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# MIXING, CHAOTIC ADVECTION, AND TURBULENCE

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## 1. INTRODUCTION

### 1.1 *Setting*

The establishment of a paradigm for mixing of fluids can substantially affect the development of various branches of physical sciences and technology. Mixing is intimately connected with turbulence, Earth and natural sciences, and various branches of engineering. However, in spite of its universality, there have been surprisingly few attempts to construct a general framework for analysis. An examination of any library index reveals that there are almost no works—textbooks or monographs—focusing on the fluid mechanics of mixing from a global perspective. In fact, there have been few articles published in these pages devoted exclusively to mixing since the first issue in 1969 [one possible exception is Hill's "Homogeneous Turbulent Mixing With Chemical Reaction" (1976), which is largely based on statistical theory]. However, mixing has been an important component in various other articles; a few of them are "Mixing-Controlled Supersonic Combustion" (Ferri 1973), "Turbulence and Mixing in Stably Stratified Waters" (Sherman et al. 1978), "Eddies, Waves, Circulation, and Mixing: Statistical Geofluid Mechanics" (Holloway 1986), and "Ocean Turbulence" (Gargett 1989). It is apparent that mixing appears in both industry and nature and the problems span an enormous range of time and length scales; the Reynolds number in problems where mixing is important varies by 40 orders of magnitude (see Figure 1).

Virtually everyone agrees that mixing is complicated. However, there is no agreement as to the source of the complications; to a rheologist, the constitutive equation is of paramount importance; to someone in turbu-



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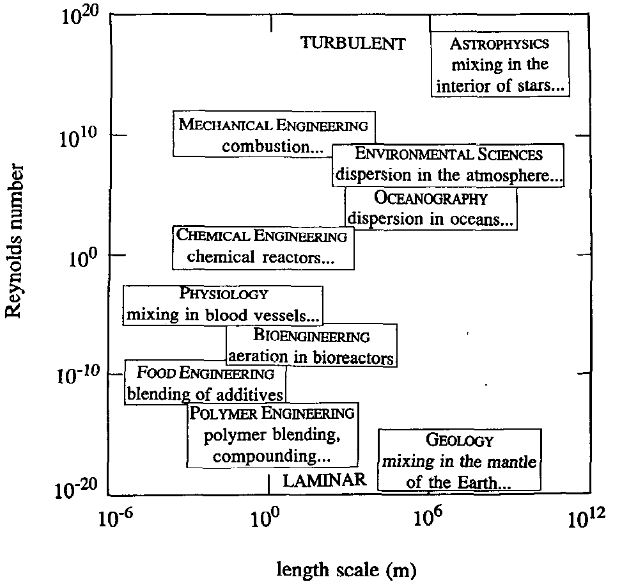


*Figure 1* Spectrum of problems studied in various disciplines in which mixing is important.

lence, it is the daunting complexities of the flow field itself. Given the ubiquity of mixing, it is very unlikely that a single explanation can possibly capture all aspects of the problem. Nevertheless, it is desirable to consider the problem from a general viewpoint. What makes mixing complex? Usually, realistic mixing problems have been regarded as nearly intractable from a modeling viewpoint owing to the complexity of the *flow* fields. Also, in many problems of interest the *fluids* themselves are rheologically complex; this is particularly important in problems with small length scales and inhomogeneous fluids, such as those typically encountered in chemical engineering. In many cases the complexity of both fluids and flows often complicates the entire picture to the point that modeling becomes intractable if one wants to incorporate all details at once. For this very reason, mixing problems have been attacked traditionally on a case-by-case basis. However, recent experimental studies and the merging of kinematics with dynamical systems and chaos are providing a paradigm for the analysis of mixing from a rather general viewpoint.

The objective of this article is to provide an overview of mixing and chaotic advection. Since one general reference on this topic is available (Ottino 1989a) we focus primarily on open problems as well as on concepts that present potential difficulties. [An introductory review to chaotic mix-

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ing is given in Ottino (1989b); a more advanced treatment, including a complete background of kinematics and chaotic dynamics as well as many examples, is given in Ottino (1989a).] In several sections the material is rather speculative (there are several references to on-going work, in particular several PhD dissertations), and the emphasis throughout is on systems where chaos is widespread rather than on small perturbations from integrability. The concepts presented can be viewed in two different ways: In the context of natural sciences the objective is understanding; in the context of engineering applications the objective is exploitation and design. Much of the understanding of chaotic mixing can be based on relatively simple pictures; in turn, these pictures can be exploited in the design of novel mixing devices.

Mixing is intimately related to stretching and folding of material surfaces (or lines in two dimensions), and theory can be couched in terms of a kinematical description. However, even though the kinematical foundations have been available for a long time (e.g. see Truesdell 1954, Truesdell & Toupin 1960), such a viewpoint was not advocated until rather recently [Ottino et al. 1981, Ottino 1982; naturally there are exceptions (e.g. Batchelor 1952)]. In a number of cases, the existing kinematical foundations are sufficient and it is possible to compute the stretching of material elements exactly—for example, in steady curvilineal flows and slowly varying flows (Chella & Ottino 1985a). Even though these cases are rather special, they are encountered routinely in polymer-processing applications (Ottino & Chella 1983, Chella & Ottino 1985b). However, they lead to poor mixing unless special precautions are taken.

Chaotic motions provide a natural way of increasing the mixing efficiency of flow. The key idea is that the Eulerian velocity field,

$$(d\mathbf{x}/dt)_{\mathbf{X}} = \mathbf{v}(\mathbf{x}, t) \quad \text{with} \quad \nabla \cdot \mathbf{v} = 0, \tag{1}$$

admits chaotic particle trajectories for relatively simple right-hand sides $\mathbf{v}(\mathbf{x}, t)$ ($\mathbf{x} = \mathbf{X}$ at $t = 0$). Chaotic mixing is intimately related to dynamical systems. However, the importance of dynamical-systems tools can be easily overestimated. Suffice it to say that at the moment it is not possible to predict a priori the "degree of chaos" in even simple two-dimensional, time-periodic laboratory experiments without actually doing the experiment first. In fact, there are two important aspects that distinguish chaotic mixing from more conventional applications of dynamical systems and chaos. The first is that we are interested mostly in *rate* processes, i.e. rapid mixing, rather than asymptotic structures and long-time behavior. The second is that the perturbations from integrability are *large*, since this is generally when the best mixing occurs. The current theoretical framework to deal with mixing needs to be extended.

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However, careful experiments, analyses, and computations have revealed some of the fundamental mechanisms operating in simple chaotic flows, and it is now possible to extract rules of general validity that help in the understanding and design of mixing systems. In Section 3, we present prototypical examples that serve as a "window" for more complicated situations. The goal here is not to construct detailed models of specific problems but rather to provide prototypes for a broad class of problems and to generate intuition about what might happen in more complex situations. Even in two dimensions, there is a gradation of difficulties: A mapping is easier to analyze than an analytical description of the velocity field. However, not all problems requiring a computational solution for the velocity field present the same difficulties and potential with respect to analysis, and most possibilities remain relatively unexplored. In some cases it is possible to devise new techniques that allow for an understanding of the system without knowing the exact form of the velocity field. One possible avenue, which is addressed in the last part of Section 3.2, is to focus on the role played by symmetries. In this case, only gross characteristics of the streamfunction $\psi(\mathbf{x}, t)$ are needed. This method is particularly useful in creeping flows, but there are indications that symmetries also play a role in the transition to turbulence.

Undoubtedly, there are many problems in mixing where turbulence plays no role (e.g. mixing in the mantle of the Earth, polymer processing; see Figure 1). Nevertheless, a general attack to the study of mixing has to delve into the connection with turbulence. Some of these matters are discussed in Section 4. In the closing section of this review we try to anticipate the future evolution of chaotic mixing and speculate on the possible connection of these ideas with various problems, such as flow in porous media. We have made an effort to produce a self-contained account. However, space limitations place constraints on the extent to which issues can be addressed (for additional material, see Ottino 1989a).

## 1.2 *Problems of Interest and Quantification of Mixing*

In the simplest case, mechanical mixing of a single fluid, an initially designated material region of fluid (Figure 2*a*) stretches and folds throughout the space (Figure 2*b*). An exact description of mixing is given by the location of the interfaces as a function of space and time. However, this level of description is rare because the velocity fields usually found in mixing processes are complex. Moreover, relatively simple velocity fields can produce exponential area growth due to stretching and folding, and numerical tracking becomes impossible. More realistic problems can take years of computer time with megaflop machines (Franjione & Ottino 1987).

In many problems it is desirable to quantify the mixing achieved (the

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earliest ideas go back to Danckwerts 1952). For example, the objective of a mixing process might be to produce a prescribed degree of "mixedness." A typical criterion might prescribe degrees of resolution and uniformity—for example, by specifying a grid size (resolution) and bounds of composition within each box of the grid (uniformity). Probably the most useful and simple measure of the state of mechanical mixing is given by the *striation thickness* $s = (1/2)(s_A + s_B)$, where $s_A$ and $s_B$ are the thicknesses of the layers of A and B (Ranz 1979, Ottino et al. 1979). The striation thickness is related to the amount of area between the fluids, or the interfacial area per unit volume, $a_v$. This quantity can be interpreted as a structured continuum property; thus, if $S$ designates the intermaterial area within a volume $V$ enclosing the point $\mathbf{x}$ at time $t$, then

$$a_v(\mathbf{x}, t) = \lim_{V \to 0} \frac{S}{V}. \tag{2}$$

If the structure is lamellar, then $a_v = 1/s$. This quantity is useful in describing mixing with diffusion and reaction (Chella & Ottino 1984).

The experiments in Figure 2*c–d* correspond to an experiment similar to that in Figure 2*b*, but in this case the blob is not passive and there is interfacial tension between the blob and the fluid (Tjahjadi 1989). If the fluids are immiscible, at some point in the mixing process the striations do not remain connected and break into smaller fragments. The coupling between the flow field and interfacial tension $\sigma$ occurs at length scales of order $\sigma/\dot{\gamma}\mu$, where $\dot{\gamma}$ is a characteristic shear rate and $\mu$ the viscosity of the continuous fluid. At these length scales the interfaces modify the surrounding flow, making the analysis considerably more complicated. It is obvious that in order to understand the processes of breakup, we have to focus on small scales; this problem is very important in its own right (Acrivos 1983, Rallison 1984, Bentley & Leal 1986, Stone et al. 1986).

Another complication is molecular diffusion. Diffusion is important when the characteristic time for diffusion, $s^2/\mathscr{D}$, is comparable with the characteristic mixing time [for example, diffusion might be unimportant in many problems involving mixing of polymers (Middleman 1977, Tadmor & Gogos 1979, Ottino & Chella 1983) and in some problems in geophysics (McKenzie et al. 1974, Hoffman & McKenzie 1985, Allègre & Turcotte 1986)]. If there is diffusion and reaction, the problem is characterized by at least three characteristic times—diffusion, reaction(s), and stretching rate of the striations; their relative importance is given by two Damköhler numbers (Chella & Ottino 1984). If the fluids are miscible, we can still track material volumes in terms of a (hypothetical) nondiffusive tracer that moves with the mean mass velocity of the fluid or any other

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suitable reference velocity. Designated surfaces of the *tracer remain connected,* and diffusing species traverse them in both directions (Ottino 1982). However, during the mixing process, connected isoconcentration surfaces of *diffusing species might break,* and cuts might reveal islands rather than striations (Gibson 1968).

Does it make sense to settle on a single, or just a few, measure(s) for the quantification of mixing? The answer is no, since the measures are highly dependent upon the problem in question. For example, there is no unique way of quantifying the degree of mixing of structures such as those of Figure 2. The problem becomes even more complex when there are large-scale inhomogeneities, such as the typical mixture of islands and stretched and folded regions characteristic of chaotic mixing (see, for example, Figures 2*e*,*f*). It is also important to distinguish clearly between the *mixing measure* and the *process* producing the mixing. In this context, it is important to remember that (*a*) the measure should be selected according to the specific application, and (*b*) the measurement has to be relatable to the fluid mechanics. The striation thickness is possibly the simplest measure that can be related to the fluid mechanics, but this choice does not work for immiscible fluids. More work in this area seems necessary, especially after recent developments in percolation theory. Choices for measures abound: fractal dimensions for structures such as Figure 2*e*,*f*, and various types of correlations and distribution functions for Figure 2*d*. However, the importance and relevance of such measures seem debatable, since they might be impossible to relate to the mixing process itself and they might be unmeasurable from an experimental view-

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*Figure 2* Chaotic mixing of passive and active (immiscible) fluids in viscous fluids. Figure (*a*) represents typical initial conditions. Figure (*b*) shows a typical snapshot during the mixing of two fluids without interfacial tension in a time-dependent flow between two eccentric cylinders; molecular diffusion is unimportant during the time scale of the experiment, and the striations of the passive yellow tracer remain sharp (Swanson & Ottino 1989). Figures (*c*, *d*) (Tjahjadi 1989) show two stages during the mixing of two fluids with interfacial tension corresponding to the initial conditions shown in (*a*). Figure (*c*) corresponds to an early stage in the stretching-breakup; the thinnest part of a long filament is already broken, whereas the thicker parts show various stages of development of capillary instabilities. Figure (*d*) shows that the black blob, placed in a chaotic region, undergoes considerable stretching and breakup; on the other hand, the red drop, which was initially placed within an island, does not break. Figures (*e*,*f*) (Leong 1989) show two snapshots during the mixing of two different passive substances in a time-periodic cavity flow; both substances undergo chaos, but the two chaotic regions do not mix. Symmetries are clearly evident; in (*e*) the eyes, the nose, and the mouth of a "monster face" are symmetric with respect to the vertical axis (the eyes, the nose, and the mouth contain elliptic-periodic points); in picture (*f*) taken shortly afterward, the face is symmetric with respect to the horizontal axis.

