LETTERS TO THE EDITOR

To the Editor:

In their recent paper (32(8), p. 1312, 1986), Li and Toor discuss in detail simultaneous mixing and reaction in a turbulent, tubular reactor. Conclusions from their experiments are:

a) The effective slab thickness δ of a diffusion-reaction model is inversely proportional to the square root of the reactant diffusivity D, when the Reynolds number ud/ν and the tube diameter d are held constant, while the reaction is changed. This finding, obtained from very rapid reactions in gas and liquid phases, was generalized as:

$$\delta \sim Sc^{-1/2} \tag{1}$$

and was also valid for a thinning slab model. They concluded, "the slab thickness δ, originally assumed to be fixed only by hydrodynamics, also depends on diffusivity."

b) From Eq. 1, they deduced, "there is no predicted effect of diffusivity on mixing or reaction" and "reactive mixing is not controlled by molecular diffusion."

Both conclusions are paradoxical, and (b) especially conflicts with most previous thinking. This letter interprets them in terms of a comprehensive micromixing model (Baldyga and Bourne, 1984).

Slab thickness and diffusivity

According to our model, the rate of micromixing (i.e., rate of dissipation of concentration variance by molecular diffusion) is highest near the Batchelor microscale λ_B, where:

$$\lambda_B = \lambda_K Sc^{-1/2} \tag{2}$$

$$\lambda_K = (\nu^3/\epsilon)^{1/4} \tag{3}$$

Using the Blasius equation for pressure drop, the rate of energy dissipation ϵ can be shown to be:

$$\epsilon \sim u^{11/4}\nu^{1/4}d^{-5/4} \tag{4}$$

Substituting Eq. 4 into the definition of the Kolmogorov scale, Eq. 3 and combining with Eq. 2 gives:

$$\lambda_B \sim d\, Re^{-11/16}Sc^{-1/2} \tag{5}$$

With constant values of d and Re, Eq. 5 simplifies to Eq. 1, which is the experimental result of Li and Toor. δ should be interpreted as a scale for micromixing, not the thickness of a quiescent region.

Diffusion and the rate-determining step

The experimental result of Li and Toor is that the concentration-time profiles for rapid second-order reactions using separate feed streams are not influenced by diffusivity, when Re, d and the stoichiometric ratio β are held constant. If neither diffusion nor chemical kinetics were rate-determining, what is? Our model incorporates molecular diffusion within stretching, energy-dissipating vortices, whereby several vortex generations are needed to complete micromixing. The average life time of such a vortex τ_w is given by:

$$\tau_w = 12\,(\nu/\epsilon)^{1/2} \tag{6}$$

The half-life time for diffusion in the absence of reaction, t_{DS}, is:

$$t_{DS} = 2\,(\nu/\epsilon)^{1/2}\,\text{arc sinh}\,(0.05\,Sc) \tag{7}$$

The condition for diffusion to be completed within each vortex generation to make the rate of generation of vortices rate-determining is:

$$t_{DS} \ll \tau_w \quad \text{or} \quad Sc \ll 4{,}000 \tag{8}$$

The Schmidt numbers in all Toor's experiments satisfied Eq. 8, and this explains why he deduced that D has no effect.

For sufficiently high Sc, Eq. 8 is no longer satisfied, and reactive mixing in, for instance, high viscosity media can become diffusion-controlled. Care is also needed with multiple reactions. Their time scales may be independent of D, but not the product-determining spatial concentration distributions.

Literature cited

Baldyga, J., and J. R. Bourne, "A Fluid Mechanical Approach to Turbulent Mixing and Chemical Reaction," *Chem. Eng. Comm.*, **28**, pp. 243 and 259 (1984).

Li, K. T., and H. L. Toor, "Turbulent Reactive Mixing with a Series-Parallel Reaction—Effect of Mixing on Yield," *AIChE J.*, **32**, 1312 (1986).

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To the Editor:

We would like to note some interesting connections between the results of Mijares, Cole, Naugle, Preisig and Holland (32(9), p. 1439, 1986) concerning criteria for selecting pairings for decentralized control and the work of Grosdidier and Morari (1985, 1986).

