Effect of Chemical Reactions on Turbulent Diffusivities

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A mixing length model describes the recently observed (Bilger et al., 1991) effect of chemical reactions on turbulent diffusivities. From this viewpoint, the coupling between microscale reactions and macroscale mixing occurs because the reaction rate is a quasitransferable property which causes more reaction in fluctuating fluid lumps leaving regions of high rate than low rate. The net effect, which raises or lowers reactant diffusivities, depends on a characteristic macromixing time and the rate at which the mean reaction rate changes with mean concentration:

$$\frac{\epsilon_A}{\epsilon} = 1 - \tau_M \frac{d\bar{r}_A}{d\bar{C}_A}$$

The model shows correct qualitative behavior and appears to agree quantitatively with much of the existing data. It adds an apparent flux, $\epsilon \tau_M (d\bar{r}_A/dy)$, to the convective diffusion equations which can enhance or hinder the normal turbulent fluxes, but is likely to have little effect on the mean concentration field.

Introduction

Two kinds of closure problems arise when the convective diffusion equations for a dilute, isothermal reacting Fickian system with nonlinear chemistry are averaged to give:

$$\frac{\partial \bar{C}_i}{\partial t} + \underline{\bar{v}} \cdot \nabla \bar{C}_i = -\nabla \cdot \underline{\bar{\phi}}_i - \bar{r}_i, \quad i = A, B, \ldots \tag{1a}$$

$$\underline{\bar{\phi}}_i = -D_i \nabla \bar{C}_i + \overline{\underline{v}' C_i'}, \quad i = A, B, \ldots \tag{1b}$$

The closure problem in the reaction term follows from nonlinear chemical rate laws which lead to correlated concentration covariance terms—terms which decrease reaction rates in nonpremixed systems. These *micromixing* (or more accurately, *microsegregation*) effects have been studied in one-dimensional systems where they can have a very large effect on reaction rates and where models have been developed to handle them in such systems (Harada et al., 1962; Toor, 1962, 1969; Vassilatos and Toor, 1965; Kattan and Adler, 1967; Mao and Toor, 1970; Treleaven and Tobgy, 1972; Mehta and Tarbell, 1983; Li and Toor, 1986; Baldyga and Bourne, 1989; Dutta and Tarbell, 1989).

On the other hand, the velocity-concentration covariance term, which has little effect in one-dimensional systems, is more likely to dominate multidimensional systems like jets and wakes (Hanks, 1991). This macromixing term has a long history in nonreacting systems where it has been usefully modeled with a turbulent diffusivity (Taylor, 1915; Hinze, 1959; Tennekes and Lumley, 1983). Because it apparently has the same significance in reacting as well as nonreacting systems, it is tempting to treat it with the normal turbulent diffusivity, that is, to assume that the reaction does not affect the turbulent diffusivity. This is probably true with slow enough reactions, but is less likely with faster reactions—reactions whose half-lives are comparable to or less than the integral time scale (Corrsin, 1974; Borghi, 1988; Klimenko, 1992), and recent experiments confirm this (Bilger et al., 1991). These experiments, which show a significant effect of faster reactions on turbulent diffusivities, cause an additional closure problem. A way around this coupling of the reaction with the turbulent diffusivity (micro-macro coupling) is to dispense with both terms and recognize that both effects are different manifestations of imperfect mixing, as is done in recent conditional mean methods (Klimenko, 1990; Bilger, 1991). The alternative, used earlier

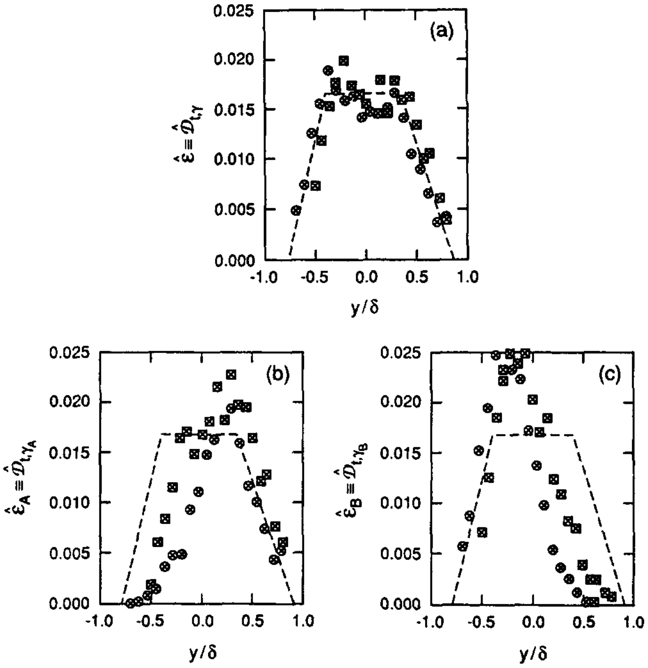

Figure 1. Turbulent diffusivity data from Figure 15 of Bilger et al. (1991).

