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J. Boissonade. Stationary structure induced along a reaction-diffusion front by a Turing symmetry breaking instability. Journal de Physique, 1988, 49 (3), pp.541-546. 10.1051/jphys:01988004903054100 . jpa-00210727

HAL Id: jpa-00210727

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Submitted on 1 Jan 1988

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Classification
Physics Abstracts
82.40 — 05.90 — 66.10

Stationary structure induced along a reaction-diffusion front by a Turing symmetry breaking instability

J. Boissonade

Centre de Recherche Paul Pascal, Université de Bordeaux I, Domaine universitaire, 33405 Talence Cedex, France

(Reçu le 26 octobre 1987, accepté le 25 novembre 1987)

Résumé. — Nous montrons qu'avec une géométrie de réacteur appropriée, une structure de Turing bidimensionnelle de réaction-diffusion peut être ramenée à une structure quasi-unidimensionnelle localisée le long d'un front de réaction. Nous donnons un exemple obtenu par simulation numérique. La géométrie proposée, qui permet de s'affranchir des contradictions habituellement rencontrées dans le maintien des conditions de non-équilibre, devrait enfin permettre l'observation expérimentale de telles structures.

Abstract. — We show that, with an appropriate reactor geometry, a two dimensional reaction-diffusion Turing pattern can be restricted to a quasi one-dimensional structure along a stationary front and support our conclusions by numerical simulations. Such a geometry should free the experimentalists from contradictory requirements and should allow them to evidence genuine isothermal sustained dissipative structures driven by pure reaction-diffusion processes.

Introduction.

Isothermal chemical systems maintained far from equilibrium by fluxes of matter can exhibit spatial concentration patterns, much the same way other physical systems do under constrained conditions, e.g. the Bénard cells or the Taylor vortices in fluids. Isothermal chemical dissipative structures have been thoroughly studied from a theoretical point of view [1-3]. Until now, most experimental studies have concerned travelling waves including the so-called target and spiral waves patterns [4-6], but very little progress has been made on another type of spatial structure predicted by Turing as early as 1952 [7]. In an initially homogeneous medium symmetry breaking instabilities leading to stationary concentration patterns can result from the competition between reaction and diffusion processes. They involve the same kind of kinetic mechanisms as the oscillating reactions, i.e. autocatalysis or substrate inhibition and arise in particular when the autocatalytic species diffuses slower than the inhibitory species [8]. In spite of their great potential interest for biology and morphogenesis, these patterns have never been clearly observed. In particular most of the transient mosaic structures reported in Petri dishes [9] have been shown to result from convective motions [10].

The major problem for the observation of Turing structures is to sustain the non-equilibrium conditions in a spatially distributed reactor able to meet two apparently contradictory requirements : on one hand one must feed the system with fresh reactants everywhere, on the other hand one must preserve the natural molecular diffusion. The common but unpractical requisite of keeping some concentrations at a uniform constant value is only made for mathematical convenience ; stationary patterns have also been predicted for systems only constrained at the boundaries [11-13]. Here we show that in a system fed this way, a Turing symmetry breaking instability can induce a stationary structure along a reaction front. The achievement of the corresponding experimental set up should be easy. To support our proposal, we produce a numerical example on the basis of a simple model.

1. Reaction front structure : general features.

The chemical system is assumed to be limited to a rectangular two dimensional thin reacting film (Fig. 1). The concentrations of the different species are fixed on the boundaries W1 and W2, parallel to the Ox axis by a continuous stream of fresh chemicals sweeping a permeable membrane. The initial re-

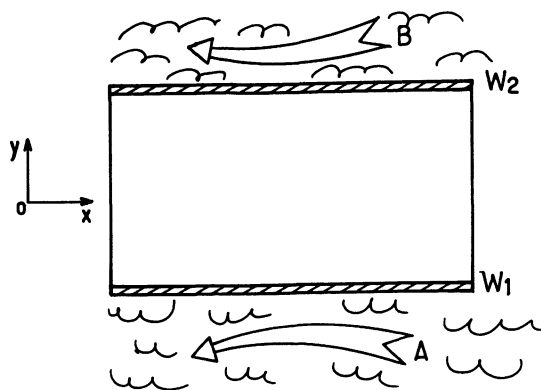


Fig. 1. — Schematic representation of the film reactor.

agents, further referred to as the *major species*, are fed and constrained to nonzero values, different on W_1 and W_2 , whereas those produced by the reaction and referred to as the *intermediate species* are maintained at a zero concentration on W_1 and W_2 . On the orthogonal sides, parallel to Oy , we shall further assume periodic boundary conditions (i.e. identical concentration profile on both sides) but no flux boundary conditions could be used without loss of generality. Inside the film transport is achieved only by molecular diffusion.

