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Experimental and theoretical scaling laws for transverse diffusive broadening in two-phase laminar flows in microchannels

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This letter quantifies both experimentally and theoretically the diffusion of low-molecular-weight species across the interface between two aqueous solutions in pressure-driven laminar flow in microchannels at high Péclet numbers. Confocal fluorescent microscopy was used to visualize a fluorescent product formed by reaction between chemical species carried separately by the two solutions. At steady state, the width of the reaction–diffusion zone at the interface adjacent to the wall of the channel and transverse to the direction of flow scales as the one-third power of both the axial distance down the channel (from the point where the two streams join) and the average velocity of the flow, instead of the more familiar one-half power scaling which was measured in the middle of the channel. A quantitative description of reaction–diffusion processes near the walls of the channel, such as described in this letter, is required for the rational use of laminar flows for performing spatially resolved surface chemistry and biology inside microchannels and for understanding three-dimensional features of mass transport in shearing flows near surfaces.

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Chemical patterning at the interface of two miscible fluids flowing laminarily and in parallel provides a method of microfabrication: examples include fabrication of microelectrodes¹ and patterning of mammalian and microbial cells.² The rational use of laminar flow for patterning and fabrication inside microchannels requires an improved understanding of the convective–diffusive transport processes near the walls of the channel. The most relevant component of this transport occurs across the interface between the flowing solutions *transverse* to the direction of the flow. In this letter, we combine experiments and theory to quantify this transverse diffusive transport as a function of the local flow speed and velocity profile.

Pressure-driven flow in microchannels generally is laminar, since the Reynolds numbers Re are small (under 100). Laminar flow patterning occurs by a chemical reaction at the interface between two miscible fluids flowing through a channel.¹ Here, we use confocal fluorescent microscopy to visualize the fluorescent product of the reaction between two nonfluorescent chemical species carried separately by the two flowing solution streams. The spatial extent of transverse diffusive mixing can be decreased by increasing the average flow speed (so long as the Reynolds number is small enough that the flow is laminar). We demonstrate experimentally that, at steady state near the top and bottom walls of the channel, the extent of transverse diffusive mixing across the fluid–fluid interface scales as the one-third power of the axial distance (z) along the channel and scales inversely as the one-third power of the maximum flow velocity. The extent of such mixing near the middle of the channel, where the flow speed is nearly uniform, scales as the more familiar one-half power of the axial distance (z) along the channel

and inversely as the one-half power of the average flow velocity. Theoretical arguments are given for these two different scaling laws.

To test the predictions of the theory, we have fabricated microfluidic channels using the “rapid prototyping” technique described previously,³ and used a single syringe pump to drive the fluids into the two inlets at a constant flow rate. Rather than deducing the extent of diffusive mixing from the flow profiles, we chose a direct approach—visualization of the region of diffusive mixing using confocal fluorescent microscopy (Leica TCS). Fluo-3 (m.w.=770; we used the pentammonium salt) is a commercially available, nonfluorescent compound that forms a strongly fluorescent 1:1 complex with a calcium ion ($K_d=0.39\ \mu\text{M}$).⁴ The formation of the complex is diffusion controlled, therefore, we could visualize the region of diffusive mixing by observing the concentration of this complex near the interface between flowing aqueous solutions ($pH\approx 6.5$) of $5\ \mu\text{M}$ fluo-3 and $1\ \text{mM}$ CaCl_2 (Fig. 1). All experiments were conducted at room temperature, and it took 2–4 s to acquire each image in the line-averaging mode. At a given axial distance z from the point where the streams join, the diffusive mixing is more extensive (i.e., the fluorescent region is broader) in the slower-moving fluid near the wall of the channel than in the middle of the channel. At low flow velocities we observe some interdiffusion in the dead volume of the Y junction [Fig. 1(b)], and we assume that its effect on the scaling behavior is negligible.⁵ Below, we argue that the transverse diffusive broadening—which, ideally, should be limited in chemical patterning applications—obeys power-law behavior as a function of distance along the channel z and the average flow speed U_a .