⊗, $N_D = 1.81$; ⊠, $N_D = 0.3$. Same dashed line drawn on each figure for orientation. (a) Nonreacting; (b) species A; (c) species B.

by Borghi and Dutoya (1978) and adopted here, is to accept the coupling and relate turbulent diffusivities in a reacting system to both the reaction and the turbulent diffusivities in the analogous nonreacting (passive) system.

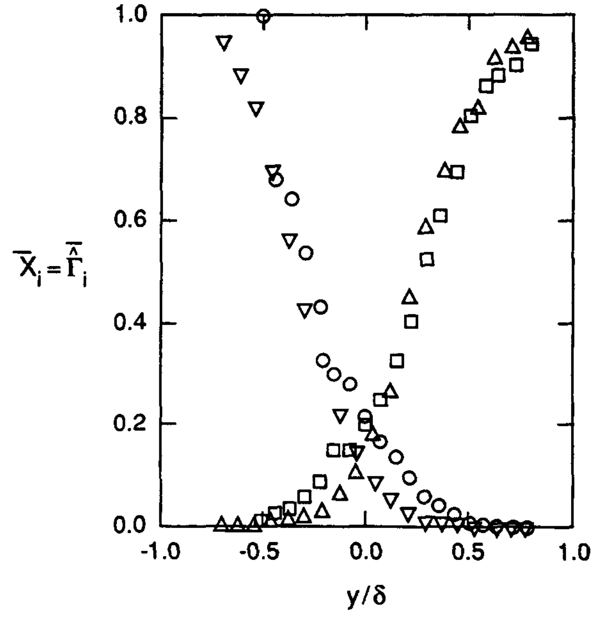

Figure 2. Mean reactant concentration profiles from Figure 8 of Bilger et al. (1991).

$N_D = 1.81$: △, species A; ▽, species B. $N_D = 0.3$: □, species A; ○, species B.

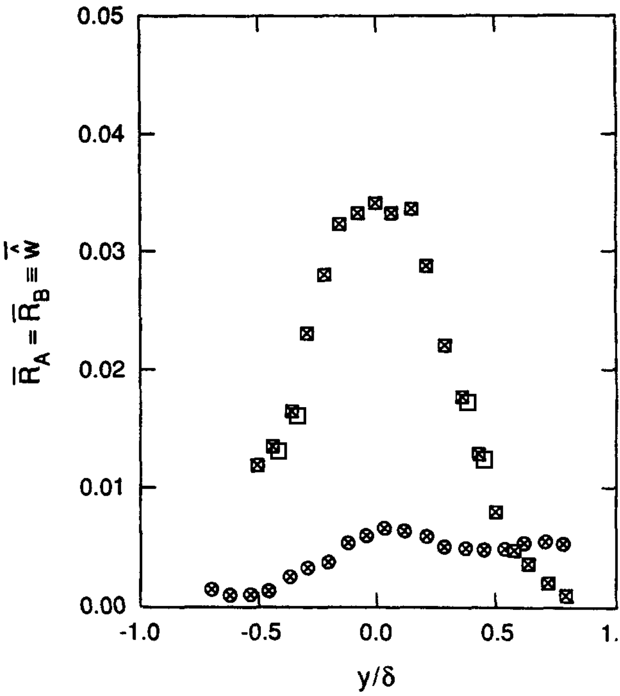

Figure 3. Mean reaction rate vs. position from Bilger et al. (Figure 18).

⊠, $N_D = 0.3$; ⊗, $N_D = 1.81$.

The Experiment

Bilger et al. (1991) studied the isothermal reaction between nonpremixed dilute nitric oxide (reactant A) and dilute ozone (reactant B) behind a splitter plate with grid-generated turbulence. With Damköhler numbers of 0.3 and 1.8, they deduced turbulent diffusivities for the reactants as well as the passive nonreacting value and their nondimensional diffusivity results for a Reynolds number (based on grid mesh and mean velocity) of 11,700 are reproduced in Figure 1. Their nondimensional mean concentration and reaction rate data are also reproduced in Figures 2 and 3. Both their nomenclature as well as that used here are shown. All the data are 21 grid spacings downstream. They pointed out that the turbulent diffusivities for the reactants "are substantially higher than those for the mixture fraction on the side of the layer from which the reactant comes but are much lower on the other side. In this latter region, the diffusivities also show a dependence on N_D with values for high N_D being smaller than those for low N_D. (The effects are seen more clearly in Figures 4 through 7.)

