

FRONT PROPAGATION IN LAMINAR CELLULAR FLOWS : AN EXPERIMENTAL STUDY

Alain Pocheau, Fabien Harambat

IRPHE, Universités Aix-Marseille I & II, 49 rue Joliot-Curie, B.P. 146, F-13384 Marseille, France

Summary Front propagation in different kinds of laminar flows is investigated experimentally. Images of front propagation as well as data on their mean velocity show that a kinematic model of propagation is more relevant here than models based on homogenization procedures.

This work is devoted to study experimentally the propagation of reaction fronts in laminar flows. This issue may concern chemical reactions in microfluidic or biological environments, chemical engineering, combustion, and more generally propagation of reactions in poorly stirred environment as found in populations dynamics or in planetary sciences (e.g. dynamics of the ozone hole) for instance. Our main objective is to model this issue in order to identify the most relevant picture of the corresponding process and its most valuable modelization. For this, we have designed different experiments in which a front propagates in a liquid medium that involves simple prescribed steady flows. The difference between experiments relies on the geometry of the flows. The comparison between the resulting front behaviour will be used to identify the salient features of the process.

EXPERIMENTAL CONFIGURATION

The experimental configuration is designed so as to study front propagation in flows created either by thermal means (i.e. thermoconvection) or by electromagnetic means (i.e. electroconvection). For this the reaction must take place in an aqueous solution [1]. We have chosen to work here on the chlorite-iodide reaction [2]. This autocatalytic oxydo-reduction reaction yields iodide I^- to be oxidized in iodine I_2 . Addition of a starch indicator makes the presence of iodide visible as a blue colour. Thus, oxydation of iodine as the wave passes through leaves a colourless medium whose frontier with the blue medium localizes the front. Front is initiated by contact with an electrode. Its velocity is about $U_L \approx 2mm/mn$ at $ph=5$. It varies with temperature or electric current in a way which has been monitored.

From the images and the velocity of the front, we have deduced its thickness $\delta \approx 1mm$, the reaction time $\tau_r \approx 6s$, and the form of the reaction term $f(\theta)$ of the corresponding 1-d reaction-diffusion equation $\partial_t \theta = D \partial_x^2 \theta + f(\theta)/\tau_r$. This term takes a polynomial shape $|f(\theta)| = \theta(1 - \theta)(1 - 0.39\theta)$ which corresponds to a KPP wave.

The convective flows are generated by electrical heating from below and by cooling from above by water circulation. To allow front visualization, an optical access is made in the cooling device and a mirror is fixed in between the fluid and the heating plate. The depth of the convective layer is $d = 5.8mm$, its width is $l = 6mm$ and its length $X = 20cm$. This yields a chain of about 30 steady rolls to appear beyond onset. The flow intensity U depends on the distance to onset. However, to accurately determine it, care has been taken of the finite thermal conductivities of the boundary plates and of the temperature variation of water physical properties. The range of flow intensity scanned was : $12 < U < 60mm/mn$. The electroconvective flows are produced by generating a current along a similar channel below which a chain of magnets, $2 * 2cm$ wide, has been placed with alternating directions of their magnetic field. The flow intensity $3 < U < 35mm/mn$ is proportionnal to the current intensity, the layer depth is $d = 3mm$, its width is $l = 12mm$ and its length is $X = 20cm$. According to the above values, our experiment involves high Péclet numbers $Pe = UL/D > 600$ where L corresponds to the vortex size ($L \approx d$ in convection and $L = 20mm$ in electroconvection). On the other hand, the Damköhler number $Da = L/U\tau_r$ is : $2 < Da < 7$ in convection ; $5 < Da < 48$ in electroconvection. This means that advection dominates diffusion but that advection and reaction have similar importances.

OBSERVATIONS AND RESULTS

Either in convective flows or in electroconvective flows, the front was rolled up by vortices, but it always succeeded in keeping its structure locally unchanged (Fig.1). In particular, no increase in front thickness has been noticed. Once contaminated by the fronts, the vortices were "burnt" at a speed roughly equal to the laminar velocity U_L . This is easily understood since perpendicularly to the flow streamlines, no advection can help the front to propagate.

The interesting phenomena come from the contamination of vortices. It arises from the "burnt" medium adjacent to the still "fresh" vortex, near the separatrix between both (Fig.1). This contamination sets the overall reaction rate since, once established, the vortex will burn in a prescribed time. It appears that the contamination is quick and roughly proportionnal to the flow intensity U . It then yields a mean relative front velocity U_f/U_L nearly proportionnal to the relative flow intensity U/U_L for both the convective and the electroconvective case (Fig.2).