In order to quantify the scaling relationships that describe diffusive mixing in the laminar flows present in our experiments, we use the convective–diffusion equation for

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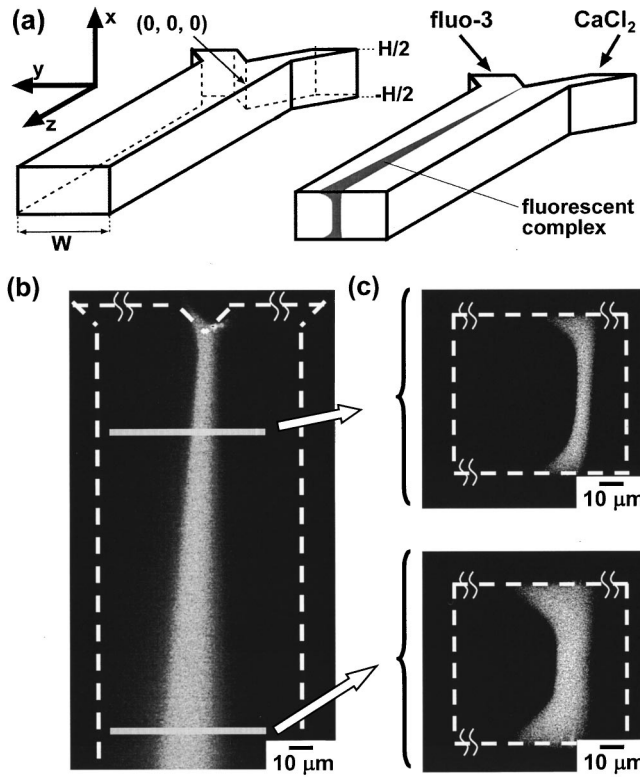


FIG. 1. (a) Schematic drawing of the experiment used to generate fluorescence in the microchannel, and the coordinate system employed. The dashed lines indicate the internal edges of the channel. H is the height of the channel. The origin of the coordinate system is located in the middle of the vertical edge shared by both channels in the Y junction. (b) and (c) Data obtained for the fluo-3/ Ca^{2+} system by confocal fluorescence microscopy in: (b) A $100 \times 250 \mu\text{m}^2$ yz slice obtained $12 \mu\text{m}$ from the top of the channel (average velocity of the flow $U_a = 0.7 \text{ cm/s}$, $\text{Re} = 0.4$; the Reynolds number $\text{Re} = \rho U_a H / \mu$, where ρ is the density of the solutions and μ is the viscosity). Some mixing is visible in the area of the dead volume of the Y junction. (c) Corresponding $90 \times 90 \mu\text{m}^2$ cross sections in the xy plane of the channel obtained $50 \mu\text{m}$ (top) and $200 \mu\text{m}$ (bottom) from the Y junction.

steady-state transport of chemical species, with concentration $c(x, y, z)$, in the flow field $\mathbf{u}(x, y, z)$, with velocities scaled by the average flow speed U_a and lengths scaled by the channel height H ,

$$P\mathbf{u} \cdot \nabla c = \nabla^2 c \equiv \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) c \quad (1)$$

[the coordinate system employed is shown in Fig. 1(a)]. Here, D is the diffusion coefficient, and P is the dimensionless Péclet number, $P = U_a H / D$, which compares the typical time scale for diffusive transport to that for convective transport in a channel with a given height H . P is large in our experiments (for $H = 6 \times 10^{-5} \text{ m}$, $D = 10^{-9} \text{ m}^2/\text{s}$, $U_a = 0.0025\text{--}0.5 \text{ m/s}$, $P = 150\text{--}30\,000$). Therefore, in Eq. (1) we neglect diffusive transport along the flow (z) direction.

If the transverse diffusive broadening $\delta(z)$ is monitored as a function of distance z down the channel, then the comparison of the high Péclet number to the ratio z/H distinguishes two regimes. For $P \gg z/H \gg 1$ (our experiments are in this regime), the diffusive broadening $\delta(z)$ also depends on x [as proposed by Kamholz *et al.*⁶ and shown in Fig. 1(c) here] because the nonuniform velocity profile affects the diffusion process. For $z/H \gg P \gg 1$, $\delta(z)$ is approximately independent of x because diffusive transport on the scale H eliminates the effects of the nonuniform velocity profile. Kamholz

et al. analyzed their data assuming the latter regime, and their experiments were indeed conducted at lower $P/(z/H)$ ratios.⁶ In this respect, the two approaches are complementary to each other.

