but when $K = 1$ Eq. 15 allows $\bar{X}_A$ to be written in terms of the moments of the PDF,

$$\bar{X}_A = \frac{1}{\beta}\sum_{n=0}^{\infty}\left(\frac{\beta - 1}{\beta}\right)^n \overline{f^{n+2}}, \tag{19}$$

with $\bar{X}_B$ and $\bar{X}_C$ from Eqs. 10 and 11. If $\beta = 1$, particularly simple results follow in terms of the first two moments,

$$\bar{X}_A = \bar{f}^2 + \sigma^2 \tag{19a}$$

$$\bar{X}_B = (1 - \bar{f})^2 + \sigma^2 \tag{19b}$$

$$\bar{X}_C = \bar{f}(1 - \bar{f}) - \sigma^2. \tag{19c}$$

Rearrangement of the preceding equations gives the nice picture,

$$(\bar{X}_A - X_A(\bar{f})) = (\bar{X}_B - X_B(\bar{f})) = -(\bar{X}_C - X_C(\bar{f})) = \sigma^2, \tag{20}$$

the "unmixedness deficit" (the difference between the local mean concentration and the value that would exist at that point if the mixing were perfect, the *homogeneous mean value*) is merely the local *tracer variance* (the second moment around the mean of the PDF).

For all other cases we will use the beta distribution, which is a better representation than the normal distribution used for the irreversible reaction (Toor, 1962),

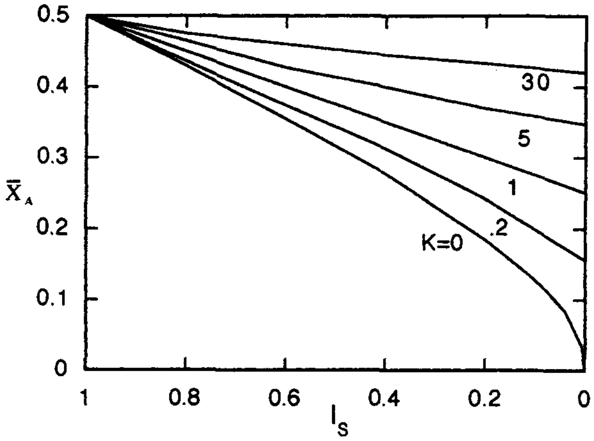

Figure 2. Mean concentration of reactant A vs. intensity of segregation, $\beta = 1$, $\bar{f} = 0.5$.

$$\phi(f) = \frac{\Gamma(p+q)}{\Gamma(p)\Gamma(q)} f^{p-1}(1-f)^{q-1} \tag{21a}$$

$$p = \bar{f}\{[\bar{f}(1-\bar{f})/\sigma^2] - 1\} = \bar{f}\left(\frac{1 - I_S}{I_S}\right) \tag{21b}$$

$$q = \frac{1 - \bar{f}}{\bar{f}}p. \tag{21c}$$

$\bar{X}_A$ is shown in Figures 2 through 5 for selected values of the parameters. They are drawn with the reactor inlet imagined to be on the left.

In the *absence* of molecular diffusion, the tracer variance and mean concentration are related by (Danckwerts, 1958; Toor, 1997),

$$\sigma_0^2 = \bar{f}(1 - \bar{f}), \tag{22}$$

so this is the *maximum* value of the variance at any $\bar{f}$, and at this maximum $\bar{X}_A = \bar{f}$. (Substituting Eq. 22 into Eq. 19a necessarily gives this result.)