To set the ideas down, we consider a reaction between two major species A and B through intermediate species X , Y , etc. The species A and B are respectively fed on W_1 and W_2 . According to the symmetries, the composition changes continuously along the Oy direction whereas the parallels $y = c$ to the Ox axis correspond to isoconcentration curves which could be visualized by appropriate color indicators. The composition changes along Oy are either smooth, or steep and localized in the region where A and B meet. These steep gradients are especially common with high order or autocatalytic reactions, the rate of which tremendously increases when the reagents are simultaneously present in a significant amount. The steep change corresponds to a reaction front which must not be confused with the usual chemical *propagating* fronts found in flames or phase waves.

We expect that if the conditions for a Turing instability are met, this instability can break the symmetry and destabilize the front, generating a structure in the direction Ox . This can be understood on the basis of the following elementary arguments. Many theoretical examples of periodic structures have been produced in one dimensional reaction-diffusion systems where the major species were kept constant and uniform, provided that some intermediate species have different diffusion coefficients. If, at first, we ignore the transverse diffusion of the intermediate species (along Oy) and assume that the consumption of A and B is negligible, an isoconcentration

curve $y = c$ can be considered as such a one dimensional system. The buffering of the major species is performed by diffusive transport from the walls. Thus, for suitable conditions, the homogeneous solution along this curve can become unstable and lead to a periodic structure in the Ox direction. Along the curve $y = c$ the concentrations are no longer constant and the isoconcentration curves should now present a periodic spatial structure in the Ox direction. The steeper are the transverse gradients, the larger should be the amplitude of this spatial modulation. In particular, if the instability conditions are satisfied, one expects a large spatial modulation of the reaction front. On the other hand, close to the walls where the concentrations of intermediate species are constrained to a uniform zero value and where instability conditions are ruled out by the absence of significant chemical activity, there is no structure.

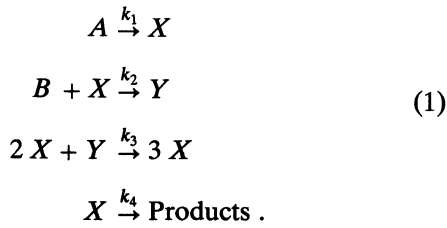
The inhomogeneous profile along the gradient direction Oy could also be destabilized and some longitudinal modes would develop along this direction [12, 13] but they should exhibit a continuous change in wavelength and amplitude due to the imposed gradient. On the contrary, the structure we expect is *transverse* to this gradient, which clearly evidence the symmetry breaking. Turing instability always occurs over a limited set of the major species concentrations. Since we consider steep fronts the largest concentration changes occur within the front. In real autocatalytic reactions, these concentrations can range over four or five orders of magnitude inside this front. Moreover, close to the wall the system is stabilized by the boundary conditions and the low rate of chemistry. Thus the conditions for instability are more likely to be fulfilled solely within the front. The strong anisotropy created by the gradient therefore precludes the formation of a longitudinal mode and the pattern is limited to the transverse mode localized along the front. The problem of the restriction of a Turing structure to a transverse mode under the effect of a strong anisotropy — here the concentration gradient — has recently been addressed by Dewel *et al.* [14] from a general point of view. The effective reduction in dimensionality results in a large decrease in the pattern complexity, which, in some respect, can be compared with the inhibition of convective rolls perpendicular to the magnetic field in Rayleigh-Bénard experiments with mercury [15].

In order to validate these heuristic arguments, which are based on simple minded assumptions we shall now give a numerical example.

2. Numerical illustration.

The reaction terms are given by the *irreversible Brusselator*, the most popular prototype of kinetic

scheme in the studies of spatial chemical dissipative structures :



A and B are the major species, X and Y the intermediate species. The third step is autocatalytic. As previous authors we introduce the following scaled variables :

$$\begin{aligned}
 A &= \left[\frac{k_1^2 k_3}{k_4^3} \right]^{1/2} [A] , & B &= \left[\frac{k_2}{k_4} \right] [B] \\
 X &= \left[\frac{k_3}{k_4} \right]^{1/2} [X] , & Y &= \left[\frac{k_3}{k_4} \right]^{1/2} [Y] \\
 t &= k_4 \tau , & D_\alpha &= \frac{\tilde{D}_\alpha}{k_4}
 \end{aligned}
 \tag{2}$$

where $[\alpha]$ is the concentration of species α , τ the time and \tilde{D}_α the molecular diffusion coefficient of species α . Using mass action law, the rate equation are :

$$\begin{aligned}
 \frac{\partial A}{\partial t} &= -k_A A + D_A \Delta A \\
 \frac{\partial B}{\partial t} &= k_B B X + D_B \Delta B \\
 \frac{\partial X}{\partial t} &= A - (B + 1) X + X^2 Y + D_X \Delta X \\
 \frac{\partial Y}{\partial t} &= B X - X^2 Y + D_Y \Delta Y
 \end{aligned}
 \tag{3}$$

with $k_A = \frac{k_1}{k_4}$ and $k_B = \frac{k_2^2}{k_3 k_4}$.