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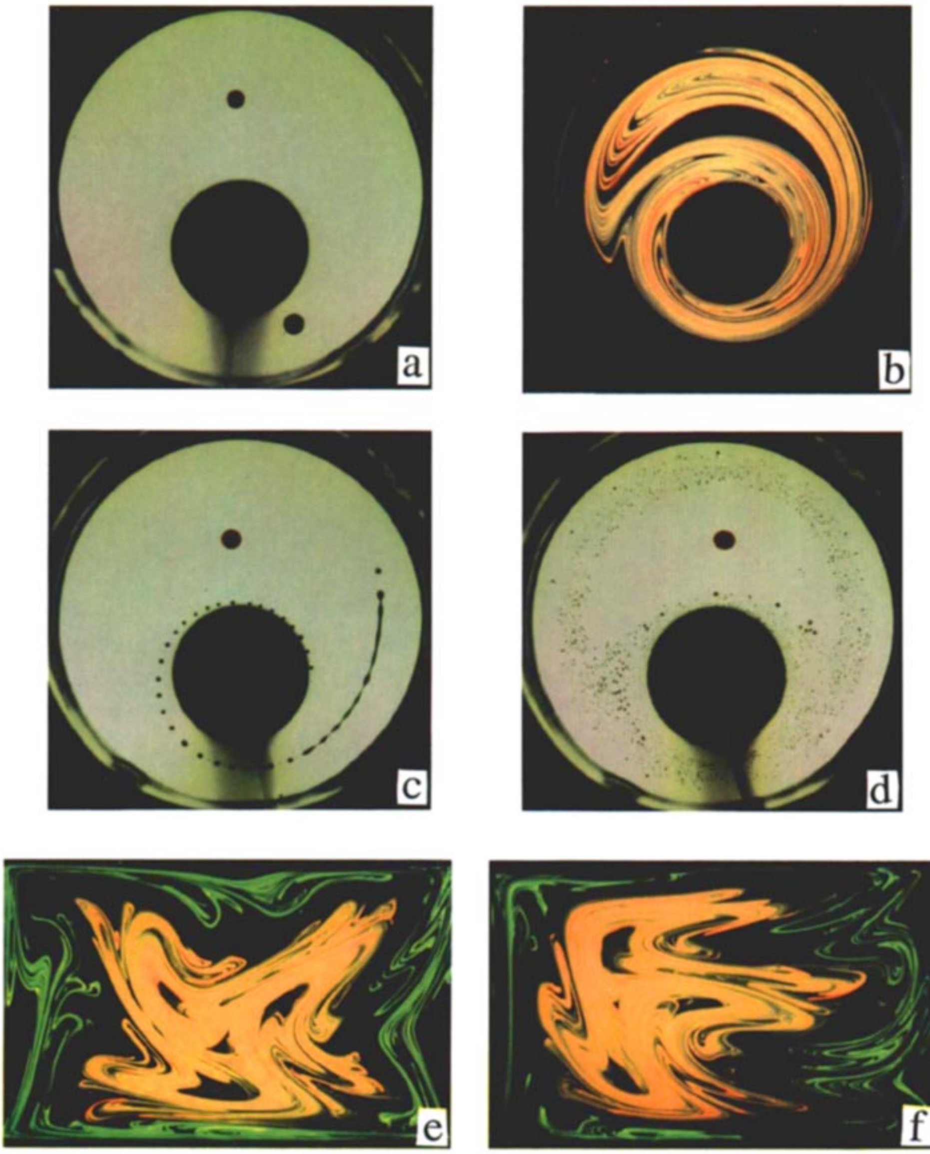

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point [except by *direct* visual observation (for example, electron microscopy) in mixed polymer samples].

## 2. KINEMATICAL FOUNDATIONS AND CHAOTIC DYNAMICS

The mixing problem starts rather than ends with the specification of the velocity field $\mathbf{v}(\mathbf{x}, t)$. The solution of Equation (1), with $\mathbf{x} = \mathbf{X}$ at time $t = 0$, gives the flow or motion,

$$\mathbf{x} = \mathbf{\Phi}_t(\mathbf{X}), \qquad \mathbf{X} = \mathbf{\Phi}_{t=0}(\mathbf{X}), \tag{3}$$

i.e. the particle $\mathbf{X}$ is mapped to the position $\mathbf{x}$ after a time $t$. The motion is required to satisfy $0 < J < \infty$, where $J = \det(\partial x_i/\partial X_j) = \det(\mathbf{F})$. [This mathematical structure is valid whether there is molecular diffusion or not (Bowen 1976).] Since $0 < J < \infty$, two particles, $\mathbf{X}_1$ and $\mathbf{X}_2$, cannot occupy the same position $\mathbf{x}$ at the same time, and one particle cannot split into two, i.e. breakup or coalescence of particles is not allowed. (Obviously there can be drop breakup and coalescence.) Note that already there are two major obstacles to a purely kinematical view of mixing. The first is that in many practical cases the Eulerian velocity $\mathbf{v}(\mathbf{x}, t)$ might be hard to obtain; the second is that even if $\mathbf{v}(\mathbf{x}, t)$ were available, the flow $\mathbf{\Phi}_t(\cdot)$ is almost never obtainable in closed form.

The basic question is the following: What are the conditions under which a deterministic flow $\mathbf{v}(\mathbf{x}, t)$ or its corresponding motion $\mathbf{x} = \mathbf{\Phi}_t(\mathbf{X})$ is able to produce widespread and efficient stretching of a material surface (line in two dimensions) throughout the space occupied by the fluid? What is the best way—assuming that a mixing measure has been adopted—to achieve a desired degree of mixing?

Time has shown that the classical ways of attacking this question are relatively unsuccessful. In fact, standard visualization tools are insufficient and give only a partial insight into the problem. The most popular way to characterize a flow field is in terms of its streamlines. However, very simple streamlines can produce very complicated streaklines (Hama 1962). Considerably more information is provided by particle paths and, particularly, by streaklines (see Section 2.2). In fact, streaklines are very useful in understanding some of the fundamental issues of chaos.

The path of particle $\mathbf{X}$ is given by the solution of $d\mathbf{x}/dt = \mathbf{v}(\mathbf{x}, t)$, with $\mathbf{x} = \mathbf{X}$ at $t = 0$. As we know from differential equations, the solution to this problem is unique and continuous with respect to the initial data if $\mathbf{v}(\mathbf{x})$ has a Lipschitz constant $K > 0$. [Note that the nonautonomous problem can be cast as an autonomous problem by introducing the time as a new

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variable (see Hirsch & Smale 1974).] The distance between two particles $\mathbf{x}_1$ and $\mathbf{x}_2$, initially at $\mathbf{X}_1$ and $\mathbf{X}_2$, evolves according to the inequality

$$|\mathbf{x}_1-\mathbf{x}_2| \leq |\mathbf{X}_1-\mathbf{X}_2|\exp(Kt), \qquad \text{where} \quad K>0. \tag{4}$$

Thus, the "error" in the position $\mathbf{x}_1-\mathbf{x}_2$ is bounded by the initial "error" $\mathbf{X}_1-\mathbf{X}_2$, and the trajectories diverge (at most) exponentially fast. Unfortunately, this result might produce the false impression that all deterministic velocity fields produce simple trajectories, that the error in the final result is of the same order of magnitude as the initial error, and that somehow the only way to uncorrelate initial conditions is to have a stochastic velocity field or a rather complex time and spatial dependence. This need not be so. In fact, many simple systems function with the equal sign in (4), indicating that initial order is lost exponentially fast. However, none of the typical examples presented in fluid-dynamics textbooks regarding streamlines, pathlines, and streaklines is able to accomplish decent mixing. Why is this so? Examples leading to good mixing cannot be integrated (Helleman 1980). In order to understand this point, it is necessary to review a few concepts of kinematics before introducing ideas about dynamical systems and chaotic dynamics.

## 2.1 *Kinematics of Deformation and Mixing Efficiency*

The deformation of a material filament $d\mathbf{X}$ to $d\mathbf{x}$ is given by

$$d\mathbf{x} = \mathbf{F}\cdot d\mathbf{X}. \tag{5}$$

Similarly, the corresponding relation for the stretching of a material area $d\mathbf{A}$ to $d\mathbf{a}$ is given by

$$d\mathbf{a} = (\det \mathbf{F})(\mathbf{F}^{-1})^{\mathrm{T}}\cdot d\mathbf{A}, \tag{6}$$

where $\mathbf{F}$ is the deformation tensor. The corresponding equations for the rates of change are

$$D(d\mathbf{x})/Dt = d\mathbf{x}\cdot\nabla\mathbf{v}, \qquad D(d\mathbf{a})/Dt = d\mathbf{a}\,D(\det\mathbf{F})/Dt - d\mathbf{a}\cdot(\nabla\mathbf{v})^{\mathrm{T}}. \tag{7}$$

The length stretch $\lambda$ and the area stretch $\eta$ are defined as

$$\lambda \equiv \lim_{|d\mathbf{X}|\to 0}\frac{|d\mathbf{x}|}{|d\mathbf{X}|}, \qquad \eta \equiv \lim_{|d\mathbf{A}|\to 0}\frac{|d\mathbf{a}|}{|d\mathbf{A}|}, \tag{8}$$

and can be obtained from

$$\lambda = (\mathbf{C}:\mathbf{MM})^{1/2}, \qquad \eta = (\det\mathbf{F})(\mathbf{C}^{-1}:\mathbf{NN})^{1/2}, \tag{9}$$

where $\mathbf{C}\,(\equiv \mathbf{F}^{\mathrm{T}}\cdot\mathbf{F})$ is the so-called right Cauchy-Green strain tensor (Trues-

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dell & Toupin 1960, Ottino 1989a). The fundamental equations for the rate of stretch are

$$D(\ln \lambda)/Dt = \mathbf{D}:\mathbf{mm}, \qquad D(\ln \eta)/Dt = \nabla\cdot\mathbf{v}-\mathbf{D}:\mathbf{nn}, \tag{10}$$

where $\mathbf{D} \equiv (1/2)[\nabla\mathbf{v}+(\nabla\mathbf{v})^{\mathrm{T}}]$ is the stretching tensor, and $\mathbf{m}$ and $\mathbf{n}$ are the instantaneous orientations ($\mathbf{m} = d\mathbf{x}/|d\mathbf{x}|$, $\mathbf{n} = d\mathbf{a}/|d\mathbf{a}|$). We say that the flow $\mathbf{x} = \mathbf{\Phi}_t(\mathbf{X})$ mixes well in a region $R$ if the time-averaged values of $D(\ln\lambda)/Dt$ and $D(\ln\eta)/Dt$ [e.g. $(1/t)\int(D\ln\lambda/Dt)dt'$] are constant and positive, regardless of the initial orientation $\mathbf{M}\,(\equiv d\mathbf{X}/|d\mathbf{X}|)$, $\mathbf{N}\,(\equiv d\mathbf{A}/|d\mathbf{A}|)$ and placement of the material elements in $R$. The Lagrangian histories $D(\ln\lambda)/Dt = \alpha_\lambda(\mathbf{X},\mathbf{M},t)$ and $D(\ln\eta)/Dt = \alpha_\eta(\mathbf{X},\mathbf{N},t)$, are called *stretching functions*. However, the values of the stretching functions, and their averages, are dependent upon the units of time. A convenient way of comparing flows is in terms of their mixing efficiencies (Chella & Ottino 1985a). The stretching efficiency $e_\lambda = e_\lambda(\mathbf{X},\mathbf{M},t)$ of the materal element $d\mathbf{X}$ and the stretching efficiency $e_\eta = e_\eta(\mathbf{X},\mathbf{N},t)$ of the area element $d\mathbf{A}$ are defined as

$$e_\lambda \equiv (D\ln\lambda/Dt)/(\mathbf{D}:\mathbf{D})^{1/2} \leq 1, \qquad e_\eta \equiv (D\ln\eta/Dt)/(\mathbf{D}:\mathbf{D})^{1/2} \leq 1. \tag{11}$$

If $\nabla\cdot\mathbf{v} = 0$, then $e_i \leq [(n-1)/n]^{1/2}$ $(i = \lambda,\eta)$, where $n$ is the number of dimensions of the flow (Khakhar 1986). The closeness to the upper bound determines the ability of the flow to produce stretching. For purely viscous fluids, the magnitude of $\mathbf{D}$ $[(\mathbf{D}:\mathbf{D})^{1/2}]$ is related to the viscous dissipation. In this case, the efficiency can be thought of as the fraction of the energy dissipated locally that is used to stretch fluid elements. A few examples are probably useful (see Section 3.1): The maximum efficiency of the blinking vortex (Khakhar et al. 1986) is 0.16, the maximum efficiency of a random sequence of simple shear flows is 0.28 (Khakhar & Ottino 1986), and the efficiency of typical mixing equipment is considerably lower (Ottino & Macosko 1980, Ottino 1983).

A complicated stretching function, with a nearly constant time average, is a symptom of "Lagrangian turbulence" (Chaiken et al. 1986, Dombre et al. 1986). [Unfortunately the name might be somewhat misleading; the "Lagrangian" description is due to Euler (see Truesdell 1954, p. 30), and much of "Lagrangian turbulence" is based on Hamiltonian mechanics.] Steady two-dimensional flows with $\nabla\cdot\mathbf{v} = 0$ cannot produce Lagrangian turbulence; the stretching function decays as $1/t$, and the efficiency decays to zero. This can be seen in various ways. A steady isochoric two-dimensional flow is characterized by the streamfunction $\psi(x,y)$. In rectangular coordinates, the velocity field can be obtained as $\mathbf{v} = \nabla\times(\psi\mathbf{e}_z)$. Level curves $\psi(x,y,t = \text{fixed})$ give the instantaneous picture of the streamlines, which in this case coincide with the pathlines and streaklines. If the flow is bounded, the flow can be divided into regions of closed streamlines. The

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stretching within each region is poor. Let $T(\psi)$ denote the period in the streamline $\psi$. It is then possible to show that $d\mathbf{x}(t)$ is mapped into $d\mathbf{x}(t+T)$ at time $t+T$:

$$d\mathbf{x}(t+T) = d\mathbf{x}(t)\cdot[\mathbf{1}-(dT/d\psi)(\nabla\psi)\mathbf{v}]+\text{higher order terms in } d\mathbf{x}. \quad (12)$$

Similarly, the orientation of the filament after $n$ cycles of the flow is given by

$$\mathbf{m}_{t+T} = \mathbf{m}_0\cdot[\mathbf{1}-(dT/d\psi)(\nabla\psi)\mathbf{v}]^n/\lambda, \quad (13)$$

where $\mathbf{m}_0$ is the initial orientation. As the number of cycles goes to infinity, the filament becomes aligned with the streamlines and the stretching $\lambda$ becomes linear with time (Franjione 1989). A similar result occurs for duct flows, i.e. flows belonging to the class

$$v_x = \partial\psi/\partial y, \qquad v_y = -\partial\psi/\partial x, \qquad v_z = f(x,y). \quad (14)$$

In this case the stretching goes at most as $t^2$ (Franjione 1989).