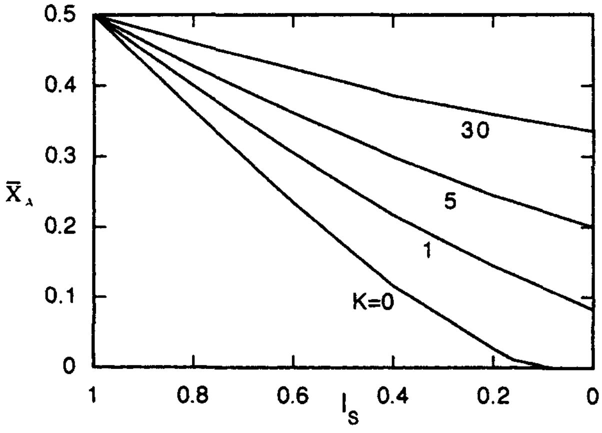

Figure 3. Mean concentration of reactant A vs. intensity of segregation, $\beta = 5$, $\bar{f} = 0.5$.

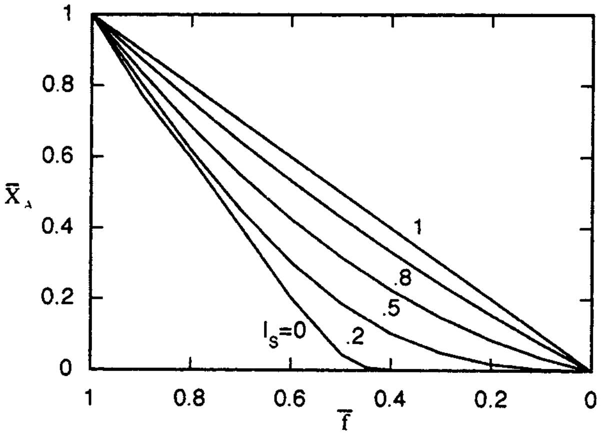

Figure 4. Mean concentration of reactant A vs. mean tracer concentration, $\beta = 1$, $K = 0$.

During reactive mixing the entering reactant concentrations decrease both by dilution and reaction, while the tracer concentration decreases by dilution alone. In an idealized one-dimensional reactor the coarse scale dilution occurs immediately (i.e., macromixing is instantaneous) followed by micromixing and reaction. Thus Figures 2 and 3 at $\bar{f} = 0.5$ represent an ideal one-dimensional reactor (or the centerline of a mixing layer) with equal flow rates of the two reactant streams in which $\bar{f}$ and $\bar{X}_A$ have been immediately diluted to 0.5. Then $\bar{f}$ remains constant, while $\bar{X}_A$ monotonically decreases as the tracer variance decays by molecular diffusion from its initial value of $\sigma_0^2 = 0.25$, reaching its final minimum value downstream when $\sigma^2 = 0$ and $f = \bar{f}$. Or in terms of the intensity of segregation, I_S decreases from its initial value of one to zero.

In reality the situation is more complicated, since dilution and reaction occur simultaneously. Consider, for example, a jet containing reactant A (with a passive tracer) entering a large tank of reactant B. As the A fluid mixes and reacts with the B fluid, $\bar{f}$ eventually decreases to zero from its initial value of 1, while σ^2 rises from its initial value of zero, reaches a maximum value (< 0.25), and finally decays to zero. Since σ^2 cannot exceed $\bar{f}(1-\bar{f})$, it is convenient to write Eq. 19a (shown in Figure 5) using the intensity of segregation,

$$\bar{X}_A = \bar{f}[\bar{f} + I_S(1-\bar{f})]. \tag{23}$$

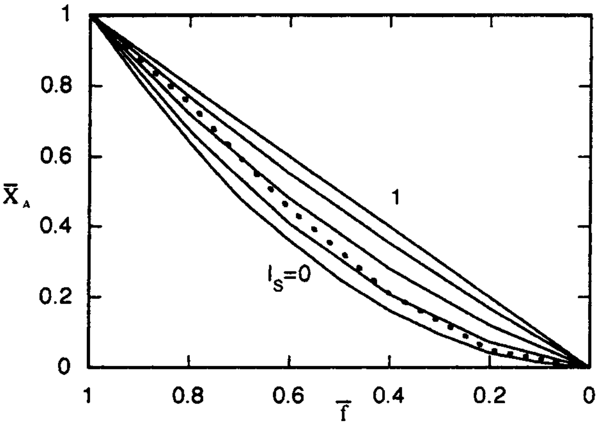

Figure 5. Mean concentration of reactant A vs. mean tracer concentration, $\beta = 1$, $K = 1$.

