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Introduction to the Focus Issue: Chemo-Hydrodynamic Patterns and Instabilities

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Pattern forming instabilities are often encountered in a wide variety of natural phenomena and technological applications, from self-organization in biological and chemical systems to oceanic or atmospheric circulation and heat and mass transport processes in engineering systems. Spatio-temporal structures are ubiquitous in hydrodynamics where numerous different convective instabilities generate pattern formation and complex spatiotemporal dynamics, which have been much studied both theoretically and experimentally. In parallel, reaction-diffusion processes provide another large family of pattern forming instabilities and spatio-temporal structures which have been analyzed for several decades. At the intersection of these two fields, "chemo-hydrodynamic patterns and instabilities" resulting from the coupling of hydrodynamic and reaction-diffusion processes have been less studied. The exploration of the new instability and symmetry-breaking scenarios emerging from the interplay between chemical reactions, diffusion and convective motions is a burgeoning field in which numerous exciting problems have emerged during the last few years. These problems range from fingering instabilities of chemical fronts and reactive fluid-fluid interfaces to the dynamics of reaction-diffusion systems in the presence of chaotic mixing. The questions to be addressed are at the interface of hydrodynamics, chemistry, engineering or environmental sciences to name a few and, as a consequence, they have started to draw the attention of several communities including both the nonlinear chemical dynamics and hydrodynamics communities. The collection of papers gathered in this Focus Issue sheds new light on a wide range of phenomena in the general area of chemo-hydrodynamic patterns and instabilities. It also serves as an overview of the current research and state-of-the-art in the field. *© 2012 American Institute of Physics*.
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Chemo-hydrodynamic patterns and instabilities appear in out of equilibrium systems when chemical reactions and diffusion interplay with advection or convection processes. Complex spatio-temporal evolution of concentrations can then set in due to the highly nonlinear character of the underlying dynamics. In this Focus issue, 13 articles discuss recent advances in the area of chemo-hydrodynamic patterns and instabilities. These articles are representative of three classes of problems in which chemo-hydrodynamic structures are commonly observed and for which simpler model systems can help unraveling key aspect of the dynamics.

I. INTRODUCTION

Pattern formation is part of the general class of nonlinear dynamics problems, which deals with the study of nonlinear differential equations and mappings, experimental model systems and the resulting complex spatiotemporal behavior, and holds a strategic position in both pure and applied sciences. Pattern forming instabilities in hydrodynamic systems often arise from the action of a driving force on the flow, such as gravity, or gradients in surface tension also referred to as thermal/solutal Marangoni effect depending on their origin. On the other hand, pattern formation in reaction-diffusion systems can result from the coupling between diffusion and nonlinear kinetics.

Hydrodynamic and reaction-diffusion systems might appear at first to be seemingly unrelated. However, in many important cases the coupling between fluid flows and chemical reactions, via, e.g., entrainment by a flow or dependencies of physical properties of the fluid on the concentration of chemicals, come to the fore and cannot be neglected. This interdependence can result in new and unexpected dynamics and pattern forming instabilities.

Two distinct situations exist for chemo-hydrodynamic patterns and instabilities. The reaction is active, i.e., it modifies dynamically the flow and in this case the corresponding fluid motion will be referred to as *convection*. Alternatively, the reaction is not affecting the flow and the chemicals are passively entrained by the flow, in which case the fluid motion will be referred to as *advection*. But even when the chemical species behave as passive scalars, their spatio-temporal distribution can nevertheless become quite complex. The evolution of the concentrations then follows *reaction-diffusion-advection*

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(RDA) equations and is strongly dependent on the properties of the imposed flow. The yield of the reaction is hence affected in a complicated fashion by the advective changes in the mixing of the reactants. Applications are numerous in fields as diverse as, for instance, combustion, astrophysics, oceanography, climatology, ecology, and of course engineering.

On the contrary, active chemistry can result when gradients in concentrations induce changes in a physical property of the fluids at hand, like their density, viscosity or surface tension. In this case, reaction-diffusion processes can induce or modify hydrodynamic flows which, in turn, will themselves modify the spatio-temporal distribution of the chemicals and the yield of the reactions. A highly nonlinear feedback then sets in between the chemical reactions and the corresponding diffusive and convective transport phenomena. The study of the associated *reaction-diffusion-convection* (RDC) patterns and instabilities is of interest, both from a fundamental point of view and with respect to their potential applications in a wide variety of natural phenomena and technological applications.