It was subsequently shown that for the particular chemistry which was used, $A + nB \rightarrow$ products and with equal molecular diffusivities of reactants (Toor, 1991), that the three turbulent diffusivities are related by:

$$(\underline{\epsilon}_A - \underline{\epsilon}) \cdot \nabla \overline{X}_A = \beta(\underline{\epsilon}_B - \underline{\epsilon}) \cdot \nabla \overline{X}_B \qquad (2)$$

Equation 2 is qualitatively consistent with this nonpremixed experiment where the gradients are of opposite sign although, as will be seen, some of the data do not quantitatively satisfy the equation. Although Eq. 2 *does* suggest that the individual reactant turbulent diffusivities will depend on the mean con-

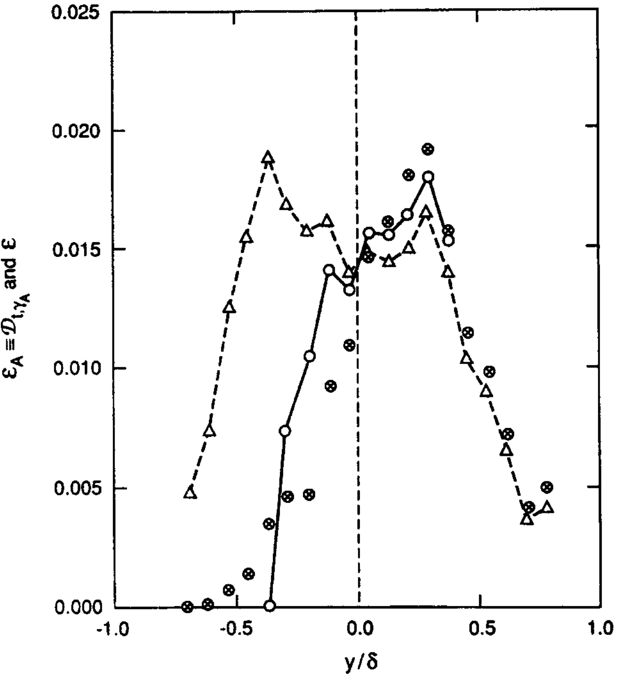

Figure 4. Turbulent diffusivities of species *A* at $N_D = 1.81$; ⊗, from Figure 1b; ○, from Eq. 12; △, nonreacting diffusivity from Figure 1a.

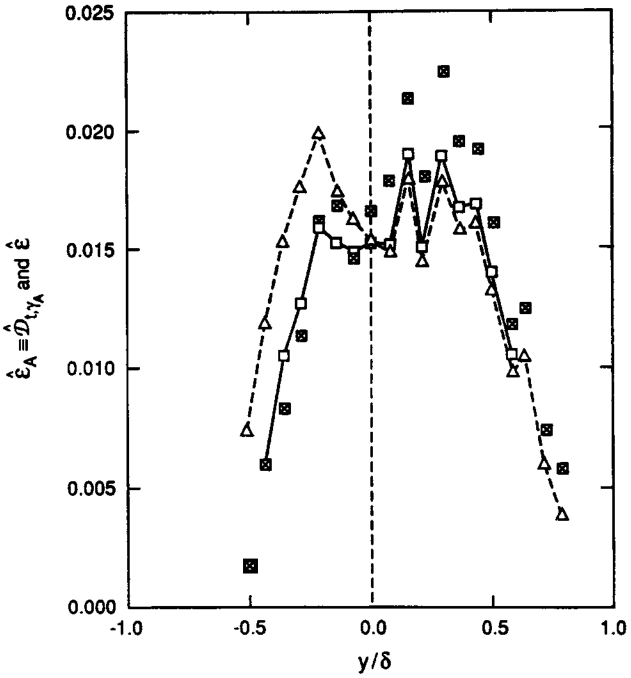

Figure 6. Turbulent diffusivities of species *A* at $N_D = 0.3$; ⊠, from Figure 1b; □, from Eq. 12; △, nonreacting diffusivity from Figure 1a.

centration gradients, it is explicit relationships for these individual reactant diffusivities which are needed for closure of Eq. 1. Although Borghi and Dutoya (1978) attempted to do this for combustion using a turbulent flux balance, we will show that an elementary mixing length model (Taylor, 1915; Hinze, 1959; Tennekes and Lumley, 1983) may be sufficient for some engineering purposes.

A Qualitative Explanation

We first show that the qualitative features of Bilger et al.—

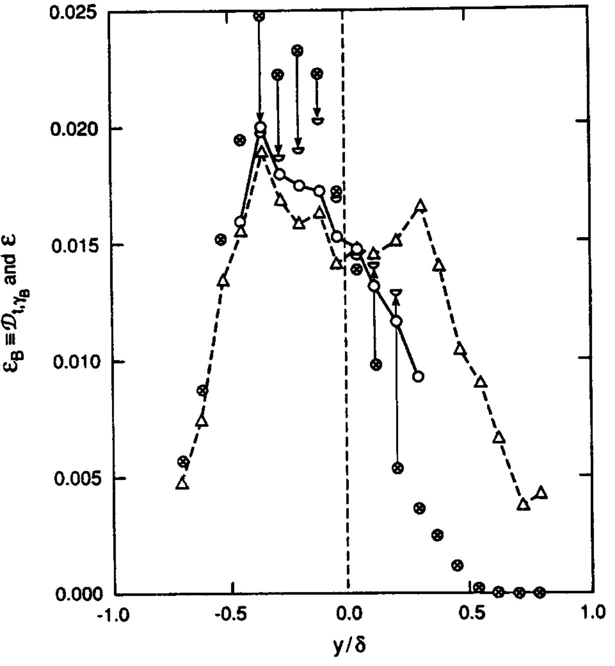

Figure 5. Turbulent diffusivities of species *B* at $N_D = 1.81$; ⊗, from Figure 1c; ○, from Eq. 17; △, nonreacting diffusivity from Figure 1a; ▽ $\hat{\epsilon}_B$ from Eq. 2.