A hyperbolic flow, $v_1 = \varepsilon x_1$, $v_2 = -\varepsilon x_2$, can produce exponential stretching (except if the elements coincide with the $x_2$-axis). However, this flow is also unbounded, which makes it less interesting from a practical viewpoint. What, then, are the possibilities to create efficient stretching in a bounded two-dimensional flow? Some reorientation mechanism is necessary to create efficient mixing in a two-dimensional bounded flow to avoid efficiency decay as $1/t$. Reorientation can be achieved by means of folding. One possibility is to compose cleverly designed motions

$$\mathbf{\Phi}_{t+r}(\mathbf{X}) = \mathbf{\Phi}_t(\mathbf{\Phi}_r(\mathbf{X})), \quad (15)$$

as is done, for example, in a static mixer or an eggbeater, to produce periodic folding (see Ottino 1989b, p. 64). However, is there any way of producing this naturally in a flow without recourse to artificiality? Chaotic flows accomplish reorientation and bending of material elements in a natural way. However, the understanding of these kinds of system requires the incorporation of a different set of tools to the arsenal of fluid kinematics. The most transparent case corresponds to two-dimensional time-periodic flows, and the following analysis is largely restricted to this case. The place to start is with the location and character of fixed and periodic points in the flow. Some of the terminology is similar to that of fluid mechanics (e.g. hyperbolic and elliptic points). However, the reader is urged to be careful; the analysis is based on the *flow* (i.e. the integral of the velocity field) and not on the velocity field itself (see, for example, the analysis in Khakhar et al. 1986).

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## 2.2 *Chaos in Area-Preserving Flows*

Given a flow $\mathbf{x} = \mathbf{\Phi}_t(\mathbf{X})$, $\mathbf{P}$ is a *fixed point* of the flow if

$$\mathbf{P} = \mathbf{\Phi}_t(\mathbf{P}) \tag{16}$$

for all time $t$ (i.e. the particle located at the position $\mathbf{P}$ stays at $\mathbf{P}$). For example, the origin in the flow $x_1 = X_1 \exp(\varepsilon t)$, $x_2 = X_2 \exp(-\varepsilon t)$, corresponding to the velocity field $v_1 = \varepsilon x_1$, $v_2 = -\varepsilon x_2$, is a fixed point. On the other hand, the point $\mathbf{P}$ is periodic, of period $T$, if

$$\mathbf{P} = \mathbf{\Phi}_{nT}(\mathbf{P}) \tag{17}$$

for $n = 1, 2, 3, \ldots$ but not for any $t < T$. That is, the material particle that happened to be at the position $\mathbf{P}$ at time $t = 0$ will be located in exactly the same spatial position after a time $nT$ [it could be anywhere for $nT < t < (n+1)T$]. For example, all the points in a circular streamline in Couette flow are periodic. Similar definitions apply to a period-$p$ point (for example, a period-2 point returns to $\mathbf{P}$ for $n = 2, 4, 6, \ldots$). Note that the concept of periodicity depends on the frame of reference. Thus, for example, there are periodic points in a moving frame in the cat's-eye portrait in a shear flow, but there are none in a fixed frame. The periodic points can be classified as hyperbolic, elliptic, or parabolic, according to the deformation of the fluid in the neighborhood of the periodic point (the parabolic case being degenerate). The character of the flow in the neighborhood of the periodic point is given by the eigenvalues of the linearized mapping:

$$D\mathbf{\Phi}_T(\mathbf{P}) \cdot \xi_k = \lambda_k \xi_k, \tag{18}$$

where $D$ denotes the matrix $\partial(\cdot)_i/\partial X_j$. According to the value of the eigenvalues $\lambda_k$, the point $\mathbf{P}$ is called hyperbolic, elliptic, or parabolic:

Hyperbolic $|\lambda_1| > 1 > |\lambda_2|, \qquad \lambda_1\lambda_2 = 1,$

Elliptic $|\lambda_k| = 1 \; (k = 1, 2) \quad \text{but} \quad \lambda_k \neq 1,$

Parabolic $\lambda_k = \pm 1 \; (k = 1, 2).$

The net motion in the neighborhood of a elliptic point is rotation; the motion in the neighborhood of a hyperbolic point is contraction in one direction and stretching in another. It is important to stress that this can occur in the absence of hyperbolic points in the streamline portrait of the velocity field; for example, in the blinking-vortex flow (see Section 3.1) the velocity field consists of just a sequence of two circular motions about two different centers.

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Hyperbolic points have associated invariant regions of inflow and outflow called the stable [$W^s(\mathbf{P})$] and unstable [$W^u(\mathbf{P})$] manifolds:

$$W^s(\mathbf{P}) = \{\text{all } \mathbf{X} \in \mathbb{R}^2 \text{ s.t. } \mathbf{\Phi}_t(\mathbf{X}) \to \mathbf{P} \quad \text{as} \quad t \to \infty\},$$

$$W^u(\mathbf{P}) = \{\text{all } \mathbf{X} \in \mathbb{R}^2 \text{ s.t. } \mathbf{\Phi}_t(\mathbf{X}) \to \mathbf{P} \quad \text{as} \quad t \to -\infty\}.$$

Fluid particles leave the neighborhood of **P** through $W^u(\mathbf{P})$ and get back to **P** via $W^s(\mathbf{P})$. Physically, the unstable manifold corresponds to a streakline injected at the periodic point. [The injection apparatus follows the motion of the point (see Figure 3).] In two-dimensional time-periodic flows the manifolds are typically represented as lines (in Poincaré sections); in three-dimensional steady flows the sets can be surfaces (see Abraham & Shaw 1985, Ottino 1989a). By definition, the sets $W^s(\mathbf{P})$ and $W^u(\mathbf{P})$ are invariant; a particle belonging to one of the sets does so permanently and cannot escape from it. Moreover, these sets cannot abruptly end in the interior of the fluid (very much like a vortex line). What, then, are the possibilities? One possibility is that somehow the outflow $W^u(\mathbf{P})$ joins smoothly into the inflow $W^s(\mathbf{P})$; in this case nothing interesting happens. This is precisely what occurs in a steady two-dimensional flow (see Figure 3*a*).

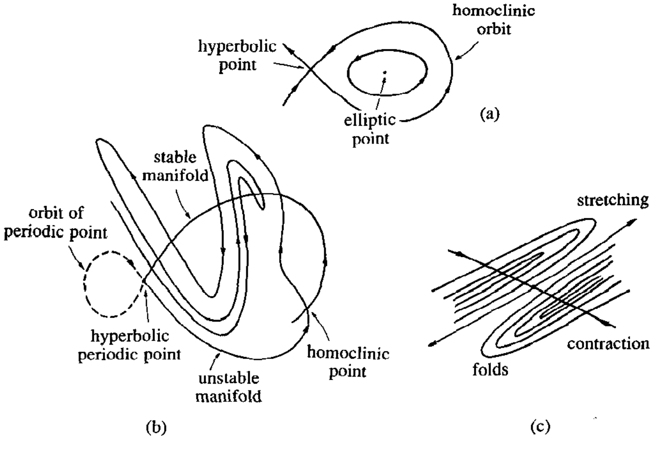


*Figure 3* (*a*) Typical portrait of an integrable system showing a homoclinic orbit; the outflow of a fixed hyperbolic point joins smoothly with the inflow. In (*b*) the structure has been perturbed; the hyperbolic point is periodic and moves in a closed orbit, and its stable and unstable manifolds cross at an angle forming a transverse homoclinic point. Since one homoclinic point implies infinitely many, the manifolds intersect again, but the distance between successive crossings diminishes as the unstable manifold is pushed by the stable manifold approaching the hyperbolic point. Figure (*c*) shows the typical structure produced by a passive tracer in the neighborhood of a hyperbolic point; such structure is evident in many experimental studies.

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However, something major happens if the intersection is transversal, i.e. the manifolds intersect nontangentially. This point is highly nontrivial, and almost everything else rests on it. The understanding of transversal intersection of manifolds requires thinking in the space $x_1$, $x_2$, $t$. In a time-periodic system, the time axis goes around the torus, and the plane $x_1$–$x_2$ ($t = nT$, $n = 1, 2, 3, \ldots$) corresponds to the cross section of a torus.

A point belonging simultaneously to both the stable and unstable manifolds of two different fixed (or periodic) points **P** and **Q** is called a *transverse heteroclinic point*. If $\mathbf{P} = \mathbf{Q}$, the point is called homoclinic. One intersection implies infinitely many and sensitivity to initial conditions (Guckenheimer & Holmes 1983). This is one of the fingerprints of chaos. [The reader might try to reconcile the fact that a point belongs to both manifolds for all times with the situation displayed in Figure 3*b*, which shows the point "jumping" from intersection to intersection. How does the jump occur?]

One of the manifestations of chaos most readily related to fluid mixing is the exponential divergence of initial conditions. The rate of divergence of a filament $d\mathbf{X}$ placed at $\mathbf{X}$ with initial orientation $\mathbf{M}_i$ is measured by means of Liapunov exponents:

$$\sigma_i(\mathbf{X}, \mathbf{M}_i) \equiv \lim_{\substack{t\to\infty \\ |d\mathbf{X}|\to 0}} \left\{\frac{1}{t}\ln\left(\frac{|d\mathbf{x}|}{|d\mathbf{X}|}\right)\right\}. \tag{19}$$

A two-dimensional area-preserving flow has two Liapunov exponents $\sigma_1$, $\sigma_2$ such that $\sigma_1+\sigma_2 = 0$; almost all $\mathbf{M}_i$'s yield $\sigma_1$ ($>0$). The Liapunov exponent is the long-time average of the specific rate of stretching, $D\ln\lambda/Dt$:

$$\sigma_i(\mathbf{X}, \mathbf{M}_i) \equiv \lim_{t\to\infty}\left\{\frac{1}{t}\int_0^t\left(\frac{D\ln\lambda}{Dt}\right)dt'\right\} = \lim_{t\to\infty}\left\{\frac{1}{t}\ln\lambda(\mathbf{X}, \mathbf{M}_i, t)\right\}. \tag{20}$$

Similarly, the average stretching efficiency can be interpreted as a normalized Liapunov exponent [with respect to $(\mathbf{D}:\mathbf{D})^{1/2}$]. However, the relationship between the (maximum) Liapunov exponent and the efficiency is not direct unless $(\mathbf{D}:\mathbf{D})^{1/2}$ is constant over the pathlines. In most cases of interest, $(\mathbf{D}:\mathbf{D})^{1/2}$ is a function of both $\mathbf{X}$ and $t$. All steady two-dimensional flows have zero Liapunov exponents. Thus, the condition for the mixing to be effective is that the flow be chaotic. However, a chaotic label does not guarantee "good mixing"; in particular, the chaos might be confined to a very small region.

How can we produce such flows and generate good mixing? Two questions come to mind: (*a*) What can we do to a steady flow in order to generate transverse homoclinic/heteroclinic intersections? (*b*) What kind

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of characteristics does a time-periodic flow have to possess in order to produce homoclinic or heteroclinic intersections?

Early attempts to find chaos in fluid flows were based on the idea of perturbing a steady streamline portrait with hyperbolic and elliptic fixed points [question (*a*)]. A two-dimensional fluid flow

$$(dx/dt)_{X,Y} = \partial\psi/\partial y, \qquad (dy/dt)_{X,Y} = -\partial\psi/\partial x, \tag{21}$$

is equivalent to a Hamiltonian system ($x \to p$, $y \to q$, $\psi \to H$; Aref 1984), and time-periodic streamfunctions $\psi$ can produce transverse intersections of manifolds ($x = X$, $y = Y$ at $t = 0$).

Chaotic mixing is a purely kinematical phenomenon and can occur in slow or fast flows. Indeed, careful experiments (Chaiken et al. 1986, Ottino et al. 1988, Leong & Ottino 1989) present incontrovertible evidence that chaos is possible even in creeping flows. [Part of the earlier belief in "kinematical reversibility" of Stokes flows in general stems from the fact that previous experiments (Heller 1960, Hiby 1962, Taylor 1972) focused exclusively on integrable flows; these flows are indeed "reversible."] This connection with Hamiltonian mechanics generated studies in the blinking-vortex system (Aref 1984), the journal-bearing flow (Aref & Balachandar 1986), and the Taylor-Couette flow (Broomhead & Ryrie 1988), as well as studies of chaos in systems consisting of a pair of vortices (Leonard et al. 1987) or roll cells (Knobloch & Weiss 1987, Weiss 1988, Weiss & Knobloch 1989) modulated by time-periodic extensional flows or waves. Various theorems and techniques describe what happens when the perturbations are small (see, for example, Guckenheimer & Holmes 1983, Wiggins 1988). However, the initial expectations regarding the usefulness of available Hamiltonian theory, e.g. KAM (Kolmogorov-Arnold-Moser) theory, have been scaled down, although some special techniques, such as the Melnikov method (Guckenheimer & Holmes 1983, Wiggins 1988) have been useful in the analysis of analytical systems with small perturbations from integrability (Holmes 1984, Leonard et al. 1987, Broomhead & Ryrie 1988, Rom-Kedar et al. 1989). In some of the most effective mixing flows (e.g. a discontinuously operated cavity flow or an eggbeater) there is no integrable picture to speak of; in many others, perturbations are greater than order one (e.g. journal-bearing flow). In fact, when viewed from the perspective of large perturbations, it makes more sense to ask question (*b*) above. From this viewpoint, the most visual construction indicative of chaos is the Smale horseshoe map (Smale 1967). For the flow to produce a horseshoe map it must be capable of stretching and folding a region of fluid and returning it—stretched and folded—to its initial location [satisfying a set of conditions known as Moser's conditions (Moser 1973); for applications, see Chien et al. (1986) and Ottino (1989a)]. A necessary condition

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is that streamline portraits at two successive times (or axial distances) show crossing of streamlines. It is important to stress that the instantaneous streamline portraits need not have any saddle points in order to produce chaos. Creative designs can be based on this idea.

## 3. EXAMPLES

It is well established, by computation and experiments, that two-dimensional time-periodic flows display chaotic behavior; the number of three-dimensional studies is less complete. A partial list of the flows studied to date is given in Figure 4. (Others are given below.) Most flows studied to date are kinematically defined, i.e. the velocity field is either steady or time periodic. In other words, there is never a question as to what the velocity at a fixed point will do at any given time. The time signal of the Eulerian velocity field is generally time periodic.