Not surprisingly, the resulting chemo-hydrodynamic patterns can be quite complex as several sources of instabilities can interplay with each other. As an example, for exothermic reactions, both compositional and thermal gradients can induce changes in density and/or viscosity triggering buoyancy-driven convection and/or viscous-driven flows. If, in addition, an interface between two immiscible fluids is present, like it is typically the case for solutions in contact with air, Marangoni-driven flows triggered by surface-tension gradients may develop. Clearly, to gain fundamental insight in such complex dynamics, the study of simpler model systems is thus necessary.

A first class of model systems ruled by RDA equations focuses on the transport of passive scalars, such as temperature or concentration, which are assumed to have no influence on the flow field. In simple cases, heat/mass is transported through unidirectional flow fields. However, effective diffusion can then already be significantly enhanced even when a small convective flow is generated in a direction normal to the originally parallel flow. For example, mass transport across a laminar falling film can increase by as much as a factor of 10 when waves appear at the interface[1] owing to the presence of a recirculation zone within the solitary waves that dominate the large-time evolution of the surface of the film.[2] Indeed, flow-assisted diffusion of heat/mass has a profound effect on a large variety of heat/mass-transport applications and understanding the enhancement mechanism is a necessary step for the development of improved transport and separation processes, crucial, e.g., in chemical engineering.

The problem of evaluating transport of passive scalars in a flow becomes rather involved in the presence of diffusive boundary layers occurring when advection dominates over diffusion (typically such layers appear in the vicinity of rigid boundaries or mobile interfaces) and/or chemical reactions in the flow.[3,4] In the latter case in particular, reaction conversion and selectivity depend largely on the topology of the flow field and dead reaction zones in the flow can easily develop, even though stirring of the flow might be vigorous. At the same time, additional complexities associated with imperfect mixing[5] and the chaotic or turbulent nature of the flow make the analysis of such systems quite involved. The so-called lamellar structures, in particular, within the microscales of a chaotic or turbulent flow (the statistical similarity of which is now well-established) largely influence chemical reactions in the flow. The prototype system here consists of one-dimensional lamellar systems where chemical reactions, e.g., of the type $A + B \rightarrow C$ (Refs. 6 and 7) or two-step (competitive-consecutive) chemical reactions,[8] take place.

A second class of model systems encompasses the so-called reaction-diffusion fronts and waves, i.e., self-organized dissipative miscible interfaces resulting from the coupling between reactions and diffusion processes. Autocatalytic chemical reactions in which one product of the reaction feedbacks onto its own production can provide reaction-diffusion fronts corresponding to self-organized interfaces separating reactants and products and traveling at a constant speed. Such propagating fronts provide an ideal model system to study RDC chemo-hydrodynamic patterns as they genuinely create internal concentration and temperature gradients and thus possible hydrodynamic instabilities. Indeed, the chemical reaction generates not only a self-organized interface between the products and the reactants when coupled to diffusion but may also induce a density, viscosity or surface tension jump between the solutions.

The influence of convective hydrodynamic currents on such miscible interfaces has long been recognized to yield acceleration and deformation of autocatalytic fronts and waves.[9–17] For $A + B \rightarrow C$ fronts, complex buoyancy-[18,19] or viscosity-driven patterns[20–22] can develop when miscible solutions containing initially separated A and B reactants are put into contact with each other. These convective patterns have been shown to have stability properties[21,22] and symmetries[18] different from those in non-reactive systems.

Finally, a third class of model systems consists of immiscible RDC fluid-fluid systems in the presence of species which are either restricted to the fluid-fluid interface or can cross the interface, and which may also undergo chemical reactions. Such systems create some sort of mass-transport-driven analogue to the thermally-driven Rayleigh-Bénard and Bénard-Marangoni convection, a paradigm of pattern formation. The pioneering work of Sternling and Scriven,[23] first for mass transport without reaction, elucidates the emergence of Marangoni-driven instabilities in such systems. Due to their accelerating effect on extraction processes, these instabilities have attracted substantial interest in engineering for many years and provide a wide variety of dissipative structures.

