An Evaluation of the Lamellar Stretch Description of Mixing with Diffusion and Chemical Reaction

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Introduction

One of the significant aspects of the lamellar stretch model of mixing with chemical reaction is the warped time transformation (Ranz, 1979), which provides a means for decoupling mechanical mixing from diffusional mixing. In essence, the convection term is eliminated from the concentration equation in the Lagrangian frame of reference. Hence, one is left with a partial differential equation in terms of the warped time and a single coordinate in space in the direction of molecular diffusion. The methodology has been used to investigate mixing with simultaneous reactions (Ou and Ranz, 1983a, b; Chella and Ottino, 1984). Further advances have been made (Ou et al., 1983, 1985) that permit the spatial distribution and time evolution of the striation thickness and intermaterial surface orientation to be determined from first principles. Thus, it appears that the lamellar stretch description of mixing has reached its maturity for practical applications. However, one should also keep in mind that the warped time transformation applied thus far allows for diffusion to take place only in one direction, i.e., across the striation thickness. In the description of a continuous-flow reactor involving unpremixed feeds, however, the material streak might not be thin enough to justify the presumption of diffusion taking place only across the striation. This recognition has prompted us to investigate the extent to which the diffusion in directions other than across the striation thickness affects the concentration distribution in space. Specifically, we have looked into the flow between two parallel plates, one of which is stationary and the other is moving at a constant speed. Our interest in such an idealized flow stems from the fact that simple shearing has been successfully employed to interpret experimental observations on chemical selectivities in a continuous-flow, stirred-tank reactor (Bourne and Rohani, 1983) and on the laminar mixing in the runner section of reaction injection molding (Bourne and Garcia-Rosas, 1985). A finite-difference scheme, the alternating direction implicit (ADI) method (McKee and Mitchell, 1970), was used to compute the concentration field of initially segregated species A and B with diffusion and with diffusion-controlled reaction. The computed results are compared to those obtained from the stretch model. The conditions then emerge for the applicability of the stretch model in terms of the values of two dimensionless groups.

Problem Description and Numerical Solution

Depicted in Figure 1a is the initial distribution of species A and B between a pair of parallel plates a distance H apart. The lower plate is stationary, and the upper plate moves at a constant speed v_O. Any point in a representative slab composed of equal amounts of A and B is specified by (ξ', η'), and the velocity field is given by

$$v_{\xi'} = \frac{v_O}{H}\eta' = G\eta' \tag{1}$$

At any time $\tau' > 0$ the orientation of species A and B is as depicted in Figure 1b. The concentration distribution can be described by the following equation:

$$\frac{\partial C_i'}{\partial \tau'} = D_i\left(\frac{\partial^2 C_i'}{\partial \xi'^2} + \frac{\partial^2 C_i'}{\partial \eta'^2}\right) - v_{\xi'}\frac{\partial C_i'}{\partial \xi'} \tag{2}$$

in which C_i' is the concentration of species A or B, τ' is the real time, and D_i is the molecular diffusivity of A or B. For the present work let us presume that $D_A = D_B = D$ and that $C_{AO}' = C_{BO}' = C_O'$.

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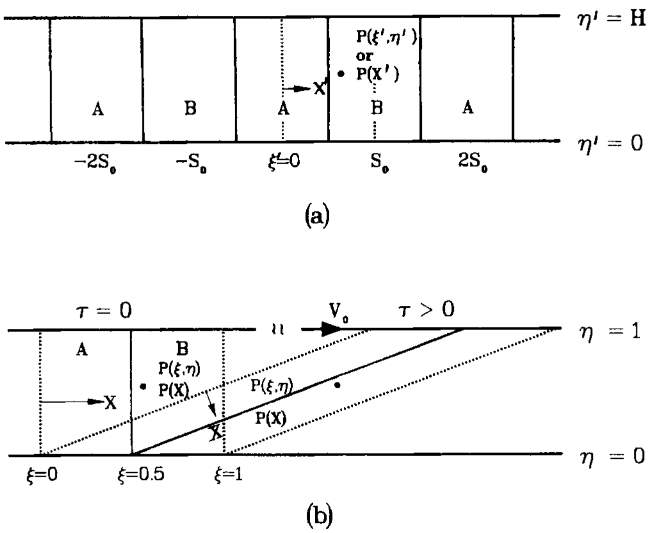

