Chemical Reaction Fronts in Ordered and Disordered Cellular Flows with Opposing Winds

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We present experiments on the motion of chemical fronts in ordered and disordered vortex flows with imposed uniform winds. Fronts in a chain of alternating vortices are found to freeze (pin to the separatrix) for a wide range of opposing winds that grows nonlinearly with the characteristic strength of the underlying vorticity. Experiments in spatially disordered flows demonstrate that freezing of fronts is common to cellular flows; furthermore, it is not dependent on boundary conditions. We therefore anticipate similar pinning in a wide range of 2D cellular flows and front-producing systems.

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Numerous chemical, biological, and physical systems are characterized by two coexisting phases and by the movement of a front that separates these phases. Front propagation is relevant to a wide variety of dynamical processes, including natural and industrial chemical processes [1], plasma systems [2], solidification [3], the spreading of a disease in a population [4], and marine ecology systems [5]. The dynamics of front propagation are well understood in stagnant fluids [6,7]; an issue of significant current interest [8] is how fluid flows affect the motion of fronts. This issue is of particular importance in light of recent and ongoing development of microfluidic devices for both chemical processing and biological diagnostic applications.

If a uniform "wind" opposing a front is applied to an otherwise motionless fluid, the front simply propagates at its reaction-diffusion (no flow) velocity minus the wind speed. If the same wind is applied to a fluid with an underlying cellular flow, however, the behavior is dramatically different. In this Letter, we present experiments showing that cellular flows can freeze the motion of a reaction front against an opposing wind. We study this phenomenon as a function of the strength of the vortex flow. Fronts produced by the ruthenium-catalyzed excitable Belousov-Zhabotinsky (BZ) reaction [9,10] are studied in both ordered and disordered vortex flows. The flows studied are time independent; nevertheless, the experimental results may have implications for many twodimensional (2D), vortex-dominated, time-dependent flows as well.

In the absence of fluid flows, a front propagates with a reaction-diffusion (RD) velocity given by the well-known Fisher-Kolmogorov-Petrovskii-Piskunov (FKPP) result [6,7] $v_0 = 2\sqrt{D/\tau}$, where *D* is the molecular diffusivity and τ is the reaction time scale. In the past 10 years, there has been growing interest in the effects of fluid advection on front propagation. Theoretical [11] and experimental [12] studies have shown that a front propagating against a simple pipe flow moves with velocity v_0 independent of the imposed flow due to no-slip boundary conditions at the

walls. No-slip conditions also play a key role in explaining anomalous front velocities in flows through a porous medium [13]. Other theoretical studies of front propagation in advection-reaction-diffusion (ARD) systems include predictions of fractal fronts in open flows with chaotic mixing [14,15]. Recent theories [16–18] have extended the standard FKPP theory to account for nondiffusive transport. However, to our knowledge, there are no general theories of front propagation in ARD systems that account explicitly for the effects of cellular flow structures, which dominate in a wide variety of natural and industrial, predominately 2D flows. The importance of such flow structures, for example, is in evidence in time-periodic vortex chains, which cause fronts to mode lock to the external forcing [19,20]. A mode-locked front propagates an integer number of vortex pairs in an integer number of oscillatory drive periods, inconsistent with straightforward FKPP approaches. Our experiments indicate the fundamental importance of coherent vortices in general descriptions of ARD dynamics.

Two flows are used for most of these experiments (Fig. 1): a chain of 20 counterrotating vortices and a random vortex flow, both created using magnetohydrodynamic forcing [21] and confined to an annulus bounded by Plexiglas rings of radii 6.1 and 8.3 cm. A radial current passing through a 2-mm-thick electrolytic solution inter-



FIG. 1. Schematics of the fluid flow for $\epsilon = 0$ (no wind). (a) Ordered chain of counterrotating vortices. The maximum vortex velocity U is measured at the separatrix. (b) Two-dimensional disordered array.

acts with a magnetic field produced by Nd-Fe-B magnets below the fluid. Two concentric rings of 3/4-in.-diameter magnets with alternating polarity are used for the ordered vortex chain, and a disordered pattern of 1/4-in. magnets is used for the random flow. The magnets are mounted on a motor that rotates at a constant rate, thus moving the vortices. In a reference frame moving with the magnets, the vortices are stationary and there is a constant, uniform wind with a velocity W equal to the drift velocity of the magnets. (This has been verified experimentally with dye and particle-tracking transport studies that agree with numerical simulations of transport in a vortex chain with an imposed wind [22].)