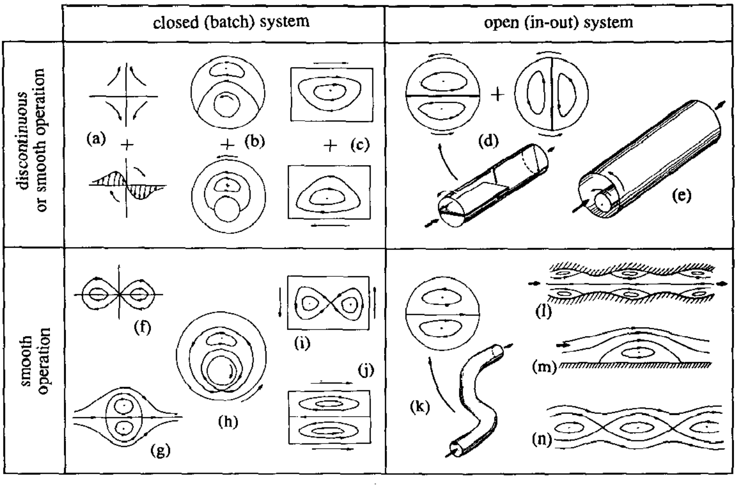


*Figure 4* Typical systems producing chaotic mixing. The systems (*a–e*) admit continuous (i.e. smooth) or discontinuous operation. The left column corresponds to closed, or "batch," systems, the right column to open, or "continuous," flows (i.e. the material enters and leaves the system). Flow types as follows: (*a*) tendril-whorl flow, (*b*) journal-bearing flow, (*c*) cavity flow, (*d*) partitioned-pipe mixer flow, (*e*) eccentric helical annular mixer flow; figure (*f*) shows the blinking-vortex flow with the two vortices "on." (This system also admits discontinuous operation.) Figure (*g*) shows the oscillating vortex-pair flow; this flow is perturbed by a hyperbolic time-dependent flow such as in the top part of (*a*). Figure (*h*) shows a typical streamline portrait of the journal-bearing flow, whereas (*i,j*) show possible streamline portraits with the cavity flow. Figure (*k*) shows a typical streamline pattern in a twisted pipe [compare with (*d*)], figure (*l*) a typical streamline pattern in a channel with wavy walls, figure (*m*) a separation bubble, and figure (*n*) the cat's-eye flow in a moving frame.

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### 3.1 *Maps*

The most primitive systems studied from the viewpoint of mixing are mappings. Maps admit detailed analytical treatment; they are related to area-preserving transformations, a classical topic in mathematics with an extensive literature (Birkhoff 1920). In principle, any time-periodic flow can be reduced to a mapping. However, obtaining the explicit expression of the mapping can be very difficult. The simplest maps, from the viewpoint of analysis, are the *tendril-whorl flow* (TW), which is a succession of elongational and rotational flows, and the eggbeater flow (J. G. Franjione & J. M. Ottino, unpublished). For example, in the TW flow it is possible to do a considerable amount of analytical work. The next simplest flow is the *blinking-vortex flow* (BV), due to Aref (1984). This was the first two-dimensional, time-periodic flow studied in the context of chaos and Hamiltonian mechanics.

THE TW FLOW The tendril-whorl flow (TW), introduced by Khakhar et al. (1986), is a discontinuous succession of extensional flows and twist maps. The physical motivation for this flow is that, locally, a velocity field can be decomposed into extension and rotation. Thus, the flow consists of vortices producing whorls that are periodically squeezed by the hyperbolic flow, leading to the formation of tendrils, and so on. The velocity field over a single period is given by

$$v_x = -\varepsilon x, \qquad v_y = \varepsilon y \quad \text{for} \quad 0 < t < T_{\text{ext}}, \qquad \text{(extensional part)}$$

$$v_r = 0, \qquad v_\theta = -\omega(r) \quad \text{for} \quad T_{\text{ext}} < t < T_{\text{ext}} + T_{\text{rot}}, \qquad \text{(rotational part)}$$

where $T_{\text{ext}}$ denotes the duration of the extensional component, $T_{\text{rot}}$ the duration of the rotational component, and $\omega(r)$ is of the form $r^2 \exp(-r)$. By examining the streamlines of the flows, it is possible to prove that the TW system is capable of producing horseshoe maps of period-1 (see Ottino 1989a).

THE BV FLOW The blinking-vortex flow (Aref 1984, Khakhar et al. 1986) consists of two corotating point vortices, separated by a fixed distance $2a$, that blink on and off periodically with a constant period $T$. At any given time, only one of the vortices is on, so that the motion is made up of consecutive twist maps about different centers. The velocity field with respect to each vortex is

$$v_r = 0, \qquad v_\theta = \Gamma/2\pi r, \tag{22}$$

where $\Gamma$ is the strength of the vortex. The system is controlled by a single parameter $\mu = \Gamma T/2\pi a^2$. When the two vortices act simultaneously, the system has the typical appearance of an integrable system (a central hyper-

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bolic point and two elliptic points). In this case we might imagine that perturbations on the integrable system are introduced by varying the period of the flow (i.e. starting from the integrable case $\mu = 0$ by increasing the value of $\mu$). In a similar fashion as with the TW mapping it is possible to prove, by construction, the existence of period-1 horseshoe functions in the flow (Ottino 1989a). The average efficiency of this flow seems to level off at 0.07 beyond $\mu \approx 3$, and the calculations indicate that it remains almost constant up to $\mu = 15$. This behavior seems to be typical of many flows, and simple models, such as shear flow with random reorientation, produce similar results (Khakhar & Ottino 1986).

Both the tendril-whorl flow and the blinking-vortex flow admit several generalizations; some are trivial, but others might reveal new physics [for example, a flow consisting of sources and/or sinks (Jones & Aref 1988) is rather contrived but illustrates that flows without circulation can produce chaotic mixing]. An obvious generalization is to operate the systems in a smooth rather than discontinuous way. (For example, the strength of the vortices can be time-periodic functions of time.) This does not seem to make a tremendous difference. Some results are relatively independent of the corners in particle trajectories produced by the discontinuous operation. In fact, just by looking at computational results (for example, a Poincaré section) it is nearly impossible to decide whether the flow producing the result was continuous or discontinuous. [The result holds for other systems as well—for example, the journal-bearing flow (see Ottino 1989a).] Other possibilities are relatively unexplored. One is to make the streamlines elliptical rather than circular. The crossing of streamlines is obviously related to chaos; thus, if $\psi_n(\mathbf{x})$ denotes the streamline portrait at $n$ and $\psi_{n+1}(\mathbf{x})$ at $n+1$, the degree of crossing is given by a color-coded plot of $\nabla\psi_n(\mathbf{x}) \cdot \nabla\psi_{n+1}(\mathbf{x})$, with $|\nabla\psi| = 1$. What is the best configuration to produce the fastest mixing? Clearly, the streamlines at $n$ and $n+1$ are insufficient to quantify the mixing, and the speed along the streamlines also matters. A plot of $\mathbf{v}_n(\mathbf{x}) \times \mathbf{v}_{n+1}(\mathbf{x})$ might be revealing. Simple diagnostics are needed to screen candidate flows.

## 3.2 *Experiments*

What information is provided by experiments that is not readily available in computations? To start with, experimental observations provide smoothness typically unavailable in standard computations, especially in chaotic regions (Ottino et al. 1988). Experiments also provide a wealth of information regarding a variety of structures, especially those with macroscopic spatial extent and low periods. The task is to identify the structures responsible for mixing in an actual fluid experiment in the absence of an analytical or computational description of the flow.

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CAVITY FLOW The *cavity flow* consists of a rectangular region capable of producing a two-dimensional velocity field in the $x$–$y$ plane. The flow region is rectangular with width $W$ and height $H$, and two opposing walls can be moved in a steady or time-dependent manner. [Other configurations are possible (see Chien et al. 1986).] This system has been thoroughly investigated: The objects that admit experimental investigation are periodic points; bifurcations, birth, and collapse of poorly mixed regions (islands); coherent structures; and symmetries. All these terms can be defined experimentally (Leong & Ottino 1989).

How can a periodic point be labeled as either hyperbolic or elliptic in an actual experiment? The first step is to show that the point is indeed periodic. Then, the order of the point is determined by the number of periods it takes the point to return to the same location. Possibly the most important by-product of the experiments, which was missed by numerous earlier computations, has been the revelation of the amazing degree of organization, coherence, and periodicity present in chaotic mixing (see Figures 2*e*,*f* herein and the figures on pp. 60–61 in Ottino 1989b). Elliptic points are surrounded by *islands*, invariant regions that translate, stretch, and contract periodically and undergo a net rotation, preserving their identity. Islands do not exchange matter with the rest of the fluid and therefore represent an obstacle to efficient mixing (Figure 5). This fact serves to locate periodic points. Carefully located blobs mark either the interior or the exterior of islands. Experimentally, if the point is elliptic, it appears as a hole or an island (not dye filled) unless the dye was located in the neighborhood of the point at the very beginning of the experiment, in which case the dye will not escape from it. Typically, blobs located outside islands quickly demarcate the boundary. Blobs placed in the interior of islands evolve very slowly. We have observed that the flow within islands is mostly rotational, that the stretching is linear, and that the rates of rotation are usually much slower than in the rest of the flow. Whorls are rare (see Figure 5).

To the extent that the system operates under creeping flow, the rate is of no consequence and only the displacement per period is important. The typical displacement per period—a sequence of top and bottom motions—is of order 1–10 cavity-width units. It is interesting to note that the maximum efficiency in a random sequence of shear flows is obtained when

*Figure 5* The stretching and folding mechanism characteristic of chaotic mixing is clearly demonstrated by this experiment, which shows the time evolution of two passive blobs in a time-periodic cavity flow. The top figure shows the initial conditions. After a few periods, one of the blobs undergoes significant stretching, whereas the other remains trapped in a regular island (Leong 1989).

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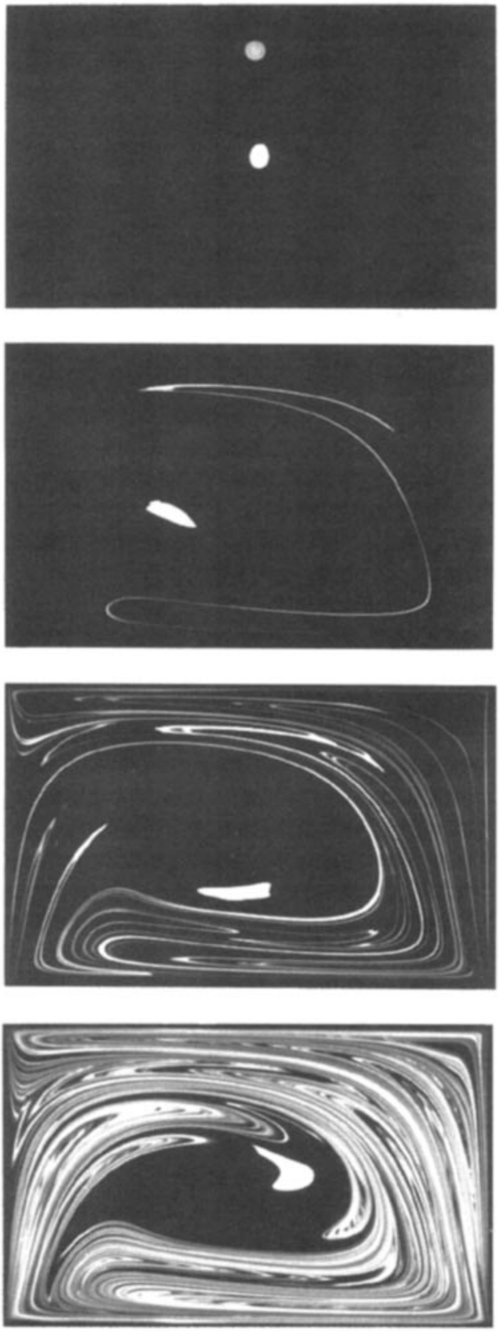

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the average strain is about five (Khakhar & Ottino 1986). The best mixing in the cavity flow occurs when the displacement of the top or bottom wall is about seven times the width of the cavity (Leong & Ottino 1989). This seems to be a good rule of thumb for effective mixing. Continuous and discontinuous flows can be compared on the basis of equal displacement. However, the most revealing comparison is on the basis of symmetry. In fact, symmetries seem to be one of the most powerful tools in the understanding of mixing (Franjione et al. 1989; see Figures 2*e*,*f*).

JOURNAL-BEARING FLOW The flow between two eccentric cylinders was the first flow studied that is amenable to both computational and experimental investigation. The solution corresponding to creeping flow has been thoroughly studied [Jeffery 1922, Wannier 1950, Ballal & Rivlin 1976; in order to achieve this condition in the laboratory, both the Strouhal number and the Reynolds numbers must be small]. Since the problem is linear, the streamfunction $\psi(\mathbf{x}, t)$ can be written as a linear combination of the forcings of the contributions corresponding to the inner and outer cylinders, i.e. $\psi(\mathbf{x}, t) = \psi_{\text{in}}(\mathbf{x})\Omega_{\text{in}}(t) + \psi_{\text{out}}(\mathbf{x})\Omega_{\text{out}}(t)$, where $\Omega_{\text{in}}(t)$ and $\Omega_{\text{out}}(t)$ are the speeds of the inner and outer cylinders, respectively (Aref & Balachandar 1986, Chaiken et al. 1987). According to the operating conditions, the flow might display one or two saddle points, and time-periodic operation might give rise to homoclinic and heteroclinic trajectories.

It is important to note two observations. The first is that under creeping flow the instantaneous streamline portrait is independent of the actual speed of the boundary and depends only on the ratio $\Omega_{\text{in}}(t)/\Omega_{\text{out}}(t)$. Also, as long as the velocity histories do not overlap [i.e. $\Omega_{\text{in}}(t) = 0$ whenever $\Omega_{\text{out}}(t) \neq 0$, and vice versa] the results for different histories, $\Omega_{\text{in}}(t)$ and $\Omega_{\text{out}}(t)$, are identical provided that the angular *displacements*

$$\theta_{\text{in}} = \int \Omega_{\text{in}}(t)\, dt \quad \text{and} \quad \theta_{\text{out}} = \int \Omega_{\text{out}}(t)\, dt \tag{23}$$

are kept the same. [In order to produce fast, widespread chaos with counterrotating cylinders, the angular displacements are of the order of several revolutions; excellent mixing can be obtained in just four periods with six revolutions of the inner cylinder and two of the outer cylinder (Swanson & Ottino 1989).] The second observation is that in order to get a good understanding of chaotic systems in general, it is convenient to start operation in such a way as to produce symmetric Poincaré sections. An important consequence of symmetry is that the search for periodic points is one dimensional rather than two dimensional. Once periodic points are located, it is possible to study the manifolds associated with the low-order points and their intersections.