Figure 1. Material distribution in terms of dimensionless coordinates.

a. Initial distribution
b. Distribution at any time $\tau' > 0$

In terms of the following dimensionless variables:

$$\xi \equiv \frac{\xi' - \tau' v_{\xi'}(\eta')}{s_o} \tag{3}$$

$$\eta \equiv \frac{\eta'}{H} \tag{4}$$

$$\tau \equiv \frac{\tau' D}{s_o^2} \tag{5}$$

and

$$C \equiv \frac{C'}{C_o} \tag{6}$$

one could rewrite Eq. 2 as

$$\frac{\partial C}{\partial \tau} = (k^2\tau^2 d^4 + 1)\frac{\partial^2 C}{\partial \xi^2} - 2k\tau d^3 \frac{\partial^2 C}{\partial \xi \partial \eta} + \frac{\partial^2 C}{\partial \eta^2} d^2 \tag{7}$$

where

$$k \equiv \frac{H^2 G}{D} = Sc \cdot Re \tag{8}$$

and

$$d \equiv \frac{s_o}{H} \tag{9}$$

It is then obvious that k is characteristic of the importance of mechanical mixing relative to diffusional mixing, and d is characteristic of the initial striation thickness relative to the distance between the two parallel plates. Equation 7 is subject to the following initial and boundary conditions:

$$\text{I.C.} \quad \left.\begin{array}{ll} C = 1, & 0 \le \xi \le \frac{1}{2} \\ C = 0, & \frac{1}{2} < \xi \le 1 \end{array}\right\} \tag{10}$$

$$\text{B.C. 1.} \quad k\tau d \frac{\partial C}{\partial \xi} = \frac{\partial C}{\partial \eta} \quad \text{at} \quad \eta = 0, 1 \tag{11}$$

$$\text{B.C. 2a.} \quad \frac{\partial C}{\partial \xi} = 0 \quad \text{at} \quad \xi = 0, 1 \tag{12}$$

for diffusion only,

$$\text{B.C. 2b.} \quad \left.\begin{array}{ll} \frac{\partial C}{\partial \xi} = 0 & \text{at} \quad \xi = 0 \\ C = 0 & \text{at} \quad \xi \ge \frac{1}{2} \end{array}\right\} \tag{13}$$

for diffusion-controlled reaction $A + B \longrightarrow$ product.

Note that Eq. 11 asserts the impermeability of the solid boundaries and is derived from $(\partial C/\partial \eta')_{\xi'} = 0$. Before Eq. 7 is solved numerically using the ADI method, let us summarize the pertinent equations resulting from a stretch model in which diffusion is assumed to occur across the striation thickness, i.e., in the direction specified as x' and x in Figure 1. Note that $x \equiv x'/s$ and that s decreases continuously with flow time. Under the same conditions as given above for the two-dimensional diffusion case, the concentration field is described by the following equation

$$\frac{\partial C}{\partial \lambda} = \frac{\partial^2 C}{\partial x^2} \tag{14}$$

where the warped time $\lambda \equiv \int_0^{\tau'} [D/s^2(t)]\, dt$. Equation 14 is subject to

$$\text{I.C.} \quad \left.\begin{array}{ll} C = 1, & 0 \le x \le \frac{1}{2} \\ C = 0, & \frac{1}{2} < x \le 1 \end{array}\right\} \tag{15}$$

$$\text{B.C. a.} \quad \frac{\partial C}{\partial x} = 0 \quad \text{at} \quad x = 0, 1. \tag{16}$$

for diffusion only, and

$$\text{B.C. b.} \quad \left.\begin{array}{ll} \frac{\partial C}{\partial x} = 0 & \text{at}\, x = 0 \\ C = 0 & \text{at}\, x \ge \frac{1}{2} \end{array}\right\} \tag{17}$$

for diffusion-controlled reaction $A + B \longrightarrow$ product.

