Wall influence on transport phenomena in microfluidic systems

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Most of microfluidic devices operate in laminar conditions, and in many cases in the creeping (Stokes) regime. Notwithstanding the relative simplicity of hydrodynamics at microscales (compared to the wealth of problems associated with the occurrence of turbulence in bench-scale and ordinary-scale fluidic systems), there are many open issues in microfluidic theory, the understanding of which has a direct impact on the device design and optimization.

Among these open problems, a crucial issue is the influence of walls on transport and dispersion of solutes and reacting species, and the scope of this presentation is to review some of the most relevant theoretical approaches developed for understanding transport, dispersion and related phenomena in microsystems.

Just because of the strong connection with the realization of microflow devices, the hydrodynamic and transport modelling in microfluidics involves mainly finite and bounded flow/mixing domains V, characterized by the presence of impermeable walls to solute transport and, in the case of open (inflow/outflow) systems, by the presence of an inlet and an outlet section (e.g., in open capillary microdevices, microchromatographic equipment, static micromixers, tubular microreactors).

Throughout this presentation we consider almost exclusively inflow/outflow systems, in the presence of a liquid incompressible stationary flow.

Consider a generic inflow/outflow device, as represented in Fig. 1, possessing a single inlet and a single outlet section, denoted as S_{in} and S_{out}, respectively. Let S_w represent the solid boundaries (walls) of the device. Within the control volume, a laminar velocity field is defined, which can be generated by different driving forces, such as pressure drops, electro-osmosis, magnetohydrodynamic effects. Typically, the flow is either known analytically, when the device possesses a simple structure (e.g. channel or annular flows), or obtained through CFD simulations, in the case of more complex geometries. In most cases of practical interest, it is possible to identify an average streamwise direction of the flow, which we denote as z, with $0 \leq z \leq L_z$, L_z being the overall length of the device. Accordingly, a generic cross-section of the device, (denoted as S_z) orthogonal to the z axis is also defined for any z. The volume delimited by the inlet section S_{in}, the generic section S_z, and the device walls, is henceforth referred to as V_z. It is worth noting, that in many applications, the geometric structure of S_z is position-dependent and can be made of a union of disjoint domains. Henceforth, we denote with $\mathbf{x} = \mathbf{x}_\perp + z\,\mathbf{e}_z$ a generic point of the control volume, where the subscript "$\perp$" indicates the cross-sectional coordinates obtained by the projection of $\mathbf{x}$ onto the generic cross-section at z, and where $\mathbf{e}_z$ is a unit vector along the z-axis. Single phase mixing is described by the advection-diffusion equation

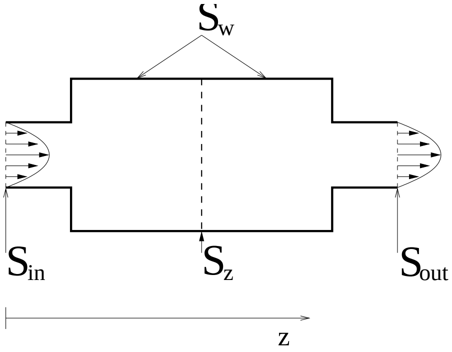

Figure 1: Schematic representation of the inflow-outflow device

$$\frac{\partial \phi}{\partial t} = \mathcal{L}_{\mathbf{v},Pe}[\phi] = -\mathbf{v}(\mathbf{x}) \cdot \nabla \phi + \frac{1}{Pe} \nabla^2 \phi \tag{1}$$

where $\phi(\mathbf{x}, t)$ is the concentration of the species being mixed, $\mathbf{v}(\mathbf{x})$ is an incompressible flow which we assume time-independent, and the Peclet number $Pe = \tau_D/\tau_c$ is a dimensionless number representing the ratio between a characteristic time for diffusion to a characteristic time for convection. Equation (1) is equipped with the boundary and initial conditions

$$\phi(\mathbf{x}, t) = \phi_{in}(\mathbf{x}, t) \quad \text{on } S_{in} \tag{2}$$

$$\nabla \phi \cdot \mathbf{n} = 0 \quad \text{on } S_w \cup S_{out} \tag{3}$$

$$\phi(\mathbf{x}, 0) = \phi_0(\mathbf{x}) \quad \mathbf{x} \in V \tag{4}$$

which assign a prescribed concentration profile at the inlet, zero diffusive flux at the system walls and at the outlet section, and a given initial profile within the control volume, respectively.

Essentially, three different classes of transport problems can be formulated in open flow systems involving a single fluid phase:

- transient phenomena within the flow domain V;
- relaxation dynamics towards stationary conditions in the case where the inlet condition is maintained steady;
- the stationary response of the flow device.

In all of these classes, the structure of the velocity field near the walls, and the interaction between advection and diffusion near the stagnation points at the solid walls play a major qualitative role in the dynamical and stationary response of the system.

Transient phenomena: dispersion in finite-length microchannels

The typical phenomenology involving transient phenomena in finite-length channels is related to solute dispersion. Dispersion phenomena in microchannels and microflow devices control the performance of many hydrodynamical and chemical processes in micro-Total Analysis and Lab-on-Chip Systems, chemical reactions, microseparations, microchromatographic measurements.

The recent Literature on fluid dispersion in microchannels has been mainly focused on the analysis and prediction of the Taylor-Aris dispersion coefficient, and on the influence of geometric parameters and of cross-sectional flow in the Taylor-Aris [1] dispersion regime [2-4]. In point of fact, Taylor-Aris theory applies to infinitely extended channels, and is not suited for predicting finite-size effects occurring in a microcapillary of finite length, occurring for very high Peclet number.