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There is a great need to develop diagnostics and tools for the analysis of mixing. Traditional studies in dynamical systems rely heavily on Poincaré sections. However, in the context of mixing, Poincaré sections do not convey a sense of the mixing structure present within chaotic regions and give no indication of mixing rate. Indeed, a naive interpretation of results might lead to erroneous conclusions. For example, breakup of KAM curves within large islands (formation of islands within islands) might suggest the presence of whorls within whorls. However, in most cases the motion within the islands does not produce significant stretching, and instead it is nearly a solid-body rotation. The rate of rotation in the smaller islands is even slower, and for all practical purposes they can be ignored as compared with the stretch in the chaotic regions. [Breakup in regular regions is very poor (see Figure 2*d*).]

The rate of spreading of a passive tracer is controlled by the unstable manifolds of the hyperbolic points. In general the spreading is controlled by the manifolds associated with the lowest order periodic points, and it is roughly proportional to the value of the eigenvalues and is inversely proportional to the period of the point. A careful analysis of the mapping of the region between stable and unstable manifolds reveals transport mechanisms within the chaotic region (Rom-Kedar et al. 1989). Extension of these studies might be used to compute the character of dispersion laws (e.g. anomalous "diffusion") as well as various types of exit-time functions.

The agreement between computations and experiments with small degrees of stretching is excellent. Chaiken et al. (1986) made direct comparisons for stretching of the order 10 or so, but not surprisingly they were unable to make comparisons for well-mixed systems (Franjione & Ottino 1987). The most successful comparison to date between experiments and computations for well-mixed systems is based on mappings of lineal stretch (Swanson & Ottino 1989). The *average stretching* is computed by averaging the stretchings over all initial orientations. The stretching is then referred back to the initial placement, and the distribution of stretching over the flow region is divided into regions of high and low stretch. (The subdivision depends on the threshold, but it is relatively insensitive to it.) Figure 6 shows the remarkable agreement between such computations and experiments; the agreement is good even after intervals as short as two periods or so. [Extensive comparisons are presented by Swanson & Ottino (1989).]

BEST MIXING AND FRACTAL SPECTRUM Islands are the most important obstruction to mixing. However, theory to date is insufficient to predict accurately their location and evolution. Let us suppose that we have an initial condition to be mixed. For simplicity, assume that we restrict the mixing to a cavity flow with discontinuous top and bottom wall motions.

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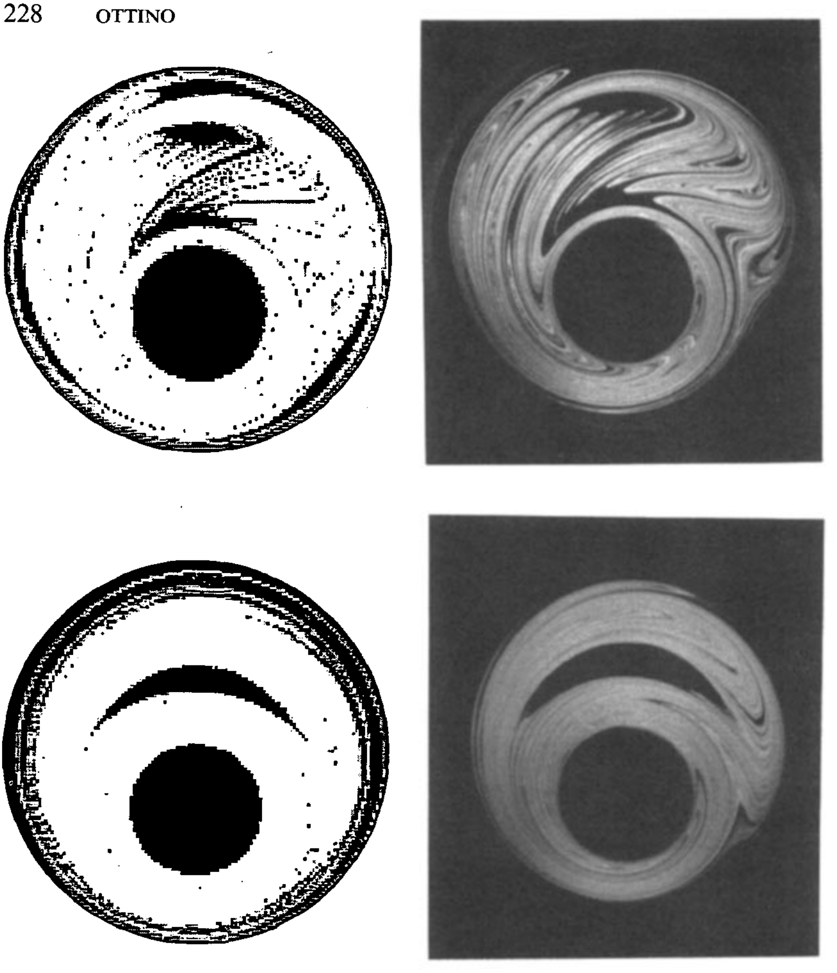

*Figure 6* Comparison between experiments (right column) and computations (left column) in the journal-bearing flow. The computations are a mapping of linear stretch; the white regions undergo considerable stretching, whereas the black regions are hardly stretched at all. The rotation of the outer cylinder per period is 270° in the top figure and 360° in the bottom figure. (The inner cylinder rotates in the opposite direction for three times the amount of the outer cylinder.) Note that the pictures are nearly symmetric with respect to the vertical axis (Swanson & Ottino 1989).

The objective is to accomplish the best possible mixing using some allotted amount of displacement that we can use any way we wish. It is obvious that chaos is desired and that we can accomplish good mixing by using a time-periodic sequence of top and bottom motions. (For simplicity, let us

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restrict the sequences to equal displacements and walls moving in opposite directions.) However, if we select a "bad" displacement per period, large islands will form (as shown in Figure 5). The first question, then, is how quickly can we recognize that the flow contains an island (again see Figure 5); the second question is how to destroy the island. However, once an island is broken, other islands will form, and these have to be destroyed too. (An island being broken is shown in Figure 6.) Can islands be systematically destroyed? The answer is yes (Franjione et al. 1989). One possibility is to exploit the symmetries of the system. Chaos coexists with symmetries, and symmetries can be discovered if systems are examined at suitable times (see, for example, Figures 2*e*,*f* corresponding to the cavity flow, as well as the comments regarding symmetric Poincaré sections in the journal-bearing flow and the partitioned-pipe mixer flow). As soon as a time-periodic sequence is chosen, islands are restricted to lie on axes of symmetries or in pairs about an axis. A periodic sequence might correspond to a series of top and bottom motions TBTBTB. A sequence leading to symmetry destruction starts with TB and is then followed by BT; the sequence TBBT is then followed by BTTB, and so on. This continuously shifts the position of possible islands and does not allow them to form in the flow. The sequence is thus . . . TBBTBTTBBTTBTBBT (the sequence is read from right to left). In a time-periodic system, a fixed probe records a time-periodic velocity signal and the power spectrum reveals only one peak. By contrast, the power spectrum in this system has a fractal structure (Mandelbrot 1982). This is probably the simplest system leading to a time-dependent Eulerian velocity signal (Figure 7).

## 3.3 *Three-Dimensional Flows and Open Flows*

THE ABC FLOW Intuition built on two-dimensional flows might be somewhat misleading in the understanding of three-dimensional flows and open flows in general. (By "open" we mean that material enters and leaves the system; the other possible word is "continuous," but it can also be misunderstood.) Chaos in three-dimensional flows is possible even if the flows are steady. In fact, the very first example of chaotic advection was a steady Beltrami flow [now called the ABC flow for Arnold-Beltrami-Childress (Dombre et al. 1986)]. In a Beltrami flow, particles spin in the direction of their motion, i.e. $\boldsymbol{\omega} = \beta(\mathbf{x})\mathbf{v}$. Since $\nabla \cdot \boldsymbol{\omega} = 0$ and $\nabla \cdot \mathbf{v} = 0$, it follows that $\mathbf{v} \cdot \nabla \beta = 0$ and that the streamlines belong to surfaces $\beta(\mathbf{x}) =$ constant, which act as a constant of the motion. However, if $\beta$ is uniform, the constraint disappears. Arnold (1965) conjectured that such flows might have a complex topology, and Hénon, in a short note (Hénon 1966), examined the case

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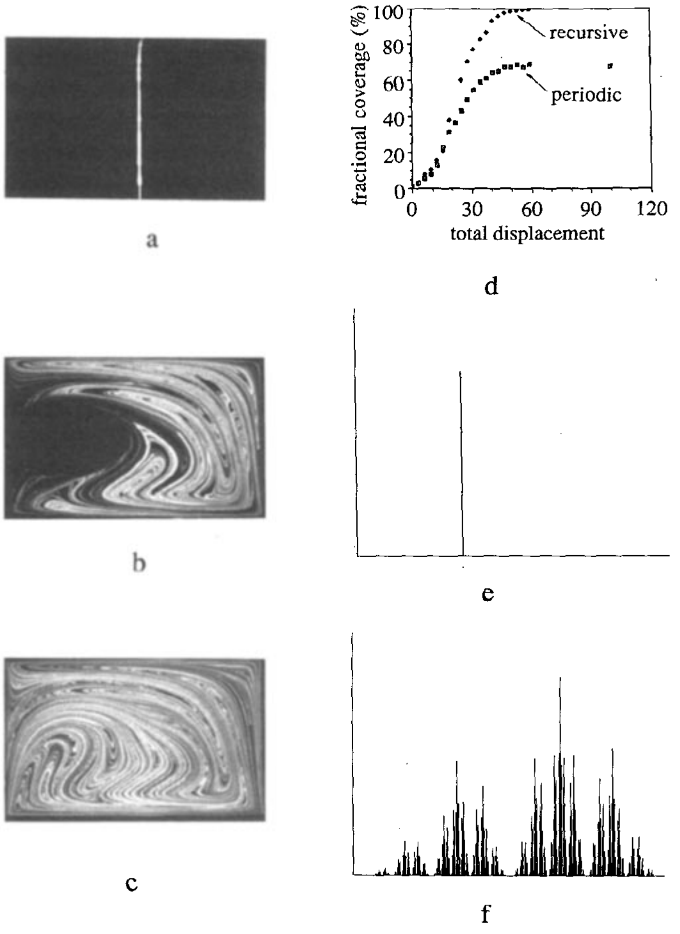


*Figure 7* Mixing in time-periodic flows and a flow generated by systematically destroying the possibility of island formation by changing the symmetries of the system. Figure (*a*) shows the initial condition, and (*b*) shows the result of mixing generated by moving the top (T) and bottom walls (B) in a time-periodic manner TBTB . . . with a displacement equal to 3.1 times the width of the cavity. In (*c*) the displacements are kept the same, but the sequence is . . . TBBTBTTBBTTBTBBT. Figure (*d*) shows the time evolution of both systems, and figures (*e*) and (*f*) show the power spectrum of the Eulerian velocity signal; clearly the spectrum (*f*) is fractal (Franjione et al. 1989).

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$$dx_1/dt = A\sin x_3 + C\cos x_2,$$
$$dx_2/dt = B\sin x_1 + A\cos x_3,$$
$$dx_3/dt = C\sin x_2 + B\cos x_1, \tag{24}$$

which is an example having $\beta = +1$. Computations reveal that this flow is chaotic (see Dombre et al. 1986); streamlines and vortex lines undergo complex paths, which can be captured in Poincaré sections. The flow has several hyperbolic points and manifold intersections that generate chaos. The condition $d\mathbf{x}/dt = 0$ implies that $\boldsymbol{\omega} = 0$ and therefore that $\nabla\mathbf{v}$ is symmetric at the fixed point (since the spin tensor is zero). It follows that the eigenvalues of $\nabla\mathbf{v}$ are real and the flow is hyperbolic (the sum of the eigenvalues is zero, since $\nabla\cdot\mathbf{v} = 0$). Chaos, however, does not imply good mixing. Since the flow is inviscid, material lines stretch as vortex lines. The length of a filament of length $\lambda_0$ placed initially to coincide with a vortex line with vorticity $\boldsymbol{\omega}_0$ evolves according to $\lambda = (|\boldsymbol{\omega}|/|\boldsymbol{\omega}_0|)\lambda_0$. Since $\boldsymbol{\omega}$ is bounded, the stretching is bounded. Due to its relative simplicity, the ABC flow has generated a number of studies and applications, primarily in magnetic fluids (e.g. Galloway & Frisch 1986, Moffatt & Proctor 1985). Some modifications of the ABC flow are easier to analyze; for example, Feingold et al. (1988) have examined a discrete version called the ABC map. A similar map was used to model the stretching of magnetic fields (Finn & Ott 1988). The problem is similar to the establishment of a statistical distribution of vorticity in a turbulent flow.

THE PPM SYSTEM The partitioned-pipe mixer (PPM) consists of a pipe partitioned into a sequence of semicircular ducts by means of rectangular plates placed orthogonally to each other, and it can be regarded as an idealized version of a static mixer or a model porous medium. The fluid is forced under Stokes flow through the pipe by means of an axial pressure gradient while the pipe is rotated about its axis relative to the assembly of plates, thus resulting in a cross-sectional flow in the cross-sectional plane in each semicircular element. If one neglects developing flows, a fluid particle jumps from streamsurface to streamsurface in between adjacent elements. Thus the flow consists of volume-preserving flow (in the ducts) followed by an area-preserving subdivision. The flow is spatially periodic, so that the most convenient choices for surfaces of section are the cross-sectional planes at the end of each periodic unit (consisting of two adjacent elements). Maps are then generated by recording every intersection of a trajectory with the surfaces of section in a very long (ideally, infinitely long) mixer and then projecting all the intersections onto a plane parallel to the surfaces. Every trajectory intersects with each surface of section, and the map captures some of the mixing in the cross-sectional flow.

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The three-dimensional structure of the flow can be obtained by plotting Poincaré sections at intermediate lengths. Each KAM curve represents the intersection of a tube with a surface of section, so that the tubes can be reconstructed by joining the KAM curves with their images in neighboring sections by smooth surfaces. The cross-sectional area of the tubes is not constant, since they explore regions with different axial speed. The KAM tubes are invariant surfaces and cannot be crossed by fluid particles; consequently, the fluid flowing within a particular tube remains in the tube and cannot mix with the rest of the fluid (Figure 8). Chaotic trajectories, on the other hand, move on two-dimensional homoclinic manifolds in the regions left free by the tubes.

