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Foundations of laminar chaotic mixing and spectral theory of linear operators

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Abstract

This article addresses the potentiality and the range of application of the spectral theory of mixing as a simple and objective way to define in a rigorous way mixing performance and stirring efficiency. Attention is mainly oriented to the class of laminar flows which is the typical condition occurring in micromixers and flow microdevices. The application of the theory is highlighted by considering the cavity flow as a case study.

Keywords: Mixing; Advection–diffusion equations; Laminar flow; Microdevices; Transport processes; Diffusion

1. Introduction

Mixing theory has attracted the interest of fluid dynamicists and engineers for its relevance in the understanding some of the fundamental problems involving fluid flows, and for its practical impact in connection with chemical, pharmaceutical and other manufacturing industrial processes (Baldyga and Bourne, 1999). The first quantitative approach to mixing dates back to Danckwerts (1952, 1958), who introduced global indices of the "degree of mixedness" such as the intensity of segregation, which essentially corresponds to the concentration variance of an advecting–diffusing dye carried by the flow, and the linear scale of segregation, yielding the average "diameter" of the partially mixed structures. Owing to its conceptual simplicity and experimental feasibility, this approach is still largely used to quantify mixing, especially when complex flows (e.g. flows in industrial equipment) are to be dealt with (Müller et al., 2004).

About two decades ago, a new approach to characterize mixing in flow systems spread out in the fluid dynamics community (Aref, 1984; Ottino, 1989). This approach stemmed from the observation that even simple, large-scale velocity fields can generate mixing structures at arbitrarily small scale. This idea constituted a major breakthrough for people involved in fluid mixing. In fact, until then, the wordings "mixing flow" and "turbulent flow" were used almost interchangeably. Methods and quantities derived from dynamical system theory were directly applied to simple flows, laboratory equipment, stirred tanks, static mixers, and constituted what is now referred to as the *Lagrangian*, *or kinematic theory of laminar mixing* (Ottino, 1989; Beigie et al., 1994). The kinematic approach has contributed to a better understanding of existing mixing devices as well as to the rational design of new ones (Harvey and Rogers, 1996; Zalc et al., 2001).

In the last few years, the development of technology for designing flow circuits at micro lengthscales (typically ranging form 1 to 100 μm), have generated renewed interest in the foundations of laminar chaotic mixing, since the flow regime in microfluidic devices is typically laminar. Microflow technology opens up new perspectives in approaching a rational design of industrial operations (Hessel et al., 2004), in the design of new micromixers (Hessel et al., 2005; Aubin et al., 2005), as well as in designing processes involving biological molecules, e.g. those associated with DNA manipulations (Tay, 2002).

This novel and promising field forces the research community involved in laminar mixing theory to a critical analysis of the state of the art of the theory, with the scope of identifying the fundamental issues that should be addressed in the next few years and, hopefully, of giving a definite answer to them. In this perspective, i.e., in the light of concrete application to microdevices, even the kinematic approach suffers some conceptual drawbacks. The most important is that it does not account for diffusion, which is, ultimately, the only mechanisms that

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ensures the approach toward a spatially homogeneous mixture. Also, even though the kinematic approach defines objective indices assessing the quality of the mixing process within each of the invariant sets associated with the flow, it does not provide any information about how this region-dependent indices should be combined together to construct a global measure of mixing in the flow domain as a whole. This last issue is clearly connected with the first in that the only flux exchange of the scalar entity (be it a chemical species or energy) that is being transported between kinematically invariant sets relies exclusively on the presence of the diffusion mechanism.

In this article, we discuss the fundamental physical and mathematical aspects associated with laminar mixing and propose a unified approach referred to as *the spectral theory of mixing* (Cerbelli et al., 2003, 2004).

The aim of this article is to show that the spectral approach provides an efficient and complete global characterization of mixing, and can be applied to general flow devices of practical interest. Throughout this article, we consider the two-dimensional cavity flow as a prototypical model for describing and highlighting the theoretical and computational methods. The discussion of the computational issues in the case of three-dimensional flows is briefly addressed in the concluding section.

2. The advection–diffusion equation and flow systems

Let us consider a closed domain Ω representing the mixing space, and the velocity field $\mathbf{v}(\mathbf{x}, t)$ defined on it. The mixing space is filled with an incompressible fluid, so that $\nabla\cdot\mathbf{v}(\mathbf{x}, t)=0$ for any t. The physical origin of $\mathbf{v}(\mathbf{x}, t)$ is immaterial in the present analysis.

Let $\phi(\mathbf{x}, t)$ be a scalar field representing the concentration of the transported entity (which may represent the concentration of some dye or temperature).

The basic phenomenology of mixing is described by means of the advection–diffusion equation for the concentration field $\phi(\mathbf{x}, t)$, which, in dimensionless form reads:

$$\frac{\partial\phi}{\partial t} = -\mathbf{v}\cdot\nabla\phi + \frac{1}{Pe}\nabla^2\phi, \tag{1}$$

where $Pe = VL/D$ is the Peclet number (V, L and D being a characteristic velocity, lengthscale and the diffusivity, respectively), and t and $\mathbf{v}$ are the dimensionless time and velocity field, respectively.