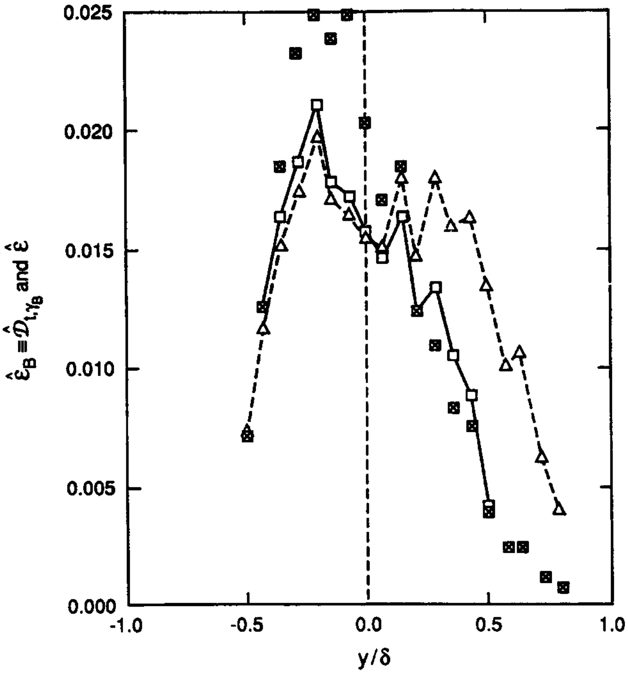

Figure 7. Turbulent diffusivities of species *B* at $N_D = 0.3$; ⊠, from Figure 1c; □, from Eq. 17; △, nonreacting diffusivity from Figure 1a.

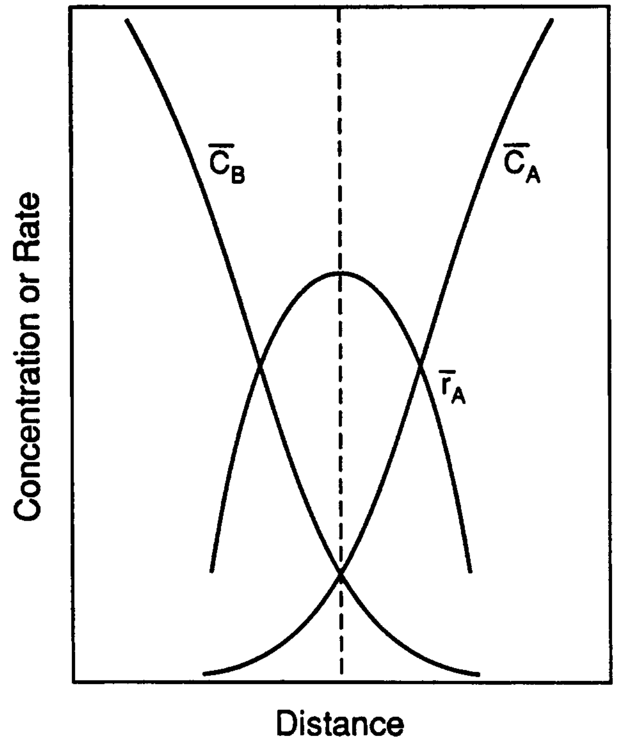

Figure 8. Mean reaction rate and species concentrations vs. position during reactant mixing.

that in their particular experiment reactant turbulent diffusivities are increased on the side from which they come and are decreased on the other side—are predicted by a mixing length model which also brings out the underlying cause. (The Damköhler Number effect is more easily seen in the next section.)

Figure 8 is a sketch of the data by Bilger et al. which shows the mean concentrations and reaction rate as a function of position transverse to the main flow. The time-average velocity in the transverse direction is zero, so transport in this direction is caused by velocity fluctuations which exchange lumps of fluid between regions of different concentrations (large-scale molecular transport is negligible). Thus, in the absence of reaction, this model leads to a positive turbulent diffusivity which depends on the intensity of the velocity fluctuations and the distance (mixing length) traveled by the lumps before they mix with the surrounding fluid, that is, before they are *micromixed.* During this motion, since the lump is not in equilibrium with its environment, it can exchange a passive tracer with the surrounding fluid by molecular diffusion leading to an effect of molecular diffusivity on turbulent diffusivity. Any *micromixing* inside a nonuniform lump, since it conserves material, could affect only the macrotransport indirectly by affecting the interchange between the lump and its surroundings, and even this "leak" will normally be small (Tennekes and Lumley, 1983). (It is this small interchange that leads to the idea of transferable properties and mixing length models.) A *reaction* which takes place *inside* a lump, a microscale phenomenon, however, does *not* conserve material even in the absence of a leak, and, as will be seen, is the apparent cause of the observed effects.