The axial flow has a major effect on the Poincaré sections, and thus on the cross-sectional mixing and dispersion. For example, the Poincaré sections corresponding to plug axial flow (perfect slip at the wall) are quite different from those corresponding to Poiseuille flow. Another parameter that has a considerable effect on the Poincaré section, and thus the mixing, is the sense of rotation in the adjacent elements. The Poincaré section for the counterrotating case, which corresponds (roughly) to the configuration in the Kenics mixer (Middleman 1977), indicates that the flow is chaotic over most of the cross section and seems to mix better than the corotating case. Symmetry considerations help to optimize the degree of mixing.

The PPM is far from being completely understood. For example, little is known about the exit times of particles injected into the flow. It is possible to have exit-age distributions with two peaks, indicating the presence of anomalous dispersion. In some sense, the exit-age distribution gives an indication of the inhomogeneity of the cross-sectional mixing; it might

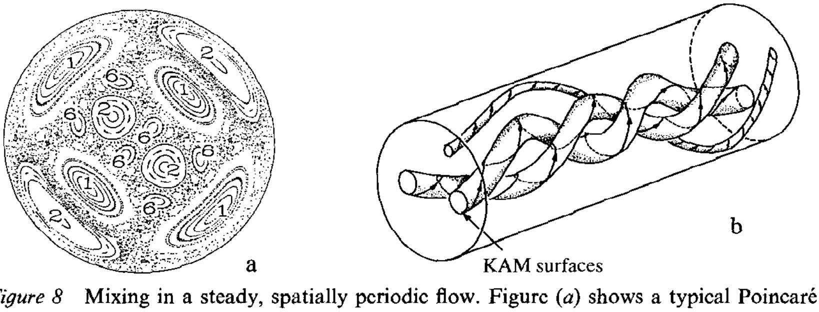


*Figure 8* Mixing in a steady, spatially periodic flow. Figure (*a*) shows a typical Poincaré section, with the numbers indicating the period of the islands; figure (*b*) shows the typical three-dimensional structure of the system. Note that the fluid within the KAM tubes does not mix with the rest of the fluid. From Khakhar et al. (1987).

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happen that fluid streams in some tubes emerge faster than others (Figure 8). However, it is in general impossible to assign speeds based on whether the particles belong to either regular or chaotic domains. Sometimes the lowest residence time corresponds to the region outside the islands; other times it is the other way around. [Obviously it is possible to include molecular diffusion in the picture and carry out simulations for a range of Péclet numbers; some work in this direction is in progress (Franjione 1989).] Our intuition can be wrong in other regards too. Two-dimensional time-periodic flows suggest that the fastest stretching takes place in the chaotic regions of the two-dimensional Poincaré sections, whereas the stretching in the regular regions is slow and inefficient. However, computational results obtained for the PPM indicate that this is not true, in general, in three-dimensional flows. In some instances, the stretching in regions identified as regular in a two-dimensional cut is larger than in chaotic regions. This seemingly contradictory result can be explained in terms of exponential stretching in the axial direction. This stretching is reminiscent of a random sequence of shear flows (Khakhar & Ottino 1986).

THE EHAM SYSTEM The eccentric helical annular mixer (EHAM) provides an illuminating counterpart for the PPM. In this case, the system is time periodic rather than spatially periodic. The cross section of this system corresponds to the journal-bearing flow, and the axial flow is a pressure-driven Poiseuille flow. There is no discontinuous jumping of particles between streamsurface and streamsurface, and the particle paths have continuous derivatives. The solution for the flow field, under creeping flow, involves no additional approximations, and the system can be studied experimentally (Kusch 1989). However, there are fewer tools available for investigation. In this case, as opposed to the PPM, the marking of intersections of trajectories of initial conditions with periodically spaced planes perpendicular to the flow reveals a blurry picture. On the other hand, intersections recorded at designated time intervals (i.e. a conventional return map) reveal no information about the axial structure of the flow. (The results are identical to those obtained for the journal-bearing flow.) The dispersion of a cloud of particles (Figures 9*a*–*c*) is relatively straightforward and yields dispersion coefficients. The most revealing visualization regarding the structure of the flow is provided by streaklines. The calculations, however, are considerably more difficult than those of the Poincaré sections, since the storage and computational requirements for smoothness increase exponentially with flow time. According to the location of injection, the streaklines can undergo complex trajectories reminiscent of the Reynolds experiment or just "shoot through" the mixer, undergoing relatively little stretching. Intermittency is also possible; since

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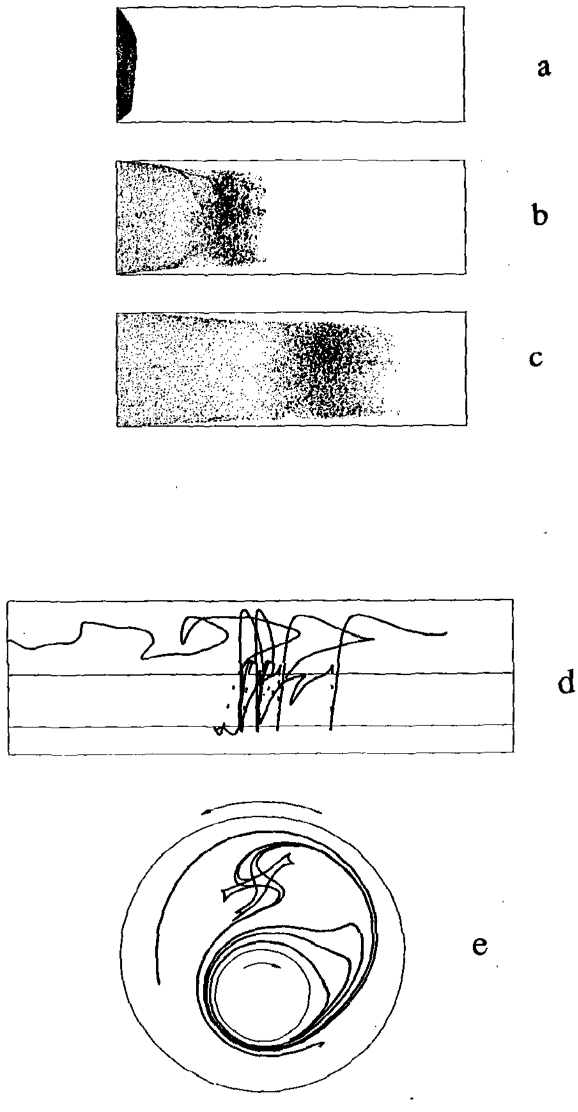


*Figure 9* Mixing in a time-dependent duct flow (EHAM). Figures (*a–c*) show the evolution of a sheet of passive particles initially located in a plane at the entrance of the mixer; (*d*) and (*e*) show lateral and end views, respectively, of a streakline that displays spatial intermittency. The flow moves from left to right. All results correspond to motion of the inner and outer cylinder of the form $V_{\theta,\text{outer}} = U\cos^2(\pi t/T)$, $V_{\theta,\text{inner}} = U\sin^2(\pi t/T)$. (The only effect of the Poiseuille flow is to compress or stretch the particle paths in the axial direction.) From Franjione (1989).

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the regular regions move, the streakline can find itself in a regular domain for some amount of time, then be trapped in a chaotic region, then escape and undergo relaminarization, and so on. It is significant that even though the axial flow always moves forward, the streaklines can "go backward," since they can wander into regions of low axial velocity, whereas other parts of the streakline can bulge forward (see Figures 9*d*, *e*). Note also that constant rate "in" does not imply constant rate "out."

SECONDARY FLOWS AND INERTIAL EFFECTS One of the practical advantages of both the PPM and the EHAM is that the axial and cross flows are independent; this allows for control of the mixing before the material exits the system. Obviously, this is not the case in flows relying on inertial effects. The secondary flow can be in a plane perpendicular to the main flow or in the same plane as the flow. Examples corresponding to secondary flows in the plane of the main flow might be based on Couette-Taylor vortices (see Swinney & Gollub 1985) or wall separation. The simplest case corresponds to the EHAM system with concentric cylinders modulated in a time-dependent fashion above the critical Reynolds number. (In this case, the strength of the vortices will be a function of time.) Another example is the time-periodic flow in a channel with wavy walls considered by Sobey (1985). The behavior of the system is dependent on the Reynolds number and the Strouhal number. A flow with a secondary flow normal to the main direction of the flow is the twisted pipe of Jones et al. (1989). The system consists of a periodic array of pipe bends [see Figure 4*k*; the bends are half-tori in the Jones et al. paper, but (*k*) corresponds to one fourth of a torus]. Within each bend there is secondary flow consisting of two identical counterrotating vortices. Several possible designs come to mind. If the planes of the bends intersect, streamline portraits taken at different axial distances reveal streamline crossing. The twisted pipe shows a KAM tube structure similar to the PPM system. Note, however, that the sense of rotations are different (compare Figures 4*d* and 4*k*). Note also that even though the system is obviously continuous and the perturbation is smooth, the analysis neglects developing flows and is basically identical to the PPM system of Khakhar et al. (1987). There are several connections between the systems in Figure 4. For example, the cavity flow can be operated in such a way as to produce two counterrotating vortices (Figure 4*j*); the position of the vortices can be varied by changing the ratio of the speeds of the walls. This makes this system somewhat similar to the cross section of the twisted pipe. Another related situation of mixing due to secondary flows arises in a meandering river; in this case, the cross flows are not mirror symmetric and the bends can belong to the same plane.

FLOWS NEAR WALLS AND FLOWS WITH FINITE REYNOLDS NUMBERS In our

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effort to make models somewhat more realistic, we are tempted to include more and more details, since we want the computer-simulated flows to do things that actual flows do. However, at what point do we stop? (This question is discussed further in Section 4.) In recent and current work (Danielson 1989), we have considered asymptotically exact solutions of the Navier-Stokes and continuity equations in two and three dimensions, constructed using a method pioneered by Perry and others (Perry & Chong 1986, 1987, Dallmann 1983). These flows are manageable and offer a good opportunity to study various aspects of the role of inertia and structural stability in three dimensions. We have focused primarily on flows near walls with separating and reattaching streamlines. The flows produce a portrait reminiscent of a heteroclinic trajectory. (In fact, things are bit more complicated, since all points at a wall with no slip, including the separation and attachment points, are parabolic.) The starting point is to expand the Eulerian velocity field in a Taylor series around a point $\mathbf{p}$, i.e.

$$\mathbf{u}(\mathbf{x}) = \mathbf{u}(\mathbf{p}) + (\mathbf{x}-\mathbf{p})\cdot\nabla\mathbf{u}(\mathbf{x})|_{\mathbf{x}=\mathbf{p}} + (1/2!)(\mathbf{x}-\mathbf{p})(\mathbf{x}-\mathbf{p}):\nabla\nabla\mathbf{u}(\mathbf{x})|_{\mathbf{x}=\mathbf{p}} + \ldots. \quad (25)$$

Each term $\nabla^{(n)}\mathbf{u}(\mathbf{x})|_{\mathbf{x}=\mathbf{p}}$ in the expansion represents a tensor of order $n+1$, which we denote $A_{ijk\ldots}$. The coefficients $A_{ijk\ldots}$ may or may not have time dependence, depending on whether the velocity field is steady or time varying (owing to some external perturbation). These tensors constitute the unknowns and are found by forcing the series to satisfy the continuity and Navier-Stokes equations, as well as the boundary and "internal conditions" of the problem in question. An enormous variety of flow topologies can be produced by this procedure. However, the number of coefficients $A_{ijk\ldots}$ grows very quickly with the order of the expansion. (For example, for a fifth-order expansion of a two-dimensional flow, the number of coefficients is 42; using both the continuity and Navier-Stokes equations eliminates just 21 coefficients, which leaves another 21 coefficients to be specified.) Typical boundary conditions are no slip at the wall and to impose the value of the vorticity at the wall (a sign change in vorticity at the wall is used to specify separation); another is to specify angles of separation and attachment. A typical internal condition is to specify the presence and location of elliptic or hyperbolic points in the flow region. It is relatively straightforward to produce flows with homoclinic or heteroclinic trajectories; time-dependent perturbations can produce chaotic trajectories in two-dimensional flows with transverse homoclinic and heteroclinic trajectories. Material can invade the bubble or leak from the bubble (see Figure 10). Time-varying perturbations generate a series of nonlinear ordinary differential equations for the time evolution of the coefficients

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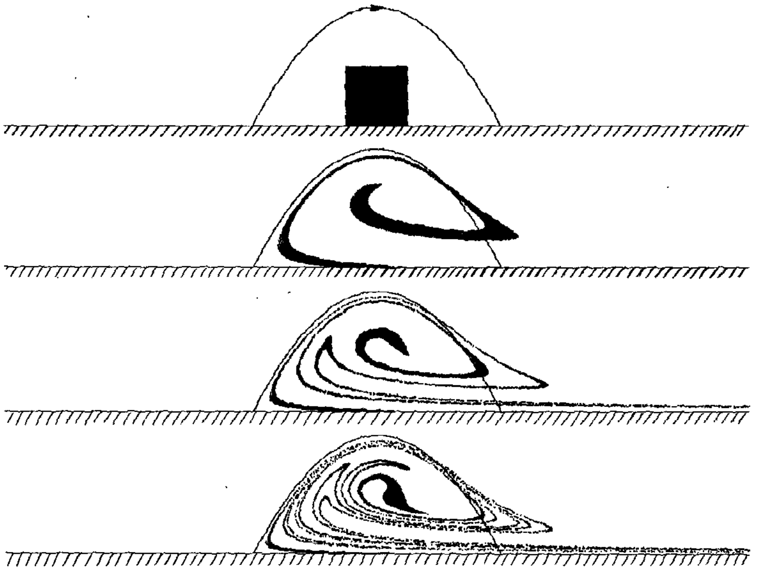

*Figure 10* Behavior of a separation bubble under a time-periodic perturbation. The top figure shows an initial square of passive particles inside the bubble; the other figures show the time evolution as the particles leak from the bubble. At all times there is a streamline attached to the wall, as shown in the top figure; the leaking particles cross the streamline (Danielson 1989).

$A_{ijk\ldots}$. Typically the system contracts volume in phase space. It seems therefore possible to generate perturbations leading to strange attractors, especially in three dimensions. This in turn implies Eulerian chaos.