The first equation does not involve any species except A . The following analytical solution can be obtained :

$$A(y) = C_1 \exp(\omega y) + C_2 \exp(-\omega y) \tag{4}$$

with

$$\begin{aligned}
 \omega &= \sqrt{\frac{k_A}{D_A}} , & C_1 &= \frac{A(0) - A(\ell) \exp(\omega \ell)}{1 - \exp(2 \omega \ell)} , \\
 & & C_2 &= A(0) - C_1 .
 \end{aligned}$$

Concentrations $A(0)$ and $A(\ell)$ are fixed on boundaries W1 ($y = 0$) and W2 ($y = \ell$) as stated in the previous section but mixed compositions of A and B were used rather than a strict separation of species. After the elimination of A , the system is reduced to three independent nonlinear partial differential equations, which were solved numerically by

the method of lines. From previous analytical and numerical results [1], obtained for one dimensional geometry with uniform A and B , the following suitable parameters were conjectured :

$$\begin{aligned}
 \text{on W1} & \begin{cases} A(0) = 3 \\ B(0) = 2 \\ X(0) = Y(0) = 0 \end{cases} \\
 \text{on W2} & \begin{cases} A(\ell) = 1 \\ B(\ell) = 6 \\ X(\ell) = Y(\ell) = 0 \end{cases}
 \end{aligned}$$

$$k_A = k_B = 10^{-2} ; \quad D_A = D_B = 12 ;$$

$$D_X = 4 ; \quad D_Y = 20$$

and where the system dimensions were $L = 50$ and $\ell = 43$ respectively in the Ox and the Oy directions.

The system was initialized in different nonhomogeneous configurations in order to let different spatial modes grow. Identical asymptotic patterns were observed but for a translation along Ox due to the periodic boundary conditions. We report different graphic representations of the X concentration pattern. The same type of profile was obtained for Y .

Figure 2 is a 3D perspective representation of the surface $X(x, y)$. The sharp peaks indicate both sharp gradients in direction Oy (reaction front) and symmetry breaking instability in direction Ox (Turing instability). Figure 3 exhibits some $X(x)$ profiles at increasing values of y . Close to the walls the concentration is constant along Ox ; far from the walls a spatial mode of wavelength $\lambda = L/3$ emerges as a result of the Turing instability with a maximum amplitude in the front region ($y = 25$). Figure 4 is the concentration map of X . This map shows how the spatial mode along Ox induces a large modulation of the isoconcentration curves in the front region. For a better visualization of the front we imagine a color indicator changing from clear to dark for $X > 1.5$ (Fig. 5a). A first color change

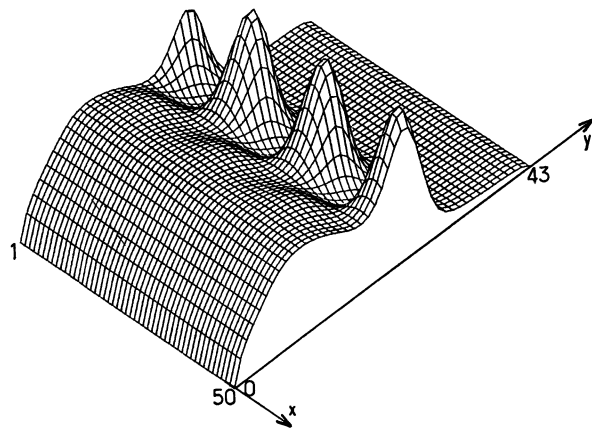


Fig. 2. — Concentration map of species X : 3-D representation.

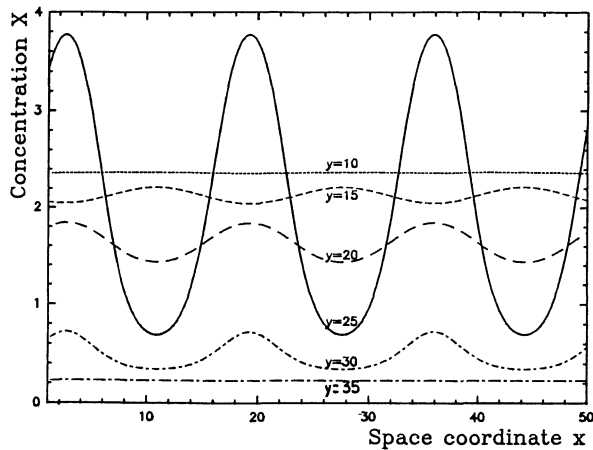


Fig. 3. — Concentration profiles of species X at various y values.

reveals the increase of X with the distance to $W1$ but no symmetry breaking. The second change reveals a sudden decrease of X within the front and the symmetry breaking related to the Turing instability. If the color change threshold is raised to 1.9 (Fig. 5b) the front even exhibits a sub-structure in relation with the sharp peaks of figure 2.