Consider the situation in Figure 8, focus on reactant A on the right side of the centerline, and consider two imaginary parallel planes perpendicular to the y direction and separated by a small distance. Lumps interchange between these planes, but now a reaction takes place *inside* the lumps—in this case removing reactant A. Because the integral-scale lumps are much larger than the Batchelor scale, the microscale at which the reaction takes place, lumps will sample the mean reaction rate as well as mean concentration of their birthplace. Consequently, lumps leaving the right plane will leave on the average with the time average concentration and time average reaction rate at that plane, so less A will reach the lefthand plane in that lump than would occur in the absence of reaction. Similarly, lumps leaving the left plane on the average leave with the time-average concentration and reaction rate in that plane, and this lump will carry less A to the right than it would if there *were no* reaction. Note, however, that if the reaction rate in the left and right leaving lumps were the same during their flight, the net transport and hence turbulent diffusivity would *not* be affected by the reaction. The transport and turbulent diffusivity are affected, because the reaction rates differ. They differ because of the small leakage from the lumps during their flight so the initially different reaction rates *persist.* (If conversion in a lump is small, the rate will be essentially conserved, but even if it is not small the two average rates will differ so long as the lumps do not immediately equilibrate with their surroundings. The reaction rate is in this sense at least a *quasitransferable* property.)

The right leaving lumps leave with a lower reaction rate (see Figure 8), and this lower rate persists so less A is lost from these lumps than is lost from the left leaving lumps which start with and continue with a higher reaction rate as they move right. Hence, as observed experimentally (Figure 1), the net transport from right to left is increased—the turbulent diffusivity of A on the right—the side from which A comes is *increased* by the reaction as observed, but it is increased because of the persistence of the reaction rate bias—it is not an eddy leak—it is a true microscale effect *within* the lumps.

On the left side of the centerline, the gradient of the mean concentration of A and its rate are both in the *same* direction. Again, consider two parallel planes on the left of the centerline. Now left leaving lumps react more slowly than right leaving lumps, so more A is lost by the right leaving lumps than the left leaving ones. Hence, less A is transported down the mean gradient—the turbulent diffusivity is *decreased* by the reaction—again as observed. Reactant B merely mirrors the behavior of A—same arguments and conclusions. This simple mixing length viewpoint is qualitatively in accord with experiment and affords a simple explanation for the coupling as well as the following quantitative result.

Quantitative Development

We will use unidirectional definitions of the turbulent fluxes:

$$\overline{\phi} = \overline{v'C'} \equiv -\epsilon \frac{d\overline{C}}{dy} \tag{3}$$

$$\overline{\phi}_A = \overline{v'C'_A} \equiv -\epsilon_A \frac{d\overline{C}_A}{dy} \tag{4}$$

If 2λ is the mixing length, lumps cross a plane carrying their transferable properties from within distances of 2λ on either side of the plane, so the *average* distance of travel to the plane is λ if statistical properties do not change over the distance 2λ. Hence, to relate ϵ and ϵ_A we consider the two imaginary planes discussed earlier (now drawn in Figure 9) to be the distance 2λ apart and determine the net flux from left to right crossing a parallel plane midway between the two planes. All the lumps crossing the midplane are thus taken to have traveled the average distance λ.

A lump moving with the velocity u from left to right carries $u\bar{C}_{A1}$ moles A/time area to the right and the amount carried across the midplane is given by:

$$u\left(\bar{C}_{A1}-\frac{\lambda}{\tilde{u}}r_{A1}\right) \tag{5}$$

It is assumed that there is no leak from the lump and that r_{A1} is the average reaction rate in these left leaving lumps based on the average flight time $\lambda/\tilde{u}$.

The amount of A crossing the midplane in right leaving lumps is then:

$$u\left(\bar{C}_{A2}-\frac{\lambda}{\tilde{u}}r_{A2}\right)$$

so, for two equal and opposite moving lumps the net flux across the midplane is:

$$\phi_A=u\left[(\bar{C}_{A1}-\bar{C}_{A2})-\frac{\lambda}{\tilde{u}}(r_{A1}-r_{A2})\right] \tag{6}$$

As usual, for small λ (Tennekes and Lumley, 1983, for other justifications):

$$\bar{C}_{A1}-\bar{C}_{A2}\approx-2\lambda\frac{d\bar{C}_A}{dy} \tag{7}$$

There are two opposing tendencies within a lump—conversion *decreases* the reaction rate while micromixing *increases* rate. The problem of evaluating the net rate change is avoided by writing:

$$r_{A1}-r_{A2}\approx-2\lambda\frac{d\bar{r}_A}{dy} \tag{8}$$

This equation, which treats $r_{A1}-r_{A2}$ like $C_{A1}-C_{A2}$, assumes that the change in rate during the motion of a lump is the same in both left and right leaving lumps and hence has no effect on the *difference* between the two rates. Combining Eqs. 6, 7, and 8 and averaging:

$$\bar{\phi}_A=-2\overline{u\lambda}\left[\frac{d\bar{C}_A}{dy}-\frac{\lambda}{\tilde{u}}\frac{d\bar{r}_A}{dy}\right] \tag{9}$$