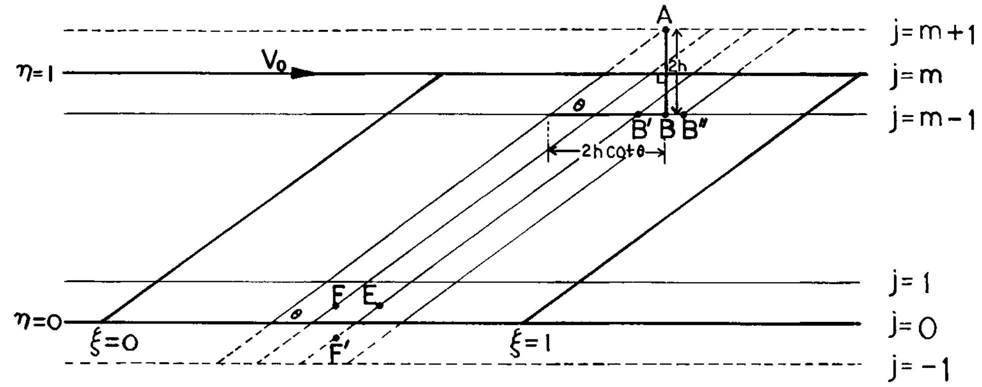

Figure 2. Treatment of boundary conditions in numerical computation.

Hence, the concentration field for the case of diffusion only is (Crank, 1975)

$$C = \frac{1}{2} + \frac{2}{\pi}\sum_{n=1}^{\infty}\frac{(-1)^{n+1}}{2n-1} \cdot \cos\left[(2n-1)\pi x\right] e^{-(2n-1)^2\pi^2\lambda}, \quad 0 \le x \le 1 \tag{18}$$

and for the case of diffusion-controlled reaction,

$$\left.\begin{aligned} C &= \frac{4}{\pi}\sum_{n=1}^{\infty}\frac{(-1)^{n-1}}{2n-1} \cos\left[(2n-1)\pi x\right] e^{-(2n-1)^2\pi^2\lambda}, \quad 0 \le x \le \frac{1}{2} \\ C &= 0, \quad \frac{1}{2} < x \le 1 \end{aligned}\right\} \tag{19}$$

Two level, alternating-direction finite-difference equations were used to approximate Eq. 7, which was discretized following McKee and Mitchell's scheme. We imposed a uniform spacing h in coordinates ξ and η and time step ℓ and solved Eq. 7 numerically in terms of

$$C(\tau, \xi, \eta) = C(n\ell, ih, jh) = C^n_{i,j} \tag{20}$$

where n, i, and j are integers with $n \ge 0$ and $0 \le i$, $j \le m$ $(\equiv 1/h)$. Boundary conditions involving partial derivatives with respect to ξ, Eqs. 12 and 13, were treated using the central-difference formula. However, with the numerical divergence problem the boundary condition given as Eq. 11 was not employed as it is. Instead, its original form prior to nondimensionalization, i.e., $(\partial C/\partial \eta')_{\xi'} = 0$, was used. The procedure is sketched in Figure 2. The idea is based on the nonpenetration of materials across solid boundaries. The concentrations at points on the imaginary boundaries, $j = -1$ and $j = m + 1$, should have the same values as their perpendicular projections on $j = 1$ and $j = m - 1$, respectively, so that no materials could diffuse across either boundary. Hence, the concentration at point A is equal to that at B, which is determined from the interpolation of