As our purpose is mainly to lay the foundation of a new experimental approach to the stationary Turing dissipative structures, we have focused our attention on the fully developed pattern. Nevertheless, it can be interesting to determine the nature of the transition by investigating the vicinity of the bifurcation point. We report here on the numerical results obtained when concentration B on wall $W2$ is taken as the bifurcation parameter. The amplitude of spatial oscillations in direction Ox at $y = 25$ was retained as the order parameter. This amplitude decreases with B and vanishes at the bifurcation

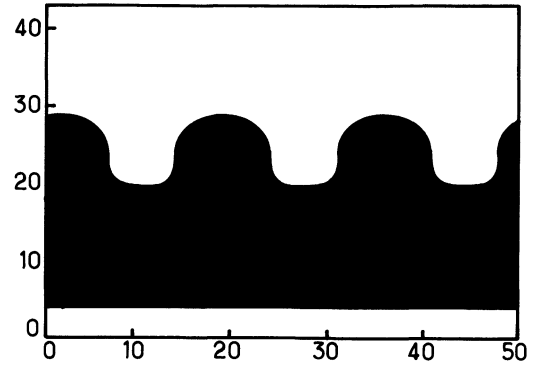


Fig. 5a. — Front structure visualized at $X > 1.5$.

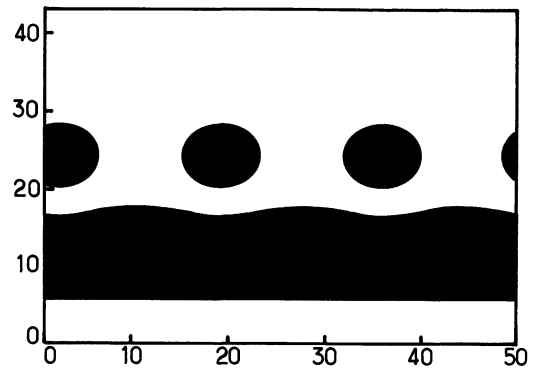


Fig. 5b. — Front structure visualized at $X > 1.9$.

point $B_c = 4.8386 \pm 0.0001$. In the limit of numerical accuracy no hysteresis was observed which indicates a supercritical bifurcation (Fig. 6a). Moreover the Log-Log plot of this amplitude over $B - B_c$ is in excellent agreement with the classical exponent 0.5 as shown in figure 6b.

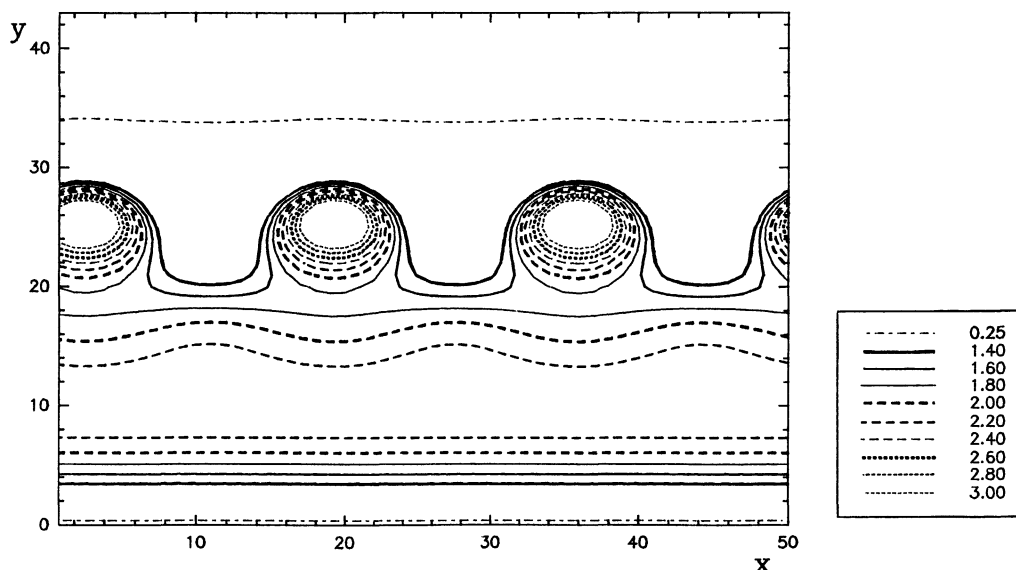


Fig. 4. — Concentration map of species X : isoconcentration curves.