In the present study, the velocity field is the uniaxial Poiseuille-like flow (in a rectangular channel with width W and height H), which, although known in the form of a Fourier series, is sufficiently complicated that no simple analytical solution can be obtained to Eq. (1).

Instead of obtaining a numerical solution, we indicate two scaling laws for transverse diffusion that are natural consequences of Eq. (1) for channel flow. We utilize the fact that the flow profile at the interface ($y=0$) is approximately parabolic in x , with a maximum velocity in the middle of the channel ($x=0$), and with zero velocity at the walls ($x = \pm H/2$). Near the middle of the channel—where $[(x/(H/2))^2] \ll 1$ and $[y/(W/2)]^m \ll 1$, $m \sim 4$ for our channels⁷—the velocity is nearly uniform and approximately equal to the maximum velocity $U_m (\approx 2U_a)$. Also, near the top and bottom boundaries, $\tilde{x}^2 \ll 1$ and $[y/(W/2)]^m \ll 1$, where $\tilde{x} = H/2 - |x|$, the velocity is approximately a linear function of the x -position, $\mathbf{u}(x, y, z) = G\tilde{x}\mathbf{e}_z$. The shear rate, $G \text{ (s}^{-1}) \approx 8(U_a/H)$, and \mathbf{e}_z is the unit vector along the axis of the channel. It is this linear variation of velocity (that increases from zero at the solid boundary) that gives rise to the one-third power law for transverse diffusion near the walls of the channel. Near the center of the channel, $\mathbf{u} = U_m \mathbf{e}_z + O[(x/(H/2))^2]$, so we can rewrite Eq. (1) in dimensional form $U_m \partial c / \partial z = D \nabla^2 c$. Thus, in the center of the channel, the transverse mass transport is simple diffusion, and by dimensional reasoning, this equation implies that an initial concentration distribution broadens in x and y as a function of the z position according to $\delta(z) \propto (Dz/U_a)^{1/2}$. We defined the thickness $\delta(z)$ of the interfacial diffusion layer as the distance over which the concentration of the fluorescent complex falls to 0.2 of its maximum value.

A special case of transverse diffusion in the linear flow near a fixed boundary is known as the L  v  que problem.⁸ This problem treats diffusion perpendicular to the boundary (along \mathbf{e}_x) and across a linear flow field, $\mathbf{u} = G\tilde{x}\mathbf{e}_z + O[(\tilde{x}/(H/2))^2]$. We will extend this classical result to account for diffusion in the other transverse direction, \mathbf{e}_y . In the traditional two-dimensional L  v  que problem, the approximate form of Eq. (1) for high-P  clet-number flow with $\partial c / \partial y = 0$ is $G\tilde{x} \partial c / \partial z = D \partial^2 c / \partial \tilde{x}^2$. Hence, diffusive transport in x will depend on the distance down the channel z as $\delta(z) \propto (Dz/G)^{1/3} \propto (DH/U_a)^{1/3}$.

We must now relate this standard L  v  que result for the diffusive transport along \mathbf{e}_x to the diffusive transport along \mathbf{e}_y , as it is this (transverse) variation that leads to mixing and to the subsequent chemical reaction of the species present in the two adjacent laminar flows in our experiments. To do this, we rewrite Eq. (1) in cylindrical coordinates with the dimensionless similarity variable $\eta = r/(Dz/G)^{1/3}$. Here, $\tilde{x} = r \cos \theta$ and $y = r \sin \theta$ for $-\pi < 2\theta < \pi$ and $C(\eta, \theta) = c(v, \theta, z)$:

$$-\frac{\eta^2 \cos \theta}{3} \left(\frac{\partial C}{\partial \eta} \right) = \frac{1}{\eta} \frac{\partial}{\partial \eta} \left(\eta \frac{\partial C}{\partial \eta} \right) + \frac{1}{\eta^2} \frac{\partial^2 C}{\partial \theta^2}. \quad (2)$$