I_S = 0, 0.2, 0.5, 0.8; 1 ——; path · · · .

I_S decreases with molecular diffusion, so as the fluid moves downstream, $\bar{f}$ and I_S *both* decrease monotonically, as does $\bar{X}_A$, which approaches $\bar{f}^2$ as I_S approaches 0. The dotted line sketched in Figure 5 represents the path followed by a parcel of jet fluid that begins in the upper lefthand corner and ends up in the lower righthand corner.

The preceding results can be used as is to describe fast-enough reactions, but the covariance terms that are evaluated below may be of value for reactions that are not fast enough to be at equilibrium.

Covariance Terms

The results to this point do not depend upon the rate equation, but the covariances do. The obvious first choice at this point is the simple mass-action rate law

$$-r_A = k(AB - KCD), \tag{24}$$

so the nondimensional mean reaction rate is

$$-\hat{r}_A = \overline{X_A X_B} - \frac{K}{\beta}\overline{X_C X_D}, \tag{25}$$

which after separating into means and fluctuations and averaging becomes

$$-\hat{r}_A = \bar{X}_A \bar{X}_B + \sigma_{AB}^2 - \frac{K}{\beta}\left(\bar{X}_C \bar{X}_D + \sigma_{CD}^2\right), \tag{26}$$

where the covariances, the means of the fluctuation products, are given by

$$\sigma_{AB}^2 = \overline{X'_A X'_B}, \qquad \sigma_{CD}^2 = \overline{X'_C X'_D}. \tag{26a}$$

The closure problem of reactive mixing is how to model these and similar covariances when reaction rates are of the order of the mixing rates.

By arguments similar to those used for nonpremixed irreversible reaction (Vassilatos and Toor, 1965; Bourne and Toor, 1977) we expect σ_{AB}^2 to be negative, and because $X'_C = X'_D$, σ_{CD}^2 must be positive.

In anticipation of behavior similar to irreversible reaction (Toor, 1969; Leonard et al., 1995), we expect the covariances to be of the order of the tracer variance, so we define the following covariance/variance ratios (CV ratios):

$$R_{AB} = \frac{\sigma_{AB}^2}{\sigma^2}, \qquad R_{CD} = \frac{\sigma_{CD}^2}{\sigma^2}, \tag{27}$$

and write Eq. 26 as

$$-\hat{r}_A = \left(\bar{X}_A \bar{X}_B - \frac{K}{\beta}\bar{X}_C \bar{X}_D\right) + R_N \sigma^2, \tag{28}$$

where

$$R_N = R_{AB} - \frac{K}{\beta} R_{CD}. \qquad (29)$$

The first term on the right of Eq. 28 is the homogeneous mean rate to which is added the net effect of the two CV ratios—necessarily negative because of the nonpremixed feed—to give the actual mean rate.

Since we are at equilibrium R_N is evaluated from

$$R_N = \frac{\bar{X}_A \bar{X}_B - \frac{K}{\beta} \bar{X}_C \bar{X}_D}{\sigma^2}, \qquad (30)$$

since the mean concentrations can be obtained from Eqs. 10, 11, 13, and 18. We can write R_N in terms of A alone by using Eqs. 10 and 11,

$$R_N = \frac{1}{\beta \sigma^2}\left[(K-1)\bar{X}_A^2 + \bar{Z}\bar{X}_A + K\bar{f}^2\right] \qquad (31)$$

$$\bar{Z} = (1+\beta-2K)\bar{f} - \beta. \qquad (31a)$$

When $K = 0$, Eq. 29 reduces to the single CV ratio for irreversible reactions,

$$R_N = R_{AB} = -\frac{\bar{X}_A \bar{X}_B}{\sigma^2}, \qquad (32)$$

but for $K > 0$ the following equations determine the individual CV ratios,

$$R_{AB} = \frac{\overline{X_A X_B} - \bar{X}_A \bar{X}_B}{\sigma^2} \qquad (33)$$

$$R_{CD} = \frac{\overline{X_C X_D} - \bar{X}_C \bar{X}_D}{\sigma^2} \qquad (34)$$

and $\overline{X_A X_B}$ and $\overline{X_C X_D}$ are obtained by numerical integration of Eq. 18 using Eqs. 10, 11, 13 and 21.