When $\bar{r}_A$ is set equal to zero, Eqs. 3 and 9 identify $2\overline{u\lambda}$ as ϵ so:

$$\bar{\phi}_A=-\epsilon\left[\frac{d\bar{C}_A}{dy}-\tau_M\frac{d\bar{r}_A}{dy}\right]\equiv-\epsilon_A\frac{d\bar{C}_A}{dy} \tag{10}$$

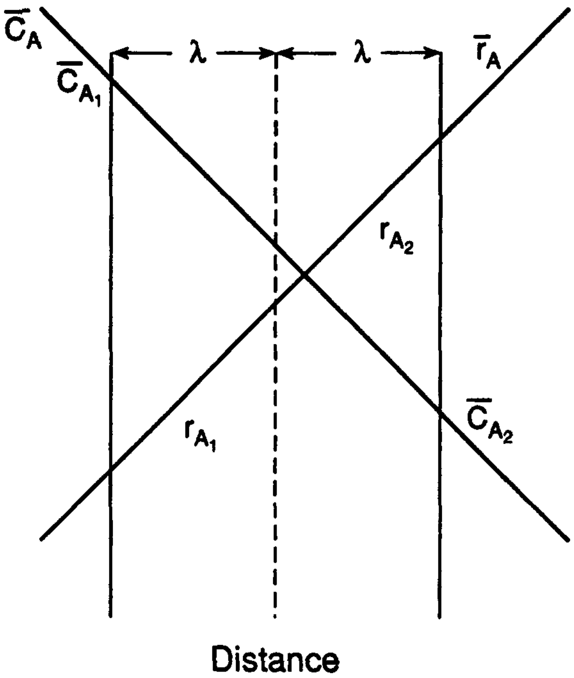

Figure 9. Mixing length model.

The flux is enhanced or diminished, depending on the relative signs of the gradients of $\bar{C}_A$ and $\bar{r}_A$.

Rearranging Eq. 10 gives the relationship between the two turbulent diffusivities which closes the macromixing term in Eq. 1:

$$\frac{\epsilon_A}{\epsilon}=1-\tau_M\frac{d\bar{r}_A}{d\bar{C}_A} \tag{11}$$

Although Borghi and Dutoya's (1978) results appear to be in qualitative agreement with Eq. 11, they are not directly comparable since they do not include the reaction rate explicitly.

The nondimensional form of Eq. 11 is:

$$\frac{\epsilon_A}{\epsilon}=1-N_{DM}\frac{d\bar{R}_A}{d\bar{X}_A} \tag{12}$$

where the Damköhler number which arises is the ratio of the flight time or characteristic *macro*mixing time, $\tau_M=\lambda/\tilde{u}$, to the characteristic reaction time, τ_R:

$$N_{DM}=\frac{\tau_M}{\tau_R} \tag{13}$$

Equation 12 is presumably valid for any chemistry, but if the reaction is described by the mass action law:

$$\bar{r}_A=k\overline{C_AC_B} \tag{14}$$

then, $\tau_R=1/kC_{BO}, \bar{R}_A=\overline{X_AX_B}$,

$$N_{DM}=\frac{\lambda}{\tilde{u}}kC_{BO} \tag{15}$$

and

$$\frac{\epsilon_A}{\epsilon}=1-N_{DM}\frac{d\overline{X_AX_B}}{d\overline{X}_A} \tag{16}$$

The equation for B is:

$$\frac{\epsilon_B}{\epsilon}=1-\frac{N_{DM}}{\beta}\frac{d\overline{X_AX_B}}{d\overline{X}_B} \tag{17}$$

β appears in this equation because of the choice of N_{DM} made in Eq. 15.

This form is not particularly useful for an instantaneous reaction, but closure is not needed here, since the solution to Eq. 1 at this limit is known in terms of the solution to the analogous nonreacting problem (Toor, 1962). This is known as the "conserved scalar" method in the combustion literature (Bilger et al., 1991).

Equation 11 can also be derived by determining the effect of the reaction on the reactant concentration fluctuations. A reaction which consumes a reactant decreases positive fluctuations and enhances negative fluctuations. Equation 11 also follows from this viewpoint because of the persistence of the bias in the reaction rates.

Proceeding any further with Eqs. 16 and 17 requires a micromixing model since

$$\overline{X_AX_B}=\overline{X}_A\overline{X}_B+\overline{X_A'X_B'} \tag{18}$$

This, however, is not needed with first-order reactions which are unaffected by micromixing, so Eq. 8 will *always* overestimate the amount of reaction in this case. With

$$\overline{r}_A=k\overline{C}_A \tag{19}$$

Eq. 11 becomes:

$$\frac{\epsilon_A}{\epsilon}=1-k\tau_M \tag{20}$$

The diffusivity is always decreased by the reaction because the reaction rate is always higher at the high reactant concentration.