The fundamental insights gained from these mass-transport-driven systems form an important basis to analyze the more complex situation of coupling of mass transport with reactions as encountered for instance in the chemical industry related to reactive extraction or phase transfer catalysis. In these processes, surfactants are often involved, e.g., as carriers and emulsifiers. Resulting convective motions have been studied both experimentally[19,24–30] and theoretically.[31–36] A related set of problems involves fluid-fluid systems with combined mass/heat transport and reactions which induce a thermal/solutal Marangoni effect, frequently encountered in chemical engineering, e.g., reactive falling films[37,38] or thin-films in the presence of reactive surfactants.[39,40] Of similar importance are "droplet" geometries, which are relevant in the form of bubble columns in chemical reactors or in the rapidly growing field of microfluidics. Chemical reactions between the liquid of the droplet and the underlying substrate may produce self-running droplets.[41–43] Another form of self-propulsion may result from the Marangoni effect when a chemical reaction in the solution leads to inhomogeneities in the surface tension.[44]

This Focus issue compiles a selection of representative articles in the above three classes, namely, passive scalars and transport, convective motions around reaction-diffusion fronts, and hydrodynamic instabilities at fluid-fluid interfaces. Our hope is that, without being exhaustive, it nevertheless captures the spirit of chemo-hydrodynamic patterns and instabilities and its current state-of-the-art through a collection of thirteen research articles by leading researchers on various aspects of this field.

A. Overview of contributions to the Focus Issue

1. Passive scalars and transport

In the first paper of this section, Neufeld[45] gives an overview, and at the same time extends previous results, on the effects of large-scale oceanic transport processes on plankton population dynamics, by considering different types of ecosystem models. By using the simplest examples of population models relevant to plankton, it is shown that increasing stirring rate in an environment where the carrying capacity is non-uniformly distributed leads to an overall decrease of the effective carrying capacity of the system.

In the next paper, Bargteil and Solomon[46] present a series of experiments on reactive front propagation in a vortex chain flow and a spatially disordered vortex-dominated flow. The flows are driven by magnetohydrodynamic effects, and the fronts are produced using the excitable Belousov-Zhaotinsky (BZ) reaction. In both of these flows, front propagation is dominated by the presence of burning invariant manifolds (BIMs) that act as barriers, similar to invariant manifolds that dominate the transport of passive scalars. Convergence of the fronts onto these BIMs is shown experimentally for all of the flows examined.

In the companion paper by Mitchell and Mahoney,[47] the underlying theory of the BIMs is developed and their role in the description of front propagation in laminar flows is analyzed. Criteria for the existence of the BIMs are offered together with classification of their stability and analysis of their barrier property as well as mechanisms by which the barriers can be circumvented. The analysis is based on the sharp-front limit with negligible feedback of the front on the fluid velocity together with a (low-dimensional) dynamical systems approach.

2. Convection around self-organized reaction fronts

Pópity-Tóth *et al.*[48] investigate how buoyancy affects the spatiotemporal evolution of exothermic autocatalytic fronts in horizontal three-dimensional covered solution layers of various thicknesses. They characterize in detail the influence of the height of the solution layer on the resulting deformation of the front. Simultaneous monitoring from the top and the side of the reactor reveals that the resulting three-dimensional front structure has a smooth leading edge independent of the solution thickness and that the structured trailing edge ends in a center cusp with a constant angle.

If similar exothermic autocatalytic fronts travel in thin horizontal solutions now open to the air, additional Marangoni flows due to gradients of surface tension can set in. To understand the specific role of such Marangoni effects, Rongy *et al.*[49] numerically investigate the possible dynamics of the fronts which can result from combined pure solutal and thermal Marangoni effects in the absence of gravity. To this end, they numerically integrate the incompressible Navier-Stokes equations coupled through the tangential stress balance to evolution equations for the concentration of the autocatalytic product and for the temperature. They show that complex oscillating regimes can be obtained when solutal and thermal Marangoni effects act in opposite direction across the front.

In a vertically oriented set-up, Levitán and D'Onofrio[50] analyze experimentally to what extent the stability properties of autocatalytic fronts with regard to a buoyancy-induced Rayleigh-Taylor instability are modified when the mean temperature of the reactor is varied. The authors demonstrate that when the temperature is increased, the density fingering of an autocatalytic front induced by unfavorable density gradients is stabilized. Above a critical temperature, the front remains stable and thus planar.

In a combination of experiments and Lattice-Boltzmann based simulations, Atis *et al.*[51] complement the foregoing works by two new elements. First, they study the interaction of autocatalytic fronts with porous media. They demonstrate how, e.g., the tortuosity of the porous media, which describes the loss in efficiency of diffusion, affects the velocity of the chemical front. Second, the authors analyze the modification of velocity and shape of the front in the presence of forced advection in analogy with the works in Sec. I A 1. They show that even steady fronts with vanishing velocity can be obtained when the direction of the Poiseuille flow is opposite to that of the chemical front. A model experiment to illustrate the origin of this behaviour is discussed.