However, irrespective of the PDF, when $\beta = 1$, the preceding equations give the simple relationship

$$R_{CD} = 1 + R_{AB}, \qquad (35)$$

and with Eq. 29,

$$R_N = -[K + (K-1)R_{AB}] = -[1 + (K-1)R_{CD}]. \qquad (36)$$

Equations 31 and 36 allow the CF ratios to be written in terms of reactant and tracer means so, still with $\beta = 1$,

$$R_{AB} = \frac{K}{1-K} + \frac{1}{\sigma^2(1-K)} \times\left[(K-1)\bar{X}_A^2 + [2(1-K)\bar{f} - 1]\bar{X}_A + K\bar{f}^2\right] \qquad (37)$$

with R_{CD} from Eq. 35.

And if K *also* equals one, from Eq. 36 the *net* effect of the microsegregation is the same as for the slow second-order irreversible bimolecular reaction (Toor, 1969),

$$R_N = -1. \qquad (38)$$

The individual CV ratios can be obtained for this situation *where both β and K are one* from Eqs. 15, 16, and 33,

$$R_{AB} = \frac{\left(\overline{f^4} - \bar{f}^4\right) - 2\left(\overline{f^3} - \bar{f}^3\right)}{\sigma^2} + 2\sigma_0^2 - \sigma^2. \qquad (39)$$

And evaluating the moments for a beta distribution (Wilkes, 1982) gives

$$R_{AB} = \frac{-\frac{(p+2q+3)(p+2)(p+1)p}{(p+q+3)(p+q+2)(p+q+1)(p+q)} + \bar{f}^3(2-\bar{f})}{\sigma^2} + 2\sigma_0^2 - \sigma^2, \qquad (40)$$

which for the important case of $\bar{f} = 0.5$ becomes

$$R_{AB} = \left(\frac{1}{4\sigma^2} + 1\right)\left(\frac{3}{4} - \sigma^2\right) - \frac{1}{8}\left(\frac{1}{\sigma^2} + 12\right)\left(\frac{3 + 12\sigma^2}{2 + 16\sigma^2}\right). \qquad (41)$$

Covariance Numbers

Examples of the CV ratios computed from the preceding equations are shown as functions of I_S in Figures 6 and 7

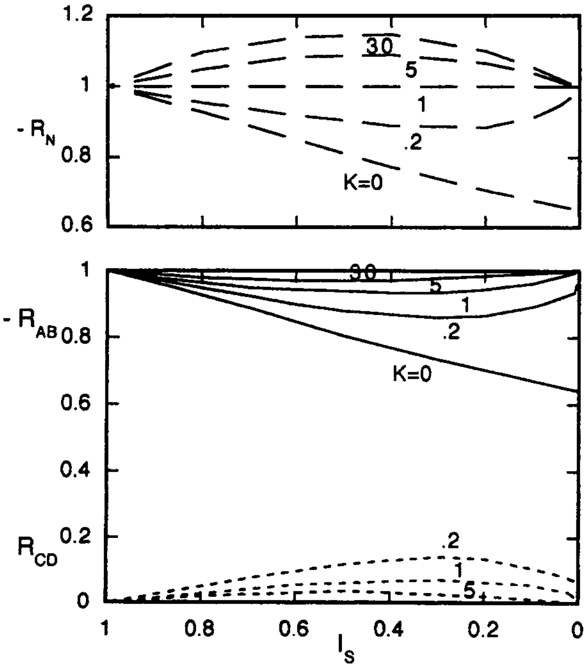

Figure 6. Covariance variance ratios vs. I_S, $\beta = 1$, $\bar{f} = 0.5$.