The electrolytic solution is composed of the chemicals for the excitable Ru-catalyzed BZ reaction [10]. In this reaction, orange Ru^{2+} ions are oxidized to a green Ru^{3+} state, forming a propagating pulselike front, which is imaged using a 12-bit CCD camera with a red interference filter. The Ru-catalyzed BZ reaction is also inhibited by blue-green light. We use photoinhibition techniques to confine the reaction to the area of interest [21]. The reaction is triggered with a silver wire. The vortices are stationary (i.e., with no wind) at the beginning of each run; the wind is gradually increased to the desired strength after the reaction is initiated. The enhanced images are digitally "decurled" into a linear chain and shifted into a reference frame moving with the vortices for analysis.

The front velocity v_f , wind speed W, and maximum vortex velocity U are all scaled by the RD velocity: $v = v_f/v_0$, $\epsilon = W/v_0$, and $\mu = U/v_0$. In the limit $\mu \to 0$, the addition of a uniform wind is the equivalent of a Galilean transformation; in a reference frame moving with the wind, there is no flow. Consequently, for $\mu = 0$

$$\nu = 1 - \epsilon. \tag{1}$$

In this limit, a front is "frozen" (i.e., $\nu = 0$) only if W is precisely equal to v_0 , i.e., if $\epsilon = 1$.

The addition of underlying vortex structures ($\mu \neq 0$) has a significant effect on front behavior. Sequences of images (Fig. 2) show the regimes of front propagation in this case. In an ordered vortex flow with small ϵ [Fig. 2(a)], the front propagates forward against the wind; it is advected around each vortex and then burns across the separatrix from one vortex to the next. The front freezes for intermediate wind speeds [Fig. 2(b)]: the wind prevents the front from burning across the separatrix into the next vortex. However, the wind does not blow the front backwards even though ϵ significantly exceeds 1; instead, the front circles in the leading vortex. In the comoving reference frame of Fig. 2, the front is *frozen*; in any other reference frame, the front is *pinned* to the motion of the leading vortex. For large enough ϵ [Fig. 2(c)], the front is pushed back by the wind, as viewed in the comoving (vortex) reference frame. In the laboratory reference frame



FIG. 2. Sequences of decurled images, showing regimes of front propagation. In all cases, the imposed wind blows from right to left. (a) Forward-propagating front; $\epsilon = 0$ and $\mu = 40$. The front is advected around the vortex and burns across the separatrix in order to propagate forward. (b) Frozen front; $\epsilon = 2.6$ and $\mu = 12$. The wind prevents the reaction from burning across the separatrix. (c) Backward-propagating front; $\epsilon = 8.6$ and $\mu = 12$. The front is initially in the leading vortex but is blown backwards by the wind. (d) Frozen front for a narrow random array of vortices; $\epsilon = 4.0$ and $\mu = 12$.

the front may still move forward, but not as fast as the vortices.

Front velocities in the comoving reference frame are plotted as a function of the opposing wind speed in Fig. 3 for three values of μ , along with the prediction from Eq. (1) for the $\mu = 0$ limit [23]. The most salient feature of Fig. 3 is the plateau, clearly visible for all three values of μ , where the front velocity $\nu = 0$. The width of the frozen-front plateau decreases with decreasing μ and the velocities approach the theoretical limit as μ approaches 0. Also note that advection due to the vortices enhances the front speeds when $\epsilon < 1$, consistent with previous studies of the $\epsilon = 0$ limit [21,24,25].

The variation with μ of the width of the pinned-front regime can be seen in a parameter-space diagram (Fig. 4). The minimum wind speed to achieve frozen fronts is



FIG. 3. Front velocities in the face of opposing wind. The data shown are for $\mu = 4$, 12, and 40 (open diamonds, filled circles, and stars, respectively) along with the theoretical limit [Eq. (1)] for $\mu = 0$ (dashed line). The inset shows the same data for a narrow random chain of vortices at $\mu = 12$. For both graphs, error bars are not visible for the frozen ($\nu = 0$) cases, since it is very easy to identify a frozen front with a high degree of certainty. A frozen front circles within the leading vortex but is pushed back from the hyperbolic fixed point.