The similarity between the propagation in the two kinds of flows stops here. In particular, the paths followed by the front inside a vortex and when crossing the separatrix with its neighbor are different in both flows. It is actually more 3-dimensionnal with convective flows than with electroconvective flows (Fig.1). In the former case, the front directly invades the middle of the vortex from the separatrix ; in the latter case, it gently rolls along the boundaries before uniformly propagating towards the center. This spatial difference is surprising since both flows are *a priori* designed to be 2-d

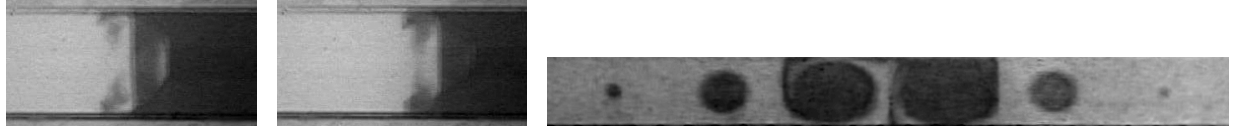


Figure 1. Images of front propagation in convective flows (left, view normal to the roll axis) and electroconvective flows (right, view along the roll axis).

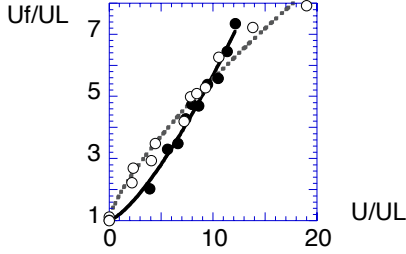


Figure 2. Relative increase of front velocity U_f/U_L as a function of the relative flow intensity U/U_L in convective flows (full circles) and in electroconvective flows (open circles).

cellular flows. We attribute it to the secondary flows (similar to Rossby flows) that develop when the vortex boundary layers separate one from the other. This occurs sooner in the convective case due to a weaker relative confinement of vortices on their axis direction. Beyond this, convection becomes quickly more efficient in increasing front propagation than electroconvection (Fig.2). This stresses the importance of the fine details of flow geometry on front propagation.

MODELS AND INTERPRETATION

The models of front propagation in a stirred medium proposed in literature rely either on the advection-reaction-diffusion-equation or on the G-equation (which describes the eikonal dynamics of a self-propagating wave surface in an advecting flow) [3, 4]. Most of them seek to determine the resulting mean front velocity U_f as the propagation velocity of an effective reaction-diffusion wave with renormalized diffusion coefficient D' and reaction time τ_r' : $U_f = (D'/\tau_r')^{1/2}$. The way these renormalized variables vary with U and U_L sets the final dependence of the mean front velocity with the flow intensity, i.e. at large Pe : $U_f \approx U^{1/4}$ for $Da \ll 1$ and $U_f \approx U^{3/4}$ for $Da \gg 1$.

None of these tendencies are recovered in our experiments, whatever the kind of flows. Moreover, direct observation of the way fronts propagate in the cellular structure reveals the essential role played by their leading edge, in contrast with the above wave picture. In particular, the mean propagation velocity is actually set by the rate of propagation of the *most advanced part* of the front, whatever the state of contamination behind it. This statement asks for viewing the process of front propagation more as the propagation of local objects (i.e. "particles") than as the propagation of a global object (i.e. a "wave"). The implications on the modelization is to turn attention to kinematic models (e.g. [5]) treating the front as an assembly of local parts, the quickest of them setting the average propagation speed in the system.

We have derived such a kinematic model for electroconvective flows and obtained a good agreement. We have not yet achieved it for the convective case because the geometry of their flows (especially their secondary flows) deserves a special attention. However, we have looked to another kind of flows produced by alternating supercritical zones and subcritical zones of convection, i.e. by alternating convection and conduction. Here too, a kinematic model based on flow geometry enabled us to recover the experimental data.

CONCLUSION

Looking experimentally to front propagation in different cellular flows at moderate Damköhler numbers led us to conclude, both from qualitative and quantitative observations, that a kinematic modelization is much more relevant than a homogenization procedure. This means that fronts behave, in average, not as a wave but as a set of dynamically independent parts, the mean velocity being set by the fastest one. Following this, front propagation in laminar flows appears as an optimization problem instead of an homogenization problem.

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