R_{AB} ——; R_{CD} - - - -; R_N— — —.

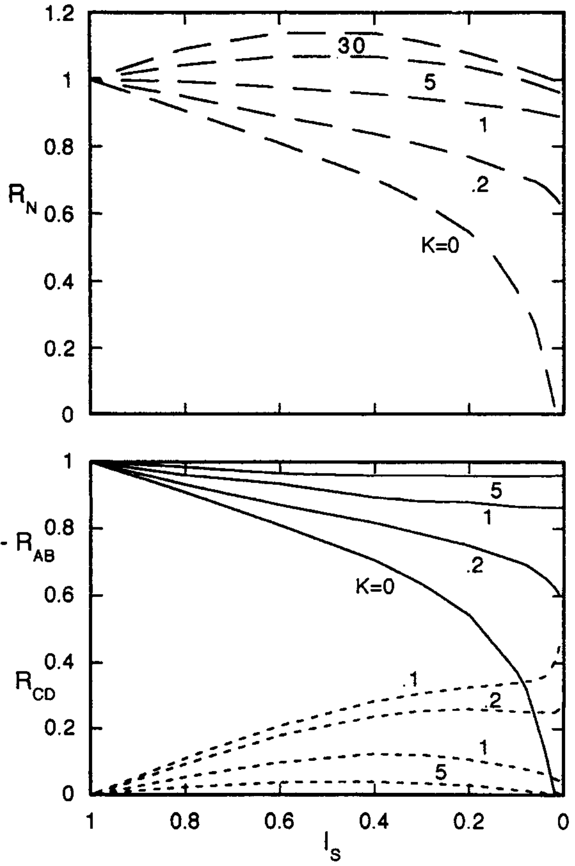

Figure 7. Covariance variance ratios vs. I_S, $\beta = 2$, $\bar{f} = 0.5$.

R_{AB} ——; R_{CD} ----; R_N— — —.

and as functions of $\bar{f}$ in Figures 8 through 11. As expected R_{AB} and R_N are always negative and R_{CD} is positive. $-R_N$ is always greater than $-R_{AB}$ since, compared to a homogeneous mixture, inhomogeneity slows down the forward step and speeds up the reverse step.

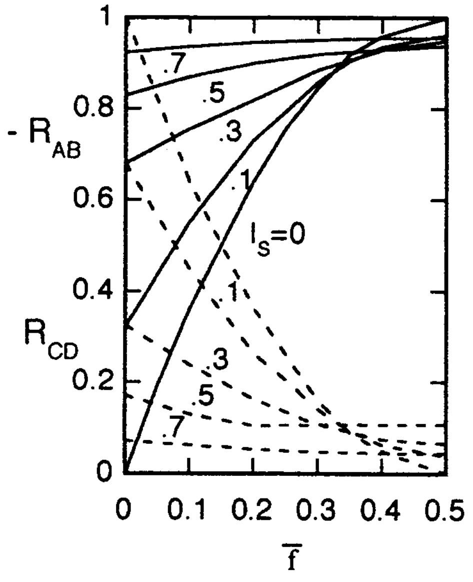

Figure 9. Covariance variance ratios vs. $\bar{f}$, $\beta = 1$, $K = 1$; $R_N = -1$ for these conditions.

Figure 6 like Figure 2 imagines a one-dimensional mixer, $-R_{AB}$ *and* $-R_N$ increase with K while R_{CD} decreases; *the more reversible the reaction the more it is slowed by unmixedness*. As I_S decreases in Figure 6, $-R_{AB}$ monotonically decreases from one for an irreversible reaction, but for the reversible reactions $-R_{AB}$ reaches a minimum not very far from one and approaches one again as I_S approaches zero. R_{CD} starts at zero, goes through a maximum, and eventually returns to zero. The net CV ratio in Figure 6, $-R_N$, has a minimum for $0 < K < 1$ and a maximum for $K > 1$.