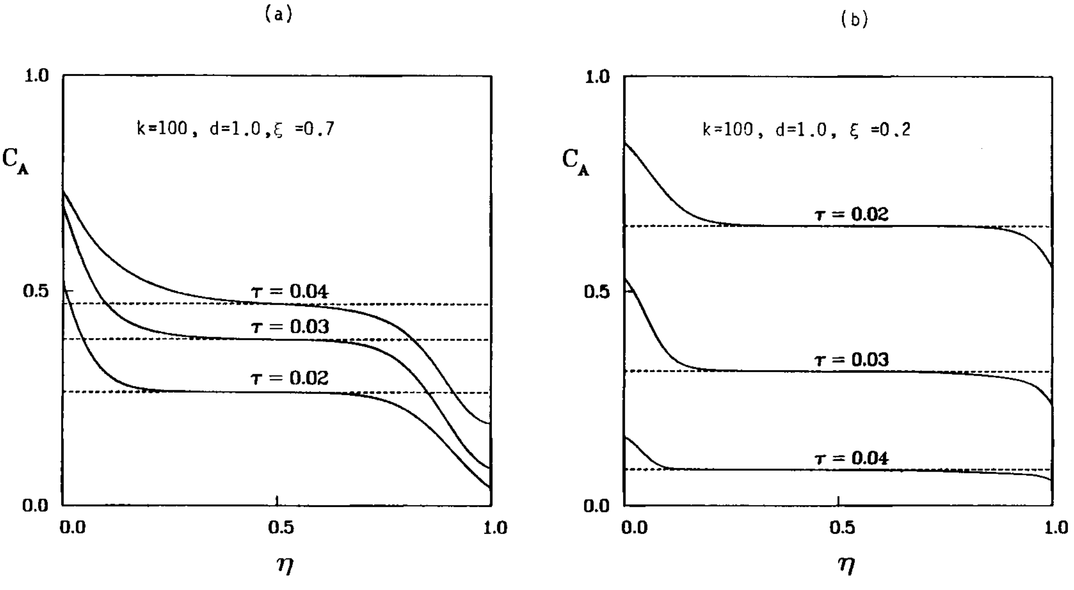

Figure 3. Time evolution of concentration profiles.

a. Nonreacting system
b. Reacting system

concentrations at B' and B''. Mesh points B' and B'' depend on the stretching angle θ, which decreases as time progresses. As we can see from the geometric sketch

$$\cot\theta = \frac{v_0\tau'}{H} = G\tau' = k\tau d^2 \tag{21}$$

By treating Eq. 11 in this manner the numerical instability problems were avoided.

The local truncation error in this scheme is on the order of $(h^2 + \ell)$. In our computations the number of grid points m was 40, 80, or 160, and the mesh ratio r $(\equiv \ell/h^2)$ between 1.25 and 0.01, depending on the value of k and d. In most cases $m = 80$ and $r = 0.625$ were adopted. To solve the difference equations one needs the solution of two tridiagonal sets of equations, which was obtained using Thomas's algorithm (Henley and Seader, 1981). The concentration distributions were computed for k varied from 1 to 10^6, and d from 1 to 10.

Results and Discussion

The computed results are used in the following discussion to investigate the accuracy of the stretch model in representing the concentration field as parameter values are varied. Figure 3 shows the evolution of concentration fields in both nonreacting $(0 \le \xi \le 1.0)$ and reacting $(0 \le \xi \le 0.5)$ systems for $k = 100$ and $d = 1.0$. It is clear that except for the region around $\eta = 0.5$, the stretch model prediction is nowhere close to reality. To reveal the effects of k for nonreacting systems, we plot in Figure 4 for $d = 1.0$ and k ranging from 1 to 10^6 at time τ when about 50% of the equilibrium condition (namely, $C_A = 0.5$ for all $0 \le \xi \le 1.0$) is reached. It appears that the accuracy of the stretch model can be explained by considering the relative contributions to the resultant concentration field from mechanical and diffusional mixing. For $k = 10^6$, where mechanical mixing turns out to dominate diffusional mixing, the model is quite accurate except in the vicinity of $\eta = 0$ or 1. Presumably, the observed error of the stretch model prediction is mainly caused by the material stretching. That the concentration gradient $(\partial C/\partial\eta)_\xi$ at $\eta = 0$ and 1 is negative is justifiable with the reflective boundary condition depicted in Figure 2. For instance, the impermeable solid boundary at $\eta = 0$ requires that the concentration at point F be identical to that at point F'. But the concentration at point E, which along with point F' lies along a line defined by a constant ξ, is lower than that at point F. Thus, $(\partial C/\partial\eta)_\xi < 0$. As the k value is reduced to 10^4 and 10^2 the computed

