

## Steady Convective Flows Traveling with Chemical Fronts

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Laurence Rongy est licenciée en Sciences Chimiques de l'Université Libre de Bruxelles depuis 2004. Son mémoire de fin d'études a porté sur l'étude théorique des fronts chimiques se propageant en présence d'effets Marangoni et a obtenu le prix annuel de la Société Royale de Chimie. Depuis octobre 2004, elle réalise une thèse sous la direction du Dr Anne De Wit dans l'Unité de Chimie Physique Non Linéaire de l'ULB grâce à une bourse d'Aspirant F.N.R.S. L'objet de sa thèse est l'étude théorique du couplage entre réactions chimiques et phénomènes convectifs induits par la chimie.

### Abstract

Chemical reactions taking place in non stirred solutions can initiate spontaneous convective motions, due to density and surface tension gradients, which in turn can affect the chemical reaction. We study this interaction with model chemical fronts showing that a convection-driven steady fluid vortex can strongly deform the front and speed up the reaction.

### Keyword

Chemical front, autocatalytic reaction, Marangoni convection, buoyancy convection

### 1. Introduction

As soon as a chemical reaction occurs in a non stirred solution, convection can be triggered by density or surface tension gradients induced by concentration or temperature gradients. An intricate interaction between chemical and transport phenomena then determines the system dynamics. A simple class of model systems used to study this interaction consists in chemical fronts resulting from the coupling between autocatalytic reactions and diffusion. Indeed, when they propagate across a solution, invading the reactants and leaving the products behind them, they give rise to an interface between two solutions with different composition and temperature. Such concentration and temperature gradients across the front are then prone to initiate convective flows, which have been shown experimentally to modify front properties drastically [1,2]. However, it is not easy to distinguish Marangoni (surface tension-driven) from buoyancy (density-driven) convection. In this context, theoretical

approaches are crucial to analyze model systems where only one type of convective flow is active. Our objective is to theoretically characterize the dynamics of a model chemical front propagating in the presence of respectively pure Marangoni convection and pure buoyancy convection, in the absence of thermal effects, to shed light into complex spatio-temporal dynamics that can exist in unstirred reactive solutions.

### 2. Physical model

Our model system is an isothermal planar chemical front propagating in a two-dimensional (2D) thin aqueous solution layer corresponding to a vertical cut in a Petri dish open to the air. The surface tension and the density of the products are different from those of the reactants and we assume no surface deformation and no evaporation. To analyze this system, we numerically integrate the incompressible Navier-Stokes (N-S) equations for the velocity of the fluid coupled to a reaction-diffusion-convection model describing the evolution of the product concentration in space and time. The chemical kinetics models a general autocatalytic reaction and the buoyancy term in the N-S equations depends on a Rayleigh number,  $Ra$ , quantifying the density changes induced by the reaction. Positive and negative  $Ra$  correspond respectively to the cases where the products are lighter or heavier than the reactants. A zero Rayleigh number is pertaining to a solution density not affected by the chemical reaction. In the same way, the surface tension changes induced by the reaction are quantified by a Marangoni number  $M$  appearing in the boundary condition at the open air-layer interface. Positive and negative  $M$  correspond respectively to the cases where the product decreases or increases surface tension.

### 3. 2D flows around chemical fronts: nonlinear dynamics

Our numerical work has allowed analyzing the dynamics of a chemical front in the presence of respectively pure Marangoni ( $Ra = 0$ ,  $M \neq 0$ ) or pure buoyancy convection ( $M = 0$ ,  $Ra \neq 0$ ).