In its ultimate essence, finding optimal mixing conditions expresses into maximizing the interplay between advection and diffusion so as to obtain a homogeneous concentration field ϕ in the shortest possible time.

Since we are considering a closed domain for which $\mathbf{v}\cdot\mathbf{n}|_{\partial\Omega}=0$ at the boundary $\partial\Omega$ of the domain Ω ($\mathbf{n}$ is the normal unit vector to $\partial\Omega$), the boundary conditions for ϕ are of Neumann type: $\nabla\phi\cdot\mathbf{n}|_{\partial\Omega}=0$, and Eq. (1) is equipped with the initial conditions $\phi(\mathbf{x}, t)|_{t=0}=\phi_0(\mathbf{x})$.

Throughout this article, we consider the two-dimensional cavity flow under creeping flow conditions, defined on the two-dimensional unit square $\Omega=\{(x, y)\,|\,0\leqslant x, y\leqslant 1\}$ as a prototypical model flow. The cavity flow stems from a streamfunction $\Psi(x, y)$ ($v_1=\partial\Psi/\partial y$, $v_2=-\partial\Psi/\partial x$, $\mathbf{v}=(v_1, v_2)$), which is the solution of the biharmonic equation $\nabla^4\Psi=0$, equipped with vanishing boundary conditions on Ψ and $\partial\Psi/\partial n$ all over the boundary but on $y=1$, for which $v_1|_{y=1}=\partial\Psi/\partial y|_{y=1}=1$.

For the scope of this article, it is sufficient to consider an approximation for the stream-function of the cavity flow developed by Chella and Ottino (1985), which derives from a first-order expansion of the solution of the biharmonic equation obtained by applying the method of weighted residuals. Within this approximation the streamfunction $\Psi(x, y)$ reads as

$$\Psi(x, y) = Cx^2(1-x)^2k_0(y), \tag{2}$$

where $C=21$, $k_0(y)=A_2\cosh(\delta_1 y)\sin(\delta_2 y)+\sinh(\delta_1 y)[B_1\sin(\delta_2 y)+B_2\cos(d_2 y)]$, where $\delta_1=4.1503$, $\delta_2=2.28582$, and the coefficients A_2, B_1, and B_2 are given by

$$A_2 = \frac{\delta_1\sinh(\delta_1)\sin(\delta_2)}{(\delta_2\sinh(\delta_1))^2-(\delta_1\sin(\delta_2))^2},$$

$$B_1 = -\frac{\delta_1\cosh(\delta_1)\sin(\delta_2)-\delta_2\sinh(\delta_1)\cos(\delta_2)}{(\delta_2\sinh(\delta_1))^2-(\delta_1\sin(\delta_2))^2},$$

$$B_2 = -\frac{A_2\delta_2}{\delta_1}. \tag{3}$$

Within this approximation, the velocity at the boundary is given by $v_1|_{y=1}=Cx^2(1-x)^2$.

The flow associated with the streamfunction Eq. (2) is the autonomous cavity flow, and cannot exhibit Lagrangian chaos since it is two-dimensional and time-independent.

In order to improve mixing, it is natural to consider time-periodic protocols giving rise to Lagrangian chaos. This is one of the simple "rules of thumb" representing the legacy of the kinematic theory of mixing. Let us consider the time-periodic (TP) cavity flow obtained by blinking alternately two steady flows associated with the streamfunction $\Psi^{(1)}(x, y)=\Psi(x, y)$ (giving rise to the velocity field $\mathbf{v}^{(1)}$) for the first half period $T_p/2$, and the streamfunction $\Psi^{(2)}(x, y)=-\Psi(x, 1-y)$ (velocity field $\mathbf{v}^{(2)}$) for the second half period. The TP cavity flow defined above originates from the instantaneous switch of the upper ($y=1$) and lower ($y=0$) walls motion, with a wall velocity oriented in positive x-direction, i.e., $v_1^{(1)}|_{y=1}=v_1^{(2)}|_{y=0}=x^1(1-x)^2$. The assumption of instantaneous switching between the two steady flows represent a fluid dynamic approximation, which is accurate at low Reynolds numbers $Re<1$ (Liu et al., 1994).

Fig. 1(A)–(D) show the Poincaré section (i.e., the global representation of the orbit dynamics $\mathbf{x}^{(n+1)}=P(\mathbf{x}^{(n)})$, where $\mathbf{x}^{(n)}=\mathbf{x}(nT_p)$, $n=0, 1, \ldots$), of the TP cavity flow for several values of the flow period T_p. For lower values of T_p, the mixing space is almost entirely covered with periodic and quasiperiodic orbits (see Fig. 1(A) at $T_p=1$). As the flow period increases, regions of chaotic motion appear, and progressively invade larger and larger portions of the mixing space (see Fig. 1 from (B) to (D)).

For a generic value of the period T_p, the phase portrait of the kinematic equations of motion is characterized by the