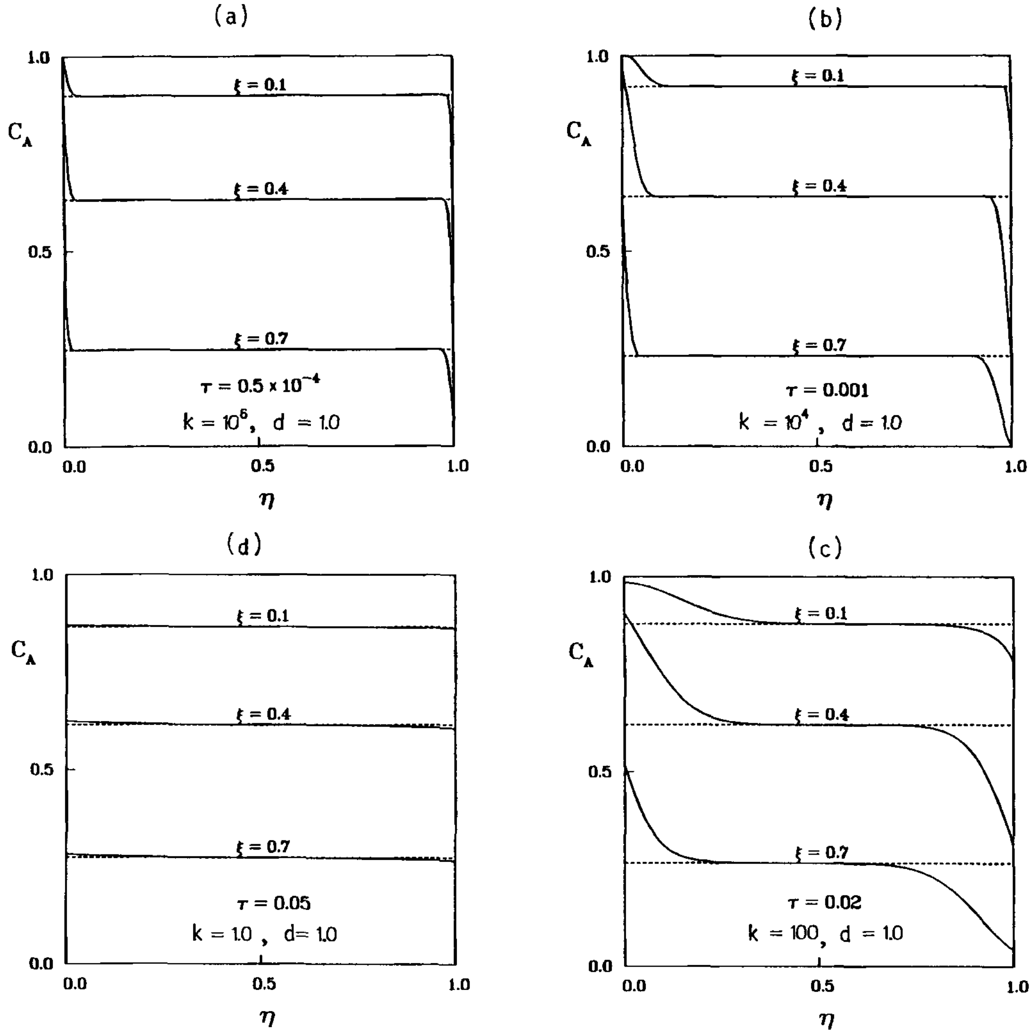

Figure 4. Effects of k values on the accuracy of stretch model predictions in nonreacting systems.