ADVECTION OF VORTICITY IN CHAOTIC FLOWS This is a convenient place to bring into perspective a few points common to the examples described so far. Any two-dimensional flow ($x$–$y$ plane) satisfies

$$\nabla^2\psi = -\omega_z. \tag{26}$$

In Stokes flow (i.e. Re $\to 0$ and Sr $\to 0$), where

$$\nabla^4\psi = 0, \tag{27}$$

the streamfunction adjusts instantly to time-dependent boundary conditions, and even though a passive scalar might be mixed chaotically by the flow, the streamfunction $\psi(\mathbf{x}, t)$ is never truly complex. In particular, the vorticity distribution *is not* advected by the flow and is simply given

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by Equation (26). This situation was encountered in the cavity flow and the journal-bearing flow; the vorticity distribution simply stays in place.

At finite Reynolds numbers, the vorticity is advected according to

$$D\boldsymbol{\omega}/Dt = \boldsymbol{\omega}\cdot\nabla\mathbf{v}+\nu\nabla^2\boldsymbol{\omega}. \tag{28}$$

In two-dimensional flows there is no vortex stretching, and the vorticity diffuses according to

$$D\omega_z/Dt = \nu\nabla^2\omega_z. \tag{29}$$

The fact that a two-dimensional chaotic flow is able to mix a scalar $c$ but unable to mix vorticity is sometimes a source of confusion. However, in the limit $\mathrm{Re} \to 0$ the passive scalar obeys the equation

$$Dc/Dt = \mathscr{D}\nabla^2 c, \tag{30}$$

where $\mathscr{D}$ is the diffusion coefficient, whereas the vorticity field is simply given by

$$\nabla^4\psi = -\nabla^2\omega_z = 0. \tag{31}$$

Evidently the two problems are not equivalent, and the scalar field (e.g. a material line stretched and folded by a chaotic flow) can be infinitely more complex than the vorticity field.

The situation is obviously different at finite and large Reynolds numbers. In the limit $\mathrm{Re} \to \infty$, the vorticity equation reduces to

$$D\boldsymbol{\omega}/Dt = \boldsymbol{\omega}\cdot\nabla\mathbf{v}, \tag{32}$$

vortex lines move as material lines, and both can be stretched and folded into complex structures characteristic of chaotic flows. The solution of Equation (32),

$$\boldsymbol{\omega} = \boldsymbol{\omega}_0\cdot\mathbf{F}^{\mathrm{T}}, \tag{33}$$

where $\boldsymbol{\omega}_0$ is the initial value of the vorticity, indicates the very same point. However, (33) might also give the mistaken impression that the evolution of vorticity can be calculated on purely kinematical grounds. This is not true; the deformation tensor $\mathbf{F}$ cannot be calculated until the velocity field is obtained by solving Euler's equation. Most of the three-dimensional flows studied to date are unable to shed much light on the connection between chaos and vortex stretching, and thus new examples must be found. In particular, if $\mathbf{v}$ is *given*, as in the ABC flow, then the vorticity distribution is simply given by $\boldsymbol{\omega} = \nabla\times\mathbf{v}$ and there is nothing else left to do.

A simpler situation occurs for two-dimensional inviscid flows. In this case, we have

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$$D\omega_z/Dt = 0, \tag{34}$$

and fluid particles conserve their initial value of vorticity, i.e. $\omega_z(\mathbf{x}, t) = \omega_{z_0}(\mathbf{X})$. [Most of these issues were ignored in the analysis of prototypical flows, such as the tendril-whorl flow, in order to highlight the chaotic and kinematical aspects of the problem (see Section 3.1).] It is also clear that examples based on singularities—e.g. point vortices, such as the blinking-vortex flow, or an oscillating pair of vortices—cannot clarify any aspect of the question of mixing of vorticity. In all cases, $\boldsymbol{\omega} = 0$, except at the vortices themselves, and there is no mixing of vorticity to speak of.

Are chaotic flows, such as the ones considered in Section 3, able to mix vorticity? The answer seems to be yes, but many questions remain: For example, it does appear that mixing of vorticity can occur in some chaotic flows, such as in our example of "flows near walls," and especially in three dimensions. However, this question has not been analyzed in detail yet, and it is instructive to consider simpler cases instead. In the next section we consider a few of these issues in terms of a shear layer; this example serves also to clarify some of the subtle connections between Lagrangian and Eulerian viewpoints.

SHEAR AND OPEN FLOWS The apparatus of dynamical systems is better suited to handle chaotic mixing in closed flows than in continuous flows. This is particularly clear in the analysis of the Kelvin's cat's-eye flow. The streamfunction with respect to a fixed laboratory frame is of the form

$$\psi(x_1, x_2, t) = ux_2 + \ln[\cosh x_2 + A\cos(x_1 - ut)], \tag{35}$$

where $u$ represents the average speed, and $A$ is a parameter quantifying the concentration of vorticity. (The case $A = 1$ corresponds to point vortices; the case of interest here corresponds to $0 < A < 1$, for which the distribution of vorticity is a smooth function of position.) With respect to a frame $x'_1$–$x'_2$ moving with the vortices, the streamfunction is time independent and has the form

$$\psi'(x'_1, x'_2) = \ln[\cosh x'_2 + A\cos x'_1]. \tag{36}$$

The flow is a succession of hyperbolic and elliptic points with connecting heteroclinic orbits (cat's-eye structure) and is an excellent candidate for chaos under a time-periodic perturbation of the form $v'_1 = \varepsilon \sin(\omega t)$. If we proceed in the same fashion as in our previous examples, the evolution equations are

$$dx'_1/dt = \partial\psi'/\partial x'_2 + \varepsilon\sin(\omega t),$$

$$dx'_2/dt = -\partial\psi'/\partial x'_1. \tag{37}$$

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Computational studies, e.g. Poincaré sections as well as analytical techniques (Melnikov method), show chaotic behavior. Significantly, the chaotic behavior is maximized for an intermediate value of the perturbation frequency [$\omega \approx 0.3$ (Danielson 1989; see also Ottino 1989a); experiments show a similar behavior (Roberts 1985), but an explanation of the experimental results probably lies outside the ability of this simple model]. However, the manifolds associated with periodic points make sense only in a moving frame. This kind of chaotic behavior means little to an observer conducting experiments in a laboratory frame (where the cat's-eye structure is seen). In the laboratory frame, the technique of choice is streaklines. However, streaklines injected with respect to the fixed frame reveal significant stretching and folding, but *far less* than anticipated based on what is observed in terms of the moving frame. In fact, rather large perturbations ($\varepsilon \approx 0.5$) show no appreciable change with respect to the integrable case ($\varepsilon = 0$). In principle, it is possible to define chaotic behavior with respect to streaklines ("streakline horseshoe"; see Rising 1989), but much more work is necessary in this regard.

The flow is obviously not turbulent. However, what is the most important single ingredient missing from it? The answer is that it does not mix vorticity. In fact, as indicated in the previous section, every flow based on perturbing an analytical expression for the streamfunction [in this case, Equations (37)] cannot possibly create a complex pattern of vorticity, since at the most the vorticity will be as complicated as the velocity field itself. What is the simplest way to produce mixing of vorticity? The simplest possibility is to regard the vorticity as purely passive and advect it according to (37). However, this does not address the most important issue: If the flow is perturbed, then $D\omega_z/Dt \neq 0$, and the equations violate conservation of momentum. Is there any simple and illustrative way (short of solving the Euler equations numerically, which takes the problem outside the boundaries of this review) to satisfy $D\omega_z/Dt \equiv 0$ and at the same time preserve the basic cat's-eye flow structure of the flow? An approximate way to take care of the problem is to augment the evolution equations (37) with the constraint $D\omega_z/Dt = 0$ by letting $A$ be a function $A(\mathbf{X}, t)$. At $t = 0$ the flow satisfies Euler's equation. Subsequently, a particle $\mathbf{X}$ carries a value of $A(\mathbf{X}, t)$ in such a way that $D\omega_z/Dt \equiv 0$ along a Lagrangian trajectory. [Each $\mathbf{X}$ originates a different history $A(\mathbf{X}, t)$; there are also other complications (T. J. Danielson & J. M. Ottino, unpublished, 1989).] Figure 11 shows the effect of such a flow on an isovorticity contour. Even though each particle in the contour has the same vorticity, the line can engulf vorticities belonging to other regions, and there is significant mixing of vorticity in the neighborhood of the hyperbolic points. A significant consequence of this example is to show that, as a first approximation, the

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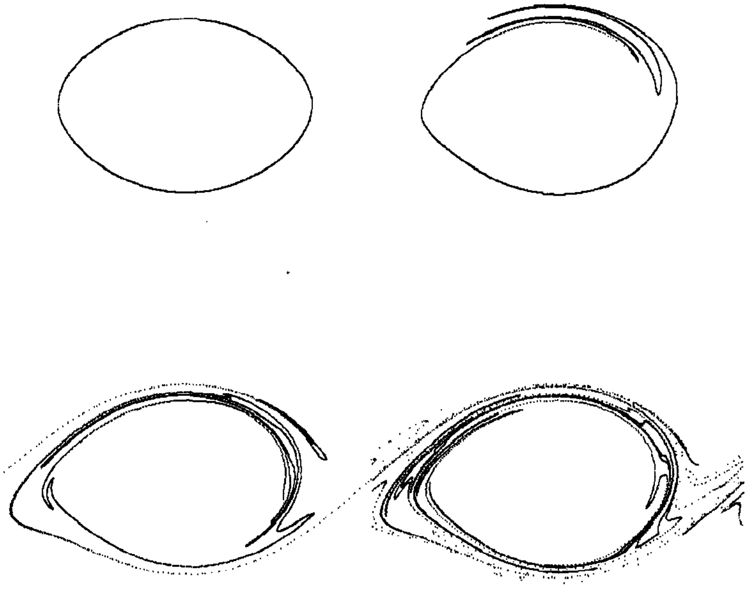

*Figure 11* Evolution of a contour of isovorticity (i.e. $\omega_z$ = constant) in the perturbed Kelvin cat's-eye flow (Danielson 1989).

vorticity can be regarded as a passive scalar advected by the flow. It shows also that Euler's equation is critical in producing chaotic distributions of vorticity, and further exploration of these issues should be continued. It should be reemphasized that more standard simulations of shear layers in terms of sequences of interacting point vortices, while capable of revealing other aspects of the problem, will not lead to mixing of vorticity, since $\omega_z = 0$ except at the vortices themselves. Vortex patches are another possibility. In this case, however, the attack has to be mostly computational, and the utility of analytical methods is somewhat lost.

## 4. KINEMATICAL CHAOS AND TURBULENCE

It is obvious that the ideas of the previous sections have modified our perception about mixing in viscous flows. A large number of problems in nature and industry fall in this category (see Figure 1). However, there has been considerable speculation regarding the connection between chaos and turbulent flows. Questions have been raised as to whether or not these new ideas will be able to "solve" the turbulence problem and allow predictions

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of practical value. It is probably a good idea to address the full problem here.

What constitutes "chaos theory"? The first thing that should be clarified is that there is not a single chaos theory. In the context of fluid mechanics we can recognize two large branches, one corresponding to volume-contracting or dissipative systems, and the other to Hamiltonian systems and area-preserving systems. [A system can be volume preserving and not be Hamiltonian; however, if it is Hamiltonian, it is volume preserving.] Another possibility, occupying a somewhat intermediate status between dynamical systems and traditional fluid mechanics, is vortex dynamics. In two dimensions and with point vortices it is analogous to Hamiltonian mechanics (see Aref 1983). In three dimensions the analogy is lost and there is less theoretical guidance; the attack is then computational (Leonard 1985).

*Dissipative systems* are associated with one-dimensional maps (such as the logistic equation), systems of ordinary differential equations [such as in the Lorenz equations (Lorenz 1963)], and strange attractors characterized by fractal dimensions where the motion is chaotic. This is the territory where truncation of infinite-dimensional systems into a finite number of (Fourier) modes becomes an issue. Typically, the phase space does not coincide with the physical space. If the model is continuous, a dissipative system must consist of at least three (autonomous) ordinary differential equations in order to exhibit chaos (as in the Lorenz model). On the other hand, if the model is represented by a mapping $\mathbf{x}_n \rightarrow \mathbf{x}_{n+1}$, its phase space is free of topological constraints present in continuous models, and it can display chaos in one dimension, i.e. with $\mathbf{x}_n$ being real (as in the logistic equation). The emphasis on dissipative systems is relatively recent, and it can be traced to the articles by Feigenbaum (1978, 1979a,b, 1980), Lorenz (1963), and Ruelle & Takens (1971). An accessible review describing various routes (scenarios) to turbulence is given by Eckmann (1981). There are at least two major reviews written on the subject in the context of fluid mechanics (Lanford 1982, Guckenheimer 1986); several books devote considerable space to the mathematical aspects of the problem, most notably Guckenheimer & Holmes (1983), Wiggins (1988), and Devaney (1986). In the context of fluid mechanics, the major applications are to closed flows, primarily the Taylor-Couette flow (e.g. Fenstermacher et al. 1979, Swinney 1985) and the Rayleigh-Bénard flow (Libchaber & Maurer 1980, Heslot et al. 1987). Applications of the concepts to open flows, such as the two-dimensional von Kàrmàn wake behind a cylinder, present more difficulties (Sreenivasan 1985). For a general review of this entire area, see Swinney & Gollub (1985).

*Hamiltonian systems and area-preserving systems* are considerably older

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and, to some extent, better understood than dissipative systems. The study of chaos in Hamiltonian systems can be traced to the beginning of the twentieth century, and to problems such as the three-body problem and the stability of the solar system. In fact, the first hints of chaos in Hamiltonian systems were noted by Poincaré in connection with his studies in celestial mechanics. A few aspects of the theory of chaos in Hamiltonian systems have been studied in considerable detail. When the perturbations from integrability are small, the problem is relatively clear and several theorems describe the behavior of the system (KAM theorem, Poincaré-Birkhoff theorem, Moser's twist theorem). However, much less is known when the perturbations are large. [For an introduction to Hamiltonian systems, see Percival & Richards (1982); a good review in the context of chaos is given by Helleman (1980); a complete treatment is given by Abraham & Marsden (1985); and an accessible review of both Hamiltonian and dissipative systems is given by Doherty & Ottino (1988).]