In this presentation we show that:

1. Whenever a finite length microcapillary is considered, a regime transition occurs for from the classical Taylor-Aris regime to a new regime referred to as *convection-dominated transport regime*, occurring for $Pe_{\text{eff}} = Pe/\alpha^2 > 5$, where $Pe = V_{\text{ref}} L_z/D$, $\alpha = L_z/W$ (V_{ref} being the characteristic mean axial velocity, D the solute diffusion coefficient, and W the characteristic cross-sectional lengthscale).

2. For parallel flows in microchannels, the average moments $m^{(n)}$, $n = 1, 2, \ldots$ of the outlet concentration profile,

$$m^{(n)} = \frac{1}{\text{meas}(S_{out})} \int_{S_{out}} t^n \, \phi(t, \mathbf{x})|_{z=L_z} \, dS \ , \quad n = 1, 2, \ldots \tag{5}$$

   (associated with a cross-sectionally uniform impulsive inlet condition) scale as a power-law of the effective Peclet number Pe_{eff}

$$m^{(n)} \sim Pe_{\mathit{eff}}^{\zeta(n)} \ , \qquad Pe_{\text{eff}} \to \infty \tag{6}$$

   where $\zeta(1) \geq 0$, and $\zeta(n) > 0$ for $n > 1$, The exponents $\zeta(n)$ depend on the local behavior of the velocity field at the solid walls, on the dimensionality of the flow domain (i.e. whether two-dimensional or three-dimensional domains are considered), and on the possible lack of Lipschitz-continuity of the channel boundary. For instance, Fig. 2 shows the cross-section and the contour plot of a cross-sectional boundary which is not Lipshitz continuous. For a cylindrical capillary one obtains $\zeta(1) = 0$, $\zeta(2) = 1/3$, for a rectangular duct $\zeta(1) = 0$, $\zeta(2) = 1/2$, while a channel with the cross-section depicted in Fig. 2 $\zeta(1) = 1/6$, $\zeta(2) = 5/6$. In this case, the lack of Lipschitz continuity enhances dispersion.

   The case where $\zeta(1) = 0$, should be understood as the fact that the first-order moment displays a logarithimic divergence with Pe_{eff},

$$m^{(1)} \sim C \, \log(Pe_{\text{eff}}) \ , \qquad Pe_{\text{eff}} \to \infty \tag{7}$$

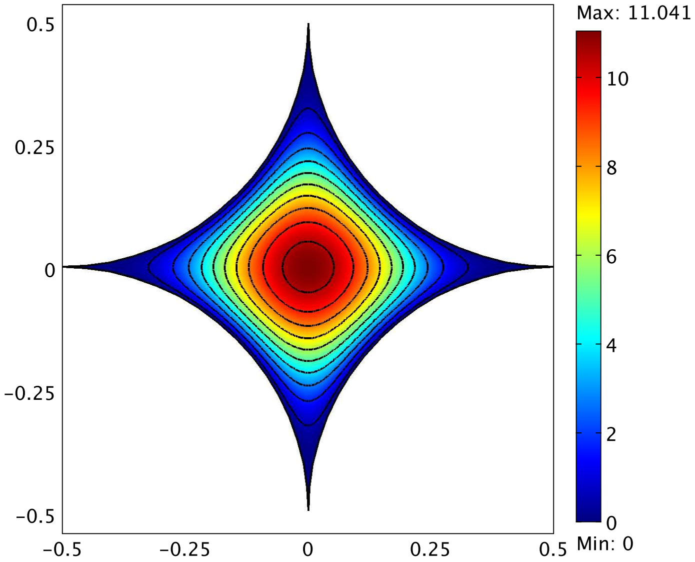

Figure 2: Contour plot of the axial velocity, solution of the Stokes equation, for a cusp channel the boundary of which is not Lipschitz continuous.

where C is a constant. We provide the result of a scaling theory for predicting the behavior of $\zeta(n)$, supported by simulation results.

3. Hydrodynamic chromatographic microchannels for relatively small aspect ratios (corresponding to the analytical application referred to as wide-bore chromatography [5]) can be used as a transport-based experimental set-up for determining the possible occurrence of slip velocity conditions at the channel solid walls, directly from the analysis of the dispersion properties of the outlet chromatograms.

Relaxation phenomena

Relaxation phenomena towards the stationary response of a microflow device has been addressed in [6], by considering the Frobenius eigenvalue of the advection-diffusion operator $\mathcal{L}_{\mathbf{v},Pe}$ associated with simple parallel flows. In inflow-outflow systems, the Frobenius eigenvalue is tipically different from zero and quantifies the timescale to reach the steady-state profile which is not, in general a perfectly mixed state. Therefore, the Frobenius eigenvalue does not quantify mixing efficiency, but rather provides a preliminary necessary requirement to motivate the analysis of mixing efficiency at steady-state, meaning that an excessively long timescale would suggest that a steady-state assessment of mixing efficiency might be inadequate, and that the whole transient behavior should be considered instead.

Stationary response of a microfluidic system

The stationary response of a fluidic system is related to several different applications of microflow devices. In particular:

1. the formation of mixing layers when two fluid streams homogenize in a microchannel channels (e.g. in a T-microjuntion) [7-8].
2. the homogenization properties of micromixers (static micromixers) operating under steady conditions.

By focusing attention on the second of these issues, we discuss the spectral characterization of mixing in simple microflow systems, and how, localization theory of the eigenfunctions of the advection-diffusion operator may unveil the occurrence of different mixing regimes, and the behavior of the homogenization exponents as a function of the Peclet number [9-10]. Examples of magnetohydrodynamic micromixers are also addressed and reviewed [11].

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