The first three papers of Sec. I A 2 deal with only one source of convection: either buoyancy or surface tension gradients. The paper by Rossi *et al.*[52] couples both effects as it is concerned with the dynamics of traveling waves under the combined effect of both Marangoni- and buoyancy-driven flows. Such an interaction can generate a rather rich spectrum of patterns, including stationary/cellular structures and chaotic wave trains. Here, the authors demonstrate, both experimentally and theoretically, the appearance of segmented wavy structures in a shallow layer of an excitable BZ solution. The segmentation phenomena were found to depend both on the depth of the solution and on the excitability of the reaction.

Extension of the study of buoyancy effects to three-dimensional systems is provided by Rogers and Morris[53] who numerically investigate the buoyancy-driven deformation of autocatalytic reaction fronts initialized in spherical configurations. Depending on the initial radius of these "autocatalytic balls," the three-dimensional front will develop in one of three different ways. In an intermediate range of ball size, the flow can evolve much like an autocatalytic plume: the ball develops a reacting head and tail that is akin to the head and conduit of an autocatalytic plume. In the limit of large autocatalytic balls, however, growth of a reacting tail is suppressed and the resemblance to plumes disappears. Conversely, very small balls of product solution fail to initiate sustained fronts and eventually disappear.

3. Chemo-hydrodynamic instabilities at fluid-fluid interfaces

Miyaoka *et al.*[54] study an oil-water system at the interface of which the formation of a complex between an anionic surfactant and metal ions takes place. A spontaneous irregular Marangoni flow, associated with temporal variation of the interfacial tension, is observed only for a limited number of metal ions, hence an ion-sensitivity occurs. The authors provide evidence that the resulting Marangoni instability is a consequence of the collective desorption of aggregated complexes, referred to as an exfoliation.

In contrast to the irregular Marangoni motions of Ref. 54, a well-ordered pattern of chemo-Marangoni cells is reported by Eckert *et al.*[55] again in an oil-water system with an *in-situ* production of surfactants by an interfacial reaction. The authors demonstrate that the transition towards periodic large-scale interfacial deformations, which emerge from the stagnation points of the chemo-Marangoni cells, is associated with an autocatalytic surfactant production after the critical micellar concentration is approached.

The systems studied in both works pose a challenge for theoretical modelling since the surfactant dynamics require a separate transport equation at the interface. To formulate this equation, a better understanding of the intricate desorption processes is mandatory in order to specify the corresponding rate terms. Furthermore, the surface activity of the surfactant remains not constant but might change due to both the reaction and, correspondingly, the change of the *p*H value. This introduces further nonlinearities into the boundary conditions, in particular into the stress balance.

For gas-liquid systems, Andres and Cardoso[56] analyze numerically the nonlinear interactions between convective fingering and a chemical reaction in a diffusive boundary layer in a porous medium when CO_2 dissolves in a liquid layer. The reaction enhances here stability by consuming the dissolved CO_2 that is unstably distributed in a gravitational field. Nonlinear simulations show that the chemical reaction can profoundly change the dynamics of the system, by introducing a steady state, shortening the evolution time, and altering the spatial patterns of velocity and concentration of the solute. For the weakly reactive case, finger growth and merger occur driving strong convective currents in a thick layer of the solute. However, as the reaction becomes stronger, finger growth is inhibited, tip-splitting is observed, and the layer of solute becomes much thinner.

An example of viscous-driven convective flows developing in the presence of an $A + B \rightarrow C$ type of reaction can be found in the contribution by White and Ward.[57] In analogy to Ref. 19, their study is performed in a horizontal Hele-Shaw cell where finite volume of aqueous $Ca(OH)_2$ is displaced by gaseous CO_2. The liquid-gas interface is deformed due to a Saffman-Taylor instability. The paper analyzes the conditions for an efficient reactive conversion of CO_2 into $CaCO_3$ precipitate by balancing the constraints, namely chemical reaction dynamics, which benefits from high pressures due to better dissolution of the gas, and the fluid displacement itself where higher pressures reduce the contact time of CO_2 and liquid and produce thicker liquid films at the Hele-Shaw cell walls through which CO_2 has to diffuse.

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