The feed concentration ratio of 1 in Figure 6 has been changed to 2 in Figure 7. Many of the curve shapes change and the relative effect of unmixedness on reversible reactions is now more pronounced.

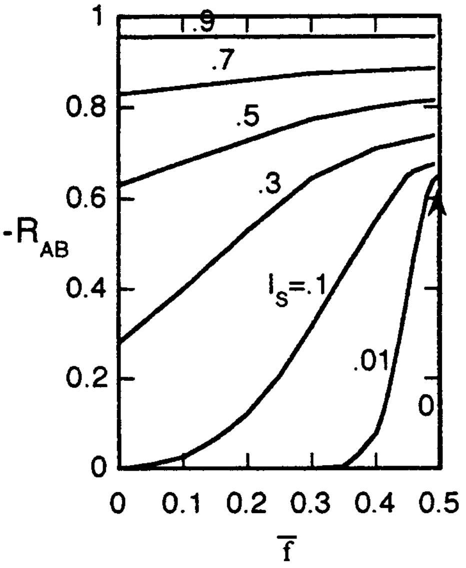

Figure 8. Covariance variance ratios vs. $\bar{f}$, $\beta = 1$, $K = 0$.

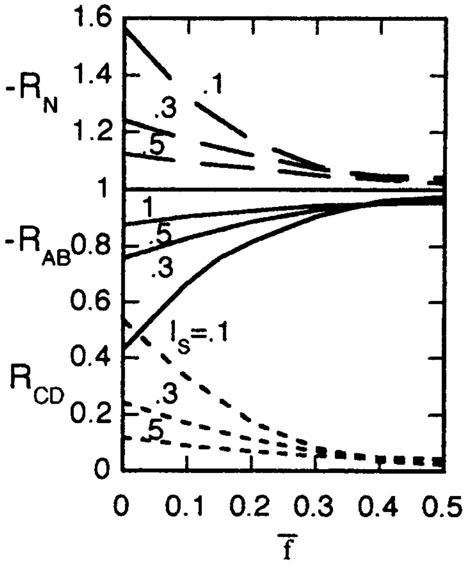

Figure 10. Covariance variance ratios vs. $\bar{f}$, $\beta = 1$, $K = 2$.

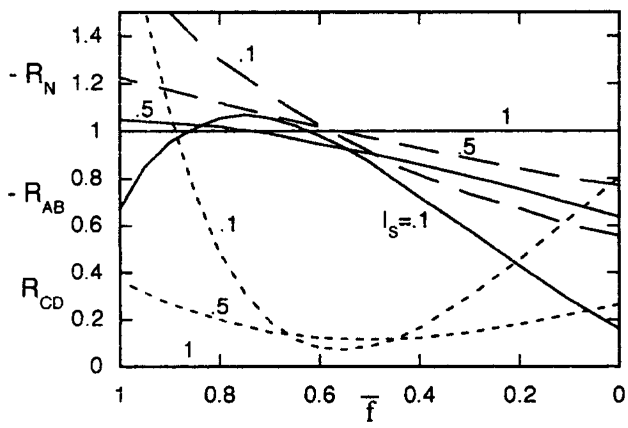

Figure 11. Covariance variance ratios vs. $\bar{f}$, $\beta = 2$, $K = 1$.

Figures 8 through 11 correspond to the multidimensional mixing of Figures 4 and 5, where both macro- and micromixing are simultaneous—$\bar{f}$ and σ^2 vary with position. Because all the CV ratios are symmetric around $\bar{f} = 0.5$ in Figures 8 to 10, the abscissas are only drawn from 0 to 0.5. For a fixed I_S, $-R_{AB}$ is a maximum and R_{CD} a minimum at $\bar{f} = 0.5$. $R_N = R_{AB}$ in Figure 8 and -1 in Figure 9. C and D do not exist in Figure 8. Although the CV ratios are greater for reversible than irreversible reactions, all the CV ratios are equal to or less than one in Figures 8 and 9, but when $K > 1$, as in Figure 10, where $K = 2$, $-R_N > 1$.