#### 3.1. Marangoni flows traveling with chemical fronts

Marangoni convection corresponds to a fluid motion initiated at the surface of a fluid towards increasing surface tension. Therefore the fluid motion is initiated at the surface towards the reactants for positive  $M$  and towards the products for negative  $M$ . In both cases, a bulk motion is induced in the opposite direction and the system reaches an asymptotic dynamics characterized by a localized steady fluid vortex traveling at a constant speed with the front and deforming it [3]. This asymptotic dynamics is presented in Fig.1 by a 2D density plot of the product concentration from  $c = 1$  (products in red) to  $c = 0$  (reactants in blue) for  $M = 100$ . For negative  $M$ , the convection roll turns anticlockwise and the front is deformed in the opposite direction. For a same absolute value of  $M$ , the front deformation and propagation speed are larger for positive  $M$ . This difference is due to the fact that the flow induced at the surface is respectively parallel ( $M > 0$ ) or antiparallel ( $M < 0$ ) to the direction of front propagation. However, in both cases, the front deformation and propagation speed are larger than without convection, and increases with  $|M|$ .

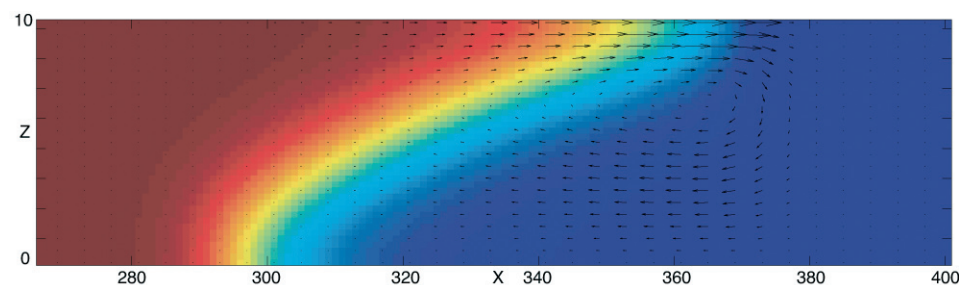


Figure 1. Focus on the Marangoni convection roll traveling with the deformed front for  $M = 100$ .

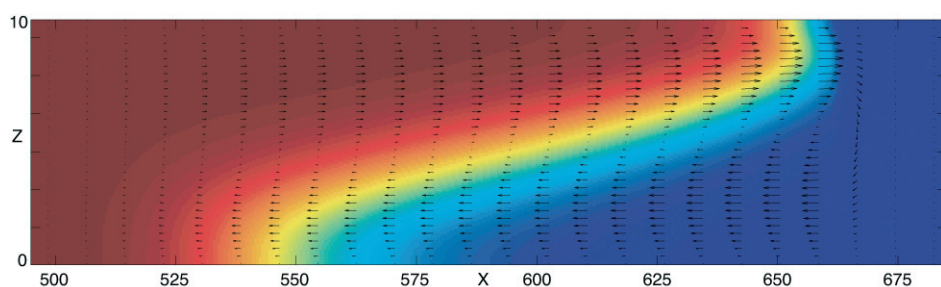


Figure 2. Focus on the buoyancy convection roll traveling with the deformed front for  $Ra = 100$ .

#### 3.2. Buoyancy flows traveling with chemical fronts

For pure density effects, the system also reaches an asymptotic dynamics with a localized fluid vortex stretching and accelerating the front (see Fig.2) [4]. The heavier reactants sink below the products that propagate faster in the upper part of the layer. The situation for negative  $Ra$  is the symmetric of positive  $Ra$  with regard to the middle of the layer thickness. The front deformation and propagation speed both increase with  $Ra$ , and smaller values of  $Ra$  than  $M$  are needed to achieve a same deformation and propagation speed.

### Conclusions

When chemical reactions occur in non stirred solutions, they can generate spontaneous convection, which in turn affects the spatio-temporal evolution of concentrations. We have shown the important effects of convection on a model autocatalytic chemical front system, pointing out that the front is strongly deformed in the layer thickness and propagates faster than without convection, increasing hereby the reaction efficiency. Interesting perspectives would be to include the reaction thermal effects and to see how both types of convection can interact in a chemical system to help understand complex dynamics observed experimentally in unstirred reactive solutions.

### Acknowledgments

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