Let G be the plant transfer function and let $\tilde{G}$ be a simplified model of this plant containing the diagonal elements of G only.

$$\tilde{G} = \text{diag}\,\{g_{11}, g_{22}, \ldots, g_{nn}\}$$

G and $\tilde{G}$ are assumed stable. A diagonal controller $C = \text{diag}\,\{c_i\}$ with integral action is to be used to control the plant. Let $\tilde{H} = \text{diag}\,\{\tilde{h}_i\} = \tilde{G}C(I + \tilde{G}C)^{-1}$ denote the closed-loop transfer matrix of the diagonal system

$$\tilde{h}_i = g_{ii}c_i/(1 + g_{ii}c_i)$$

Based on stability arguments for inverting a matrix, Mijares et al. propose to use the "Jacobi Eigenvalue Criterion" for selecting the best pairing of controlled and manipulated variables: The pairing which minimizes $\rho(E)$ is selected, where $E = (G - \tilde{G})\tilde{G}^{-1} = G\tilde{G}^{-1} - I$, and E is evaluated at steady state ($\omega = 0$). $\rho(E)$ is the spectral radius that is defined as the magnitude of the largest eigenvalue of E. [Mijares et al. consider the matrix $A = I - \tilde{G}^{-1}G$, but this does not change the condition since $\lambda_i(E) = -\lambda_i(A)$ (λ_i denotes the eigenvalue)].

This criterion may also be derived from Corollary 2.1 in Grosdidier and Morari

(1986) that states:

"Assume that all individual loops are stable (i.e., $\tilde{h}_i$ stable) and have been chosen to have identical transfer functions, i.e., $\tilde{H} = \tilde{h}I$ [for example, $c_i(s) = k(s)/g_{ii}(s)$]. Then the overall system with all loops closed is stable if

$$|\tilde{h}(j\omega)| < \rho^{-1}(E(j\omega)) \quad \forall\omega\text{"} \quad (1)$$

In particular, this condition shows that decentralized control with integral action ($\tilde{h}(0) = 1$) is always possible if $\rho(E(0)) < 1$, and a reasonable criterion for selecting pairings is to choose the one with the smallest $\rho(E(0))$. However, if the process dynamics were known, this information should also be used and $\rho(E(j\omega))$ should be kept small as seen from Eq. 1. Thus, Condition 1 also extends the "Jacobi Eigenvalue Criterion" to nonzero frequencies. Condition 1 is derived by Grosdidier and Morari (1986) using the Nyquist criterion which leads to the stability condition $\rho(\tilde{H}E) < 1$. The approach taken by Mijares et al. is less general, but may be helpful, for example, for persons with a background in process design rather than in control.

Condition 1 is only sufficient, and a decentralized controller with integral action may be possible even if $\rho(E(0)) > 1$. To illustrate this, consider the controller $C(s) = k/s\hat{C}(s)$ where $\hat{C}(s)$ is diagonal and satisfies $\hat{C}(0) = \tilde{G}^{-1}(0)$. According to Theorem 7 in Grosdidier et al. (1985), there exists a k^* such that this particular controller results in a stable closed-loop system for any $k\epsilon(0,k^*]$ (integral controllability), if and only if $\mathrm{Re}\,\{\lambda_i(G\tilde{G}^{-1}(0))\} > 0, \forall i$. From the identity $\lambda(G\tilde{G}^{-1}) = \lambda(E + I) = \lambda(E) + 1$, we see that this is equivalent to requiring $Re\,\{\lambda_i(E(0))\} > -1, \forall i$. This interesting condition is given by Mijares et al. and is proved here to complement their derivation. Consequently, decentralized control with integral action is possible also with $\rho(E(0)) > 1$ when the real parts of the eigenvalues of $E(0)$ are all larger than -1.

One restriction of Eq. 1 is the assumption of identical loop responses. While this is always satisfied at steady state, where $\tilde{H}(0) = I$, this is not likely to be satisfied at nonzero frequencies. Starting from the stability condition $\rho(\tilde{H}E) \leq 1$, Grosdidier and Morari (1986) derive a generalized version of Eq. 1, which also applies when the responses $\tilde{h}_i$ for each loop are *not* identical;

$$|\tilde{h}_i(j\omega)| < \mu^{-1}(E(j\omega)) \quad \forall\omega, \quad \forall i \quad (2)$$

μ is the Structured Singular Value and is computed with respect to a diagonal structure. Note that $\mu(E) \geq \rho(E)$, and therefore Eq. 1 always gives the least restrictive bound on $|\tilde{h}_i|$. By replacing $|\tilde{h}_i|$ by $\bar{\sigma}(\tilde{H})$, condition 2 may easily be extended to cases where C is block-diagonal.

Literature cited

Grosdidier, P., M. Morari, and B. Holt, "Closed-Loop Properties from Steady-State Gain Information," *Ind. Eng. Chem. Fund.*, **24,** 221 (1985).

Grosdidier, P., and M. Morari, "Interaction Measures for Systems Under Decentralized Control," *Automatica,* **22**(3), 309 (1986).

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