This first-order reaction is not very interesting in its own right, but it does allow examination of the approximation introduced by Eq. 8, since the reaction rate in a lump is now simply given by Eq. 19. The result obtained by summing the reaction effects in all lumps originating within a distance 2λ on either side of a given plane is:

$$\frac{\epsilon_A}{\epsilon}=\frac{1-(1+2k\tau_M)e^{-2k\tau_M}}{2(k\tau_M)^2} \tag{21}$$

Equation 20 approximates Eq. 21 only for small values of the macro Damköhler number, $k\tau_M$. Unfortunately, this does not help much in determining the error in Eq. 11 for other kinds of reactions which *are* sensitive to micromixing. (Borghi and Dutoya, 1978, do obtain an explicit result for the first-order reaction and if their time constant is taken as τ_M, they predict a smaller effect than given by either Eq. 20 or 21.)

Comparison with Experiment

Returning to Eq. 11 (or 12), we first note that with its counterpart for reactant B it satisfies the consistency condition of Eq. 2, because $\overline{r}_B=n\overline{r}_A$ for the single-step reaction. Secondly, we observe that Eq. 12 has the correct overall behavior (the behavior observed by Bilger et al., 1991) for their experiment: (1) the reactant turbulent diffusivities are higher than the ordinary (passive) values on the side from which the reactant comes (because of the sign of $d\overline{R}_A/d\overline{X}_A$); (2) they are *much* lower than the ordinary values on the other side (because the rate of change of reaction with concentration is *much* larger on that side); and (3) in this latter region there is a Damköhler effect with diffusivity values for the high N_D smaller than those for the low N_D (obvious from Eq. 12).

Because τ_M is not precisely fixed by the model it would be reasonable to take τ_M as an adjustable parameter. However, it is interesting to evaluate τ_M with measured quantities. The mixing length 2λ should be approximately the integral length scale (Tennekes and Lumley, 1984), $\tilde{u}$ the RMS velocity fluctuation, and since Bilger et al. (1991) have measured these values we can estimate a single value of τ_M from their measurements. Using the average measured integral scale of 0.25 m and $\tilde{u}$ of 0.025 m/s (Saetran, 1989) gives an average micromixing time, τ_M, of 5.0 s.

The macromixing Damköhler number in Eq. 12 is related to that used by Bilger by:

$$N_{DM}=\frac{\lambda}{M}\frac{\overline{U}}{\tilde{u}}\frac{\beta}{1+\beta}N_D$$

The mean velocity U is 0.55 m/s and M is 0.32 m. $\beta=0.94$ in the experiment with $N_D=1.81$ and 1.03 in the experiment with $N_D=0.3$. Then, for $N_D=1.8$, $N_{DM}=7.61$ and for $N_D=0.3$, $N_{DM}=1.30$.

We read $\overline{R}_A$, $\overline{X}_A$, and $\overline{X}_B$ off Figures 2 and 3, determined $(d\overline{R}_A/d\overline{X}_A)$ and $(d\overline{R}_B/d\overline{X}_B)$ by finite differences and then interpolated to obtain a value of $d\overline{R}_A/d\overline{X}_A$ for each reported value of $\hat{\epsilon}$. Equation 12 was then used to calculate $\hat{\epsilon}_A$ and Eq. 17 to calculate $\hat{\epsilon}_B$. Figures 4 through 7 compare these values with experiment. They all show $\hat{\epsilon}$ and both experimental and predicted values of $\hat{\epsilon}_A$ or $\hat{\epsilon}_B$ and $\hat{\epsilon}$.

Figures 4 and 5 are at the higher N_D of 1.81. In Figure 4 the experimental values of $\hat{\epsilon}_A$ left of the centerline are much lower than $\hat{\epsilon}$. We do not have an estimate of the uncertainty in these values, but the predictions of Eq. 12 capture this behavior surprisingly well, as it also does on the other side, the side from which A comes. There, Eq. 12 predicts a small increase in $\hat{\epsilon}_A$ for all points—close to experiment for most of the data (some of the reaction rate data here is questionable so we have used the rate data from the left side of Figure 3). $\hat{\epsilon}_B$, shown in Figure 5 is predicted almost as well by Eq. 17, except for the cluster of very high points near left center. It is possible that these $\hat{\epsilon}_B$ data are incorrect, since the reported $\hat{\epsilon}_A$ and $\hat{\epsilon}_B$ data do not satisfy Eq. 2 which should hold in this system.

We have recalculated $\hat{\epsilon}_B$ from Eq. 2 by assuming that the reported $\hat{\epsilon}_A$ are correct and these corrected values, shown in Figure 5, are much closer to the predictions of Eq. 17, which may lend some credence to the suggestion that the data are in error here.

Figures 6 and 7 repeat Figures 4 and 5 at a lower N_D. The decrease in diffusivities, on the left in Figure 6 and right in Figure 7, less than at the higher Damköhler number in Figures 4 and 5, is very nicely predicted by Eqs. 12 and 16. The small increase on the other side is also handled well, although there are more anomalous high data points in both figures. The reported values of $\hat{\epsilon}_A$ and $\hat{\epsilon}_B$ are inconsistent with Eq. 2 in such a random manner that there seems little point in recalculating $\hat{\epsilon}_B$ (or $\hat{\epsilon}_A$) as was done before.

In all the calculations, we assumed τ_M to be constant, the mixing length 2λ to be the measured integral length scale, $\tilde{u}$ to be the measured average RMS velocity fluctuation and this obtained reasonably satisfactory agreement with experiment—unexpectedly good in many cases, even though the model cannot explain the very high values of $\hat{\epsilon}_A$ and $\hat{\epsilon}_B$ in Figures 5-7. Is this a failure of the model or are these data erroneous? Further data are needed to answer the question.

Convective Diffusion Equation

Generalizing Eqs. 3, 4, and 12 to three dimensions gives:

$$\underline{\overline{\phi}}_i = -[(D_i\underline{\underline{\delta}} + \underline{\underline{\epsilon}}) \cdot \nabla \overline{C}_i - \underline{\underline{\epsilon}} \cdot (\underline{\underline{\tau}}_M \cdot \nabla \overline{r}_i)], \quad i = A, B, \ldots \tag{22}$$

The new term contains the macroproperties ϵ and τ_M, and the reaction term which depends on micromixing. This is a true macro-micro coupling which causes an apparent flux, $\underline{\underline{\epsilon}} \cdot (\underline{\underline{\tau}}_M \cdot \nabla \overline{r}_l)$, which can either enhance or hinder the normal turbulent flux, $-\underline{\underline{\epsilon}} \cdot \nabla \overline{C}_i$.

Even though the apparent flux can be a significant fraction of the normal flux, it is unlikely to have a significant effect on the mean concentration field because

$$\left| \frac{\epsilon \tau_M \dfrac{d^2 \overline{r}_A}{dy}}{\overline{r}_A} \right| \sim O\left(\frac{\lambda}{\delta}\right)^2 \tag{23}$$

Thus, since Eq. 8 requires that λ/δ be small (λ/δ is about 0.08 in the experiments of Bilger et al., 1991), in Eq. 1a the contribution of the apparent flux term will be smaller than the reaction term.

Equation 18 still requires evaluation of the concentration covariance term or terms that appear in the mean reaction rate term. There is much less information available in multidimensional than in unidimensional systems as to how to handle these micromixing effects. Bilger et al. (1991) found that the closure, which assumes that the covariance term, $\overline{C_A'C_B'}$, which arises with the single-step bimolecular reaction is the value which would exist with an instantaneous reaction (Toor, 1969), predicted their data moderately well. Assuming perfect micromixing ($\overline{C_A'C_B'} = 0$) did less well, but this avoids the micromixing problem altogether and closes Eq. 18 for all chemistries. Because macrotransport tends to control in multidimensional systems (Hanks, 1991), this naive closure may well give acceptable engineering results, but this speculation requires further study.

Acknowledgment

Equation 23 is due to Da-Ming Wang and J. M. Tarbell. Their help is greatly appreciated, as were discussions with Dennis C. Prieve.

Notation

A, B = reactants A and B
C = concentration in nonreacting systems, mol/m^3
$\overline{C}_i$ = concentration of reactant i, mol/m^3
$\overline{C}_{A1}, \overline{C}_{A2}$ = average concentration of A in lumps leaving positions 1 and 2, respectively, mol/m^3
D_i = molecular diffusivity of i, m^2/s
k = reaction velocity constant, m^3/mol·s for a second-order reaction, s for a first-order reaction
M = pitch of grid in experiment
n = stoichiometric coefficient
N_D = Damköhler no., $(M/U)k(C_{AO}+C_{BO})$
N_{DM} = macro Damköhler no., τ_M/τ_R; for a second-order bimolecular reaction $= (\lambda/\tilde{u})kC_{BO}$
r_i = reaction rate mol/m^3·s
r_{A1}, r_{A2} = average reaction rate during flight in left and right leaving lumps, respectively, m^3/mol·s
R_i = nondimensional reaction rate, $r_i\tau_R/C_{iO}$; for a second-order bimolecular reaction $= r_i/kC_{AO}C_{BO}$
t = time, s
$\underline{u}$ = lump velocity, m/s
$\overline{U}$ = mean experimental flow velocity, m/s
$\tilde{u}$ = average lump speed, m/s
v = fluid velocity, m/s
v' = velocity fluctuation
X_i = nondimensional concentration of i, C_i/C_{iO}
y = distance normal to flow toward the A side, m

Greek letters

β = stoichiometric ratio of feed, C_{BO}/nC_{AO}
δ = width of mixing layer, m
ϵ = passive turbulent diffusivity—no reaction, m^2/s
ϵ_A, ϵ_B = reactant turbulent diffusivities, m^2/s
ϕ = diffusive flux in absence of reaction, m^2/s
ϕ_i = diffusive flux with reaction, m^2/s
λ = half mixing length, m
τ_M = characteristic macromixing time, $\lambda/\tilde{u}$, s
τ_R = characteristic reaction time, $1/kC_{BO}$ for a bimolecular second-order reaction, $1/k$ for a first-order reaction, s

Subscripts

i = reactant A or B
O = initial value
$\underline{\ }$ = vector
$\underline{\underline{\ }}$ = tensor

Superscripts

$\overline{\ }$ = time mean value
$\hat{\ }$ = nondimensionalized by $1/\overline{U}M$
$'$ = fluctuation

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