(within error) the RD front velocity: $\epsilon = 1$. This can be understood by considering the behavior near the separatrices, where the wind is perpendicular to the underlying vortex flow and where forward propagation is not aided by advection. If ϵ exceeds 1, the wind is stronger than the forward-burning RD velocity, and the front stalls at the separatrix. The end point of the plateau is significantly below $\mu + 1$ (i.e., the sum of the RD velocity and the advective velocity), and it diverges from this line in a nonlinear fashion. We are currently investigating this nonlinear behavior, particularly in light of secondary flows [26] and no-slip boundary conditions in the experiments.

Frozen fronts are not limited to ordered chains of vortices; our results extend to random, 2D vortex arrays, which better approximate 2D flows found in nature. Frozen fronts in narrow random vortex flows are shown in Fig. 2(d) and the inset of Fig. 3. The freezing mechanism remains the same—the front is pinned at the (more convoluted) separatrices. The minimum ϵ necessary for frozen fronts is greater than 1, however. The separatrices are not typically perpendicular to the wind for random flows; as a result, higher W is required for the perpendicular component of the wind to exceed v_0 and thus prevent the front from burning across a separatrix.

Freezing of fronts is even more general; it is not restricted to flows with a limited number of vortices, nor is it dependent on no-slip boundary conditions in a confined geometry. A frozen front is shown in Fig. 5 for a significantly wider annulus (inner and outer radii 2.6 and 8.3 cm), along with a streak photograph showing the underlying disordered pattern of vortices. Because of the annular geometry, we can no longer approximate a uniform wind;



FIG. 4. Parameter-space diagram, showing the increase in the range of the frozen-front regime with increasing μ . Open triangles denote forward propagation of the front against the wind, closed diamonds represent pinned fronts, and open circles denote backward (downwind) front propagation. The solid line shows the boundary between pinned and unpinned fronts. The error bars show the uncertainty in μ and are representative for all points with the same value of μ . The dashed line is at $\epsilon = \mu + 1$, the normalized sum of the RD and advective velocities.

W grows by a factor of 3 between the inner and outer edges. Despite this significant variation in W fronts freeze across the entire annulus, a further indication of the robustness of this phenomenon. As seen in Fig. 5(b), the frozen front generally follows the separatrices, although it is pushed back slightly from the edge of the vortices at larger radii due to the increased wind.



FIG. 5. Frozen front in a wide, 2D, random array of vortices. The images are decurled with the inner and outer radii at the bottom and top of the image, respectively. (a) Image of the frozen reaction front. The normalized wind ϵ ranges from 1.8 to 5.8 from the bottom to the top of each image. (b) Streak photograph of the underlying vortex flow with $\epsilon = 0$. The front from (a) (black curve) is superimposed on the section of the flow where it freezes. (c) Snapshots of frozen front.

Our experiments indicate that pinning of reaction fronts should occur in a wide range of steady 2D vortex flows, regardless of the spatial pattern of the vortices. Furthermore, these results help interpret and predict front behavior in time *-dependent* flows. We can use front pinning, for example, to explain mode locking in an oscillating vortex flow [19,20]. In a comoving (i.e., oscillating) reference frame, the vortices are stationary in the presence of an oscillating wind. For sufficiently large oscillatory velocity, the wind pins the front to a vortex during a significant fraction of each oscillation period. Thus, the front can only propagate during a well-defined segment of each period, tying the front propagation speed to the oscillation frequency.

Moving vortices in other 2D, time-dependent flows can be expected to pin reaction fronts similarly, if only temporarily, in a manner that fundamentally alters the speed and method of front propagation. In our experiments with ordered vortex flows, fronts pin in a single vortex in the chain. Consequently, pinning in general should be possible with only one vortex. Consider the case of a vortex and a front moving in the same direction. If the vortex passes through the front with a sufficient speed, the front will pin and be dragged forward [27]. (In the reference frame of the vortex, this is equivalent to the frozen-front case.) Even if the vortex later slows down or speeds up such that it can no longer hold the front, it will have already significantly altered the shape and location of the front. Transient and long-lived coherent vortices are often found even in 2D turbulent flows [28-30] and may temporarily pin fronts as they move through the fluid. These predictions are being tested in ongoing experiments that we are conducting on the role of vortices in front propagation in time-dependent flows. Ultimately, a general theory is needed of front propagation in ARD systems that accounts for pinning of fronts by vortices in the flow.

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