Symmetry is lost when $\beta > 1$ as in Figure 11, and $-R_N$ and R_{CD} both significantly exceed one when $\bar{f}$ is near one.

Discussion

The previous results can be extended by unconstraining the stoichiometric coefficients, and/or considering feedstreams other than pure reactants, and/or using other rate expressions. The rate law has no effect on the reactant concentrations since we are at equilibrium, but it does effect the covariances. Take as an example an irreversible reaction with a rate law of the form

$$-r_A = kA^2B. \tag{42}$$

Expanding, averaging, and nondimensionalizing gives

$$-\hat{\bar{r}}_A = \bar{X}_A^2\bar{X}_B + R_{A^2B}\sigma^2, \tag{43}$$

where

$$R_{A^2B} = -\frac{\bar{X}_A^2\bar{X}_B}{\sigma^2} = 2\bar{X}_A\frac{\sigma_{AB}^2}{\sigma^2} + \bar{X}_B\frac{\sigma_{AA}^2}{\sigma^2} + \frac{\sigma_{A^2B}^2}{\sigma^2}. \tag{44}$$

Although R_{A^2B} is now more complicated than earlier, it is readily evaluated because $\bar{X}_A$ and $\bar{X}_B$ are unchanged.

The other changes will modify Eqs. 10 through 13, requiring further integrations for mean concentrations and variances, but the results should handle any reversible reactions that are fast enough to be close to equilibrium during mixing. Although the beta distribution has been used for the PDF, this can also be replaced by a more appropriate one if necessary.

The covariances have been determined for two reasons. First, at present, other than one-dimensional, there are no suitable data with complex chemistry that can be used to test closure models. These reversible reactions represent a kind of complex chemistry, so they should be useful in testing such models, particularly because of the richness of the behavior of the covariances.

Second, the covariances evaluated here for very fast reactions can probably be used as a first approximation closure for slower reversible reactions without great error, because as intrinsic reaction rates decrease relative to mixing rates, and these covariance values become less accurate, reaction rates become less sensitive to them. Indeed for the special case of $\beta = K = 1$, where $R_N = -1$ for both very slow and very fast reactions, a reasonable hypothesis is that it is -1 for all reaction speeds, an extension of an earlier hypothesis (Toor, 1969) to this particular reversible reaction.

Acknowledgment

The author is indebted to Joseph Kadane and Arthur Westerberg for helpful advice.

Notation

A,B,C,D = species or species concentrations
a,b,c,d = stoichiometric coefficients
D = diffusivity
f = nondimensional tracer concentration
$I_s = \sigma^2/\sigma_0^2$
k = forward rate constant
K = equilibrium constant
L = operator defined by Eq. 6
p,q = coefficients of beta distribution, Eq. 21
r_A = rate of reaction of A
R_N = net covariance/variance (CV) ratio, Eq. 29
t = time
v = velocity
$X_A = A/A_0$
$X_B = B/B_0$
$X_C = C/A_0$
$X_D = D/A_0$
β = feed concentration ratio $= C_{B_0}/C_{A_0}$
ϕ = probability density function, PDF
σ^2 = tracer variance $= \bar{f}'^2$
σ_0^2 = tracer variance in absence of diffusion, Eq. 22
$\sigma_{AA}^2 = \overline{X_A' X_A'}$
$\sigma_{AAB}^2 = \overline{X_A' X_A' X_B'}$

Subscripts and superscripts

e = equilibrium
i = species A, B, C, D
$\bar{\ }$ = time or ensemble mean
$'$ = fluctuation

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Manuscript received May 28, 1996, and revision received Aug. 29, 1996.