As opposed to model development in dissipative systems, Fourier expansions do not play an important part in the model development in Hamiltonian systems. The physics is contained in the Hamiltonian structure itself; there are no modes to speak of, and the system is defined by such quantities as (generalized) position and momentum. In particular, in Hamiltonian systems there are no stable steady states, the phase space does not contract, and there are no attractors, strange or otherwise. However, there is "recurrence," and in a few simple cases it can be shown that there is ergodicity.

Probably, the closest point of connection between dissipative, Hamiltonian, and volume-preserving systems is that chaos can be traced back to a stretching-and-folding mechanism in phase space. Stretching and folding is the fingerprint of chaos. Stretching and folding of fluid elements (not just in phase space) occurs also during mixing in turbulent flow. In fact, stretching and folding is also the fingerprint of mixing (Ottino 1989b). Apparently, mixing is the clearest connection between turbulence and chaos. Is there any real connection between chaotic advection and turbulence? Let us consider now the nature of the questions asked by chaos and classical approaches to turbulence (Table 1), along with the classical symptoms defining turbulence [Table 2, based on Tennekes & Lumley (1980, Section 1.1)]. [A similar classification to that of Table 1 was proposed by Chapman & Tobak (1985); for a glimpse into the early questions in turbulence, see the report by von Neumann (1963).]

There is no universally accepted way of defining chaos, although several definitions are equivalent from a mathematical viewpoint. Generally, a system can be classified as chaotic if it satisfies any of the following criteria: (*a*) the flow produces either transverse homoclinic or transverse

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**Table 1** Approaches to turbulence

- CLASSICAL HYDRODYNAMIC STABILITY (e.g. classical analysis of Taylor-Couette flow)
  *premise*: Analysis is based on small perturbations of known deterministic (laminar) flows.
  *questions*: How does a flow become unstable? How is turbulence generated?
- STATISTICAL THEORY (e.g. homogeneous isotropic turbulence and extensions)
  *premise*: Flow is disorganized to start with.
  *questions*: How is turbulence maintained? How is energy transferred between scales?
- STRUCTURAL STUDIES (observation of coherent structures, turbulent spots, etc.)
  *premise*: Turbulent flows might exhibit some degree of organization.
  *questions*: What large-scale structures are present within various types of turbulent flows?
- DETERMINISTIC CHAOS OR "CHAOS THEORY"
  *premise*: Complex behavior is possible in systems with few degrees of freedom.
  *questions*: How does a system become chaotic? What are the fingerprints of chaos? What are the typical structures characteristic of chaotic flows? What is the relationship between these structures and actual turbulent flows?

heteroclinic intersections, (*b*) the flow has a positive Liapunov exponent, or (*c*) the flow produces horseshoe maps. All these definitions are amenable to mathematical proof. Often the definitions are computationally based. For example, the visual appearance of numerically computed Poincaré sections is taken as evidence of chaos. In the context of dissipative systems, "chaos" can be interpreted as a system that has an attractor with at least one positive Liapunov exponent (a so-called "strange attractor"). Much of the experimental work on chaos involves the case of a time-dependent signal $x(t)$; in such cases various other definitions of chaos are used (Swinney 1985). One common definition is to compute the power spectrum of $x(t)$; an indication of chaos is a continuous spectrum. Another possibility is to compute the correlation function $c(\tau)$ of the signal $x(t)$. Thus, when reading the literature, it is important to determine what type of definition is being used by the author. On the other hand, turbulence is a collection of symptoms and does not admit a simple one-line definition (see Table

**Table 2** Turbulence: a list of symptoms

- The signals of Eulerian quantities, such as velocity $\mathbf{v}(\mathbf{x}, t)$ and pressure, are complicated (temporal disorder).
- The flow has the ability to mix.
- There is energy transfer from large to small scales.
- There is mixing of vorticity (spatial disorder), and there is vorticity intensification until intensification is balanced by dissipation.
- The flow is characterized by a large Reynolds number.
- Intermittency (both temporal and spatial) is observed to occur.

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2). A flow is diagnosed to be turbulent if it presents the proper symptoms. (By no means will all researchers agree as to which ones are the "right symptoms," but it is generally agreed that the flow has to be able to produce both spatial and temporal disorder.) It is simplistic to seek a clean answer to the questions of whether turbulence is chaotic or chaos is turbulent. It is clear that there are chaotic flows that are not turbulent. As to whether turbulent flows are chaotic, the answer depends largely upon the definition.

None of the chaotic flows studied to date can mimic all of the essential symptoms of turbulence (see Table 3). In fact, the specification of $\mathbf{v}(\mathbf{x}, t)$ precludes the existence of turbulence from the outset. It would appear that any sensible theory of turbulence has to produce the Eulerian velocity, or its statistical properties, as an *output*, and that it would not make much sense to regard $\mathbf{v}(\mathbf{x}, t)$ as an *input* into the problem. However, there seems to be some middle ground; we have seen how some flows lead to a fractal velocity signal at a fixed point, how strange attractors might appear in the context of volume-preserving velocity fields, and how a constraint of conservation of vorticity along a Lagrangian trajectory might lead to mixing of vorticity. Various simple systems display a symmetry-breaking process characteristic of the transition from laminar to turbulent flow (Beloshapkin et al. 1989); other systems, such as Rayleigh-Bénard flow, can be studied from the viewpoints of both strange attractors and chaotic advection and provide a bridge between the two viewpoints (Solomon & Gollub 1988). These systems are a beginning. It is clear that if we put all details into a model we will get more and more of the symptoms listed in Table 2, but then the attack of the problem must be purely computational. In fact, once the flows become more realistic, the key question seems to be the following: How "realistic" should the flow be in order to capture expected phenomena but at the same time preserve the utility of the dynamical-systems tools? There are probably two ways to look at this

**Table 3** Properties of chaotic flows taking $d\mathbf{x}/dt = \mathbf{v}(\mathbf{x}, t)$ as a dynamical system

ESTABLISHED
- The flow stretches and folds, i.e. it is able to mix effectively.
- Simple Eulerian velocity fields $\mathbf{v}(\mathbf{x}, t)$ produce complex particle paths $\mathbf{x} = \mathbf{\Phi}_t(\mathbf{X})$.
- There is coherence of large-scale structures.
- There are symmetry-breaking transitions.

MORE STUDIES NECESSARY, BUT POSSIBLE IN PRINCIPLE
- The flow displays spatial intermittency.
- The flow produces Eulerian turbulence.
- There is mixing of vorticity in two dimensions.
- There is stretching and folding of vorticity; flow produces a fractal set of vorticity.

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question. On the one hand, there should be exploration of simple models in a progression from simple to complex; most of the flows presented in this review belong to this category. What are the common causes for complex behavior? Can they be isolated in simple pictures? On the other hand, if the ideas are to have any impact in systems of engineering interest, work should be done in trying to mimic actual experimental occurrences in classical turbulent flows. A highly developed model attempting to bridge the two camps in the context of a turbulent boundary layer was developed recently by Aubry et al. (1988).

## 5. PROGNOSIS

Chaotic mixing can clarify problems in natural sciences and inspire the design of mixing devices. Applications have been found in areas as varied as explaining the generation of magnetic fields in the Universe (Finn & Ott 1988) and the anomalous dispersion of pollutants in oceans (Pasmanter 1988). Other applications will undoubtedly follow (see Figure 1), and open problems abound. In many cases a computational determination of the velocity field is the only route. In this case the analysis is necessarily more restricted than an analysis based on analytical descriptions, and there is a need for establishing bounds on the errors introduced by numerical schemes (for example, location of manifolds, establishing the size of small islands, etc.). In this regard, computations based on boundary-integral methods might be more accurate than standard finite-element simulations. However, standard finite-difference schemes are most definitely sufficient to capture most of large-scale detail—folds, incomplete horseshoes—in experiments such as the bottom frame of Figure 5 (Leong 1989). Nevertheless, substantially more work is also necessary at the basic mathematical level; for example, we cannot yet predict with certainty the spatial extent, location, and movement of islands in two-dimensional time-periodic flows. Fundamental questions remain at the very core of chaos, especially regarding the role of symmetries and bifurcations. In this regard it is important to note that experiments can be used as a prototype for the understanding of chaos from a mathematical viewpoint, since they provide excellent resolution of chaotic regions (Ottino 1989b). An important question to keep in mind throughout the examples of Section 3 is, What constitutes a complete analysis or understanding of a chaotic mixing flow? A loose definition as to what constitutes complete understanding is the following: A system can be regarded as "completely understood," from a practical viewpoint, when the answer to the $n+1$ question in a program of analysis can be qualitatively predicted from the previous $n$ answers (examples of such questions might be the following: What size have the

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outermost KAM curves associated with islands of period-2? How "large" is the region invaded by the manifolds belonging to the period-1 hyperbolic points? At what parameter values do large period-1 islands bifurcate?). Such an understanding would signify that most of the basic aspects of the flows have been grasped, and design of mixing processes from first principles would therefore be possible. However, from this viewpoint very few of the flows sketched in Figure 4 can be regarded as completely understood. Little is known about three-dimensional volume-preserving flows; more insightful examples are necessary to establish the analogies and differences in the behavior of spatially periodic and time-periodic continuous flows (such as the PPM and EHAM flows; can the twisted-pipe flow produce two peaks in an exit function?). Another major problem is open flows—in particular, establishing the behavior of streaklines and connecting the pictures of chaos in moving and fixed frames.

The simplest problems of mixing in chaotic flows involve a single fluid (i.e. mixing of nearly identical fluids or passive particles). However, other processes can be imagined to occur on the fabric of a chaotic flow. One possibility is coagulation; others are molecular or Brownian diffusion and the behavior of active particles, which present resistance to stretching and might break. Stirring can unmix (e.g. starting with Figure 2*d*, go back to Figure 2*a*). One of the simplest problems is coagulation in a globally chaotic two-dimensional flow; a study is presented by Muzzio & Ottino (1988). The main result is that even though there is no molecular diffusion, the coagulation process is governed by Smoluchowski's equations; the kinetic constants can be calculated based on simple arguments and depend on the strength of the flow. These kinds of problems can be extended in several directions. Clear possibilities are adding molecular diffusion, inertia, volume growth, and growth of ramified structures; in particular, breakup processes might admit some statistical treatment.

Molecular diffusion can be incorporated in two different ways. The simplest possibility is to add white noise to the flow $\mathbf{\Phi}_t(\cdot)$ itself. However, if the problem involves interdiffusion and reaction between two fluids, an approach in which particles are followed is very inconvenient and hard to generalize for complex chemical reactions. The most important aspects of the problem can be incorporated in terms of thinning striations [lamellar models (Ranz 1979, Ottino et al. 1979)]. At the moment little is known regarding the character of the striation-thickness distributions generated by chaotic flows; however, a possible approach can be based on the work by Ott & Antonsen (1988) and especially Finn & Ott (1988). Their approach yields the fractal dimension of well-mixed structures produced by variants of the baker's transformation using twist-and-fold maps. Another important problem relevant to combustion is fast reactions in lamellar

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structures. Recent work indicates that the decay of concentration fluctuations and the rate of conversion are nearly independent of the original distribution due to the evolution of the striation-thickness distribution into a scale-invariant universal distribution (Muzzio & Ottino 1989).

Another problem in which molecular diffusion is important is the flow of tracers in porous media. In this regard it is interesting to notice that a steady two-dimensional model of a porous medium is incapable of generating chaotic particle trajectories, since streamlines obviously do not cross. [An early investigation of kinematical reversibility in two-dimensional porous media is due to Hiby (1962).] Systems related to the PPM might serve as model analogs of porous media. The system has the advantage that it is mathematically tractable and can generate trajectories reminiscent of a disordered porous medium. Computations of dispersion should account for a range of Péclet numbers.

How much computational work is possible in the design of mixing devices? Open problems involve stretching and breakup (see Figure 2) and mixing of viscoelastic fluids (C.-W. Leong & J. M. Ottino, unpublished); opportunities abound at both the experimental and theoretical levels. What is the smallest size droplet that can be generated by chaotic dispersion? (Drops can be extended several order of magnitude without breakup. It should be possible to tailor motions to produce large stretching, break the filaments by capillary instabilities, and then disperse the resulting droplets; however, these small droplets might be hard to break.) Can chaotic mixing be used in conjunction with Taylor diffusion? Can chaotic advection be used to enhance separation of diffusing substances? (Aref & Jones 1989). Can the theory be extended to predict the rate of mass or heat transfer between the walls and the bulk of the fluid in the PPM or EHAM? [These devices should be useful in many biomedical applications; a good model is the work of Sobey (1985) in a system that originated in the development of high-performance mass-transfer devices.] Other practical examples include the cleaning of small cavities with jets of fluids, or the mass transfer between a cavity and a time-dependent fluid stream. (These sort of problems appear in the microelectronics industry.) Chaotic advection should greatly increase the efficiency of the mass-transfer rate and the cleaning efficiency. Other possibilities should be explored in the polymer industry. The theoretical challenge in much of the current equipment is that the mixing geometry is a function of time (for example, in twin-screw extruders). The theory and analysis developed to date should inspire the design of new geometries, especially in continuous flows such as the PPM.

Finally much more should be possible regarding the connection between chaotic advection and turbulence, and tractable model flows should be

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developed to mimic the symptoms of Table 1. In turn these models should be useful in establishing the bounds of what are reasonable and unreasonable expectations in direct simulations. Recent models have focused on "generic" patterns that arise in hydrodynamical systems in the transition to turbulent flow. Steady flows (the ABC flow is an example) produce symmetric, crystallike structures separated by a web of chaotic streamlines; the next step in the evolution should be to explore models that produce temporal chaos. We have seen how symmetries, and lack thereof, might be intimately bound to turbulence, and how a systematic elimination of islands leads to a fractal Eulerian spectrum (see Section 3.2); all of these concepts might be related at a deeper level. Probably the biggest contribution of chaotic advection in the context of turbulence might be at the level of small-scale quasi-laminar flows (as advocated by Khakhar et al. 1986) or in the understanding of preturbulent flows. It seems profitable to try to mesh these ideas with some of the more classical concepts in Eulerian turbulence.

As usual when a new idea is put forward, there is a considerable degree of overshooting and hurried claims; it is often perceived as if one paradigm is being completely replaced by another (Kuhn 1970, pp. 153–56). A total replacement will not occur in this case. In fact, we are of the opinion that the new developments should be able to coexist peacefully with previous approaches to turbulence and hopefully complement and augment existing concepts. In contrast to viscous mixing, practical applications to turbulence appear to be some way off. Nevertheless, it is apparent that the new developments will affect our perception of what constitutes turbulence and how it is generated.

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