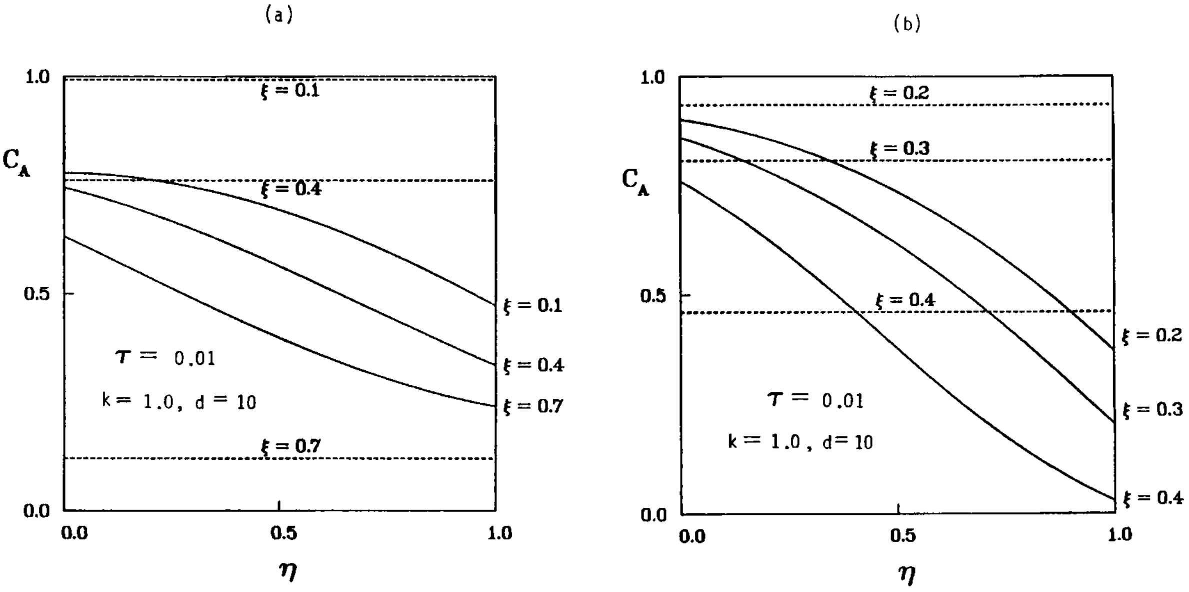

Figure 5. Evaluation of stretch model prediction.

a. Nonreacting system
b. Reacting system

results suggest that the concentration gradients created by mechanical mixing are consistently suppressed by diffusional mixing, which obviously has taken place since more time is allowed. However, as the k value is further reduced to 1, the extent of mechanical mixing is rather small, so that the stretch model representation deviates from reality by only ±1%. Therefore, one is led to conclude that only at extremely high or low mechanical mixing relative to diffusional mixing can the stretch model represent actual concentration distributions satisfactorily. In the former situation, there is almost no time allowed for diffusion to take place, while in the latter there is almost no stretching, so that free diffusion across a finite slab is predominant. However, as the value of d is increased to 10 while keeping $k = 1$, the stretch model representation turns out to be rather poor (Figure 5). This is anticipated in view of the extent of stretching measured by $\theta = \cot^{-1}(k\tau d^2) = 45°$ in Figure 5 being higher than that encountered in Figure 4d, for which $\theta = 87°$. Specifically, the higher concentration gradient in the η direction created by mechanical mixing coupled with the resultant diffusional mixing is responsible for the differences observed between Figures 5 and 4d. Hence, given identical k values, the lower the value of d the more accurate the stretch model.

In summary, the present work has carried out an evaluation of the stretch model applied in the context of mixing with chemical reaction. The two dimensionless groups, k and d, emerging naturally from nondimensionalizing Eq. 2, serve as a basis for justifying the applicability of the model. For more complex fluid flows the importance of mechanical mixing relative to diffusional mixing, as well as the initial striation thickness relative to the mixer-reactor dimension, should be assessed before the stretch model can be employed to simplify the concentration equation via warped time transformation. The method and results reported here for simple shear flow should be instrumental in such assessment. Furthermore, by way of the example of simple shear we have found the inadequacy of the one-dimensional stretch model in the description of fluid mixing. One could then draw a sensible conclusion that in a more complicated flow situation the two-dimensional description should be a better approximation of reality than the stretch model currently in wide application.

Notation

A, B = chemical species
C = concentration
D = molecular diffusivity
d = defined as s_O/H
f = a numerical parameter in computation
G = velocity gradient
h = spacing in coordinates ξ and η
H = distance between parallel plates
k = defined as H^2G/D
ℓ = time step
m = mesh number
Re = defined as Hv_O/ν
r = mesh ratio, ℓ/h^2
s = striation thickness
Sc = defined as ν/D
t = a dummy integration parameter
v_O = constant velocity of the upper plate
x' = coordinate defined in Fig. 1
x = defined as x'/s

Greek letters

η', ξ' = coordinates defined in Fig. 1
η = defined as η'/H
ξ = defined as $[\xi' - \tau' v_\xi(\eta')]/s_O$
τ' = real time
τ = defined as $\tau' D/s_O^2$
λ = warped time, defined as $\int_0^{\tau'} [D/s^2(t)]\,dt$

Subscripts

A, B = species identification.
O = initial value.

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