Turbulent Diffusivities in Reacting Systems

H. L. Toor
Dept. of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213

When two reactants with equal molecular diffusivities are mixed, the convective diffusion equations and stoichiometry for the reaction $A + nB \rightarrow$ products yield a linear relationship among the reactant concentrations and a nonreacting tracer (Hawthorne et al., 1949; Toor, 1962; Lin and O'Brien, 1974; Li and Toor, 1986; Bilger et al., 1991):

$$x_A - \beta x_B = (1 + \mathrm{B})f - \beta \qquad (1)$$

Then, in a turbulent system where x_A, x_B, and f fluctuate:

$$\overline{x_A} - \beta\overline{x_B} = (1 + \beta)\bar{f} - \beta \qquad (2)$$

$$x_A{}' - \beta x_B{}' = (1 + \beta)f' \qquad (3)$$

$$\overline{\underline{v}'x_A{}'} - \beta\overline{\underline{v}'x_B{}'} = (1 + \beta)\overline{\underline{v}'f'} \qquad (4)$$

Define conventional scalar turbulent diffusivities:

$$\overline{\underline{v}'f'} \equiv -\epsilon\nabla\bar{f} \qquad (5)$$

$$\overline{\underline{v}'x_A{}'} \equiv -\epsilon_A\nabla\bar{x}_A \qquad (6)$$

$$\overline{\underline{v}'x_B{}'} \equiv -\epsilon_B\nabla\bar{x}_B \qquad (7)$$

From Eq. 2:

$$\nabla\bar{x}_A - \beta\nabla\bar{x}_B = (1 + \beta)\nabla\bar{f} \qquad (8)$$

Then, Eqs. 4 to 8 give a relationship among the three turbulent diffusivities:

$$(\epsilon_A - \epsilon)\nabla\bar{x}_A = \beta(\epsilon_B - \epsilon)\nabla\bar{x}_B \qquad (9)$$

Equation 9 can also be derived by starting with the time averaged convective diffusion equations.

Equation 9 allows equality among the turbulent diffusivities, $\epsilon_A = \epsilon_B = \epsilon$, which would be expected to be the case for slow reactions, but apparently not for faster reactions since measurements of Bilger et al. (1991) in a gas behind a splitter plate with Damköhler Numbers of 0.3 and 1.8 show significant differences among the three turbulent diffusivities. Their results appear to be consistent with Eq. 9, which predicts that the absolute deviation in reactant diffusivity from the normal tracer diffusivity will be larger for the reactant with the smaller mean gradient when the reactants are not premixed so that the mean gradients and the deviations are of opposite signs.

Although it is possible to obtain the mean reactant and product concentrations from Eq. 1 when the reaction is *instantaneous* (Toor, 1962; Lin and O'Brien, 1974; Bilger et al., 1991), surprisingly enough, this does not seem to fix the turbulent diffusivities of the reactants. This is a situation in which we know the solution to the time-averaged, convective diffusion equations, but *not* the explicit terms in the equations themselves!

Even though turbulent reactant diffusivities are not needed with instantaneous reactions, they would normally have to be used to close the time-averaged, convective diffusion equations—the equations that must be solved to compute mean concentrations with finite speed reactions. The fact that these diffusivities may be reaction-dependent is troublesome; at best they may be useful, even though lacking the conventional meaning as a fluid mechanical quantity, and at worst new closure methods may be required.

Acknowledgment

This work was supported by National Science Foundation under Grant CBT-8703890. Helpful comments were received from R. W. Bilger.

Notation

C_A = concentration of reactant A
C_B = concentration of reactant B
f = nondimensional tracer concentration, C_A^o/C_{Ao}^o
n = stoichiometric coefficient
$\underline{v}$ = velocity vector
x_A = nondimensional concentration of reactant A, C_A/C_{Ao}
x_B = nondimensional concentration of reactant B, C_B/C_{Bo}

Greek letters

β = equivalence ratio, $C_{Bo}/n\ C_{Ao}$
ϵ = turbulent diffusivity of nonreactive tracer
ϵ_A = turbulent diffusivity of reactant A
ϵ_B = turbulent diffusivity of reactant B

Subscripts and superscripts

o = concentration of nonreacting tracer

$\bar{\ }$ = mean
$'$ = fluctuation
o = initial value

Literature Cited

Bilger, R. W., L. R. Saetran, and L. V. Krishnamoorthy, "Reaction in a Scalar Mixing Layer," *J. Fluid Mech.*, in press (1991).

Hawthorne, W. R., D. S. Weddel, and H. C. Hottel, *Symp. Combustion and Flame and Explosion Phenomena*, p. 266, Williams and Wilkins, Baltimore (1949).

Li, K. T., and H. L. Toor, "Chemical Indicators as Mixing Probes: a Possible Way to Measure Micromixing Simply," *I&EC Fund.*, **25**, 719 (1986).

Lin, C.-H., and E. E. O'Brien, "Turbulent Shear Flow Mixing and Rapid Chemical Reactions: an Analogy," *J. Fluid Mech.*, **54**, 195 (1974).

Toor, H. L., "Mass Transfer in Dilute Turbulent and Nonturbulent Systems with Rapid Irreversible Reactions and Equal Diffusivities," *AIChE J.*, **8**, 70 (1962).

Manuscript received June 13, 1991, and revision received Aug. 19, 1991.

Errata

- In the article titled "A Series Solution for Mass Transfer in Laminar Flow with Surface Reaction" (September 1991, p. 1341), three lines above Eq. 9 on p. 1342 "a denotes the scaled Damköhler number, d_1Da/d_2, where . . . " should read "a denotes the scaled Damköhler number, d_1D/d_2, where . . . " Equation 33 on p. 1345 should read:

$$F_m(\xi) = e^{-\xi}[1 + (-\xi)^{1/3-m}\gamma^*(1/3 - m,\ -\xi) + (-\xi)^{2/3-m}\gamma^*(2/3 - m,\ -\xi)]$$

In addition, case a) of Appendix A on p. 1352 should start with $\Sigma t_m(-p)^m$, not $\Sigma \tau_m(-p)^m$. And Eq. 19 on p. 1344 should read:

$$\bar{N}(p) = \frac{1}{d_1} \sum_{m=1}^{\infty} t_m(-\eta) \sum_{i=1}^{m} (-p)^{m-i} + \sum_{m=1}^{\infty} \alpha_m \eta^{3m} p^{m-1}$$

- In the article titled "A Transformation Approach to Nonlinear Process Control" (July 1991, p. 1082), the following reference was inadvertantly omitted from the section on Literature Cited:

Alsop, A. W., and T. F. Edgar, "Nonlinear Heat Exchanger Control Through the Use of Partially Linearized Control Variables," *Chem. Eng. Comm.*, **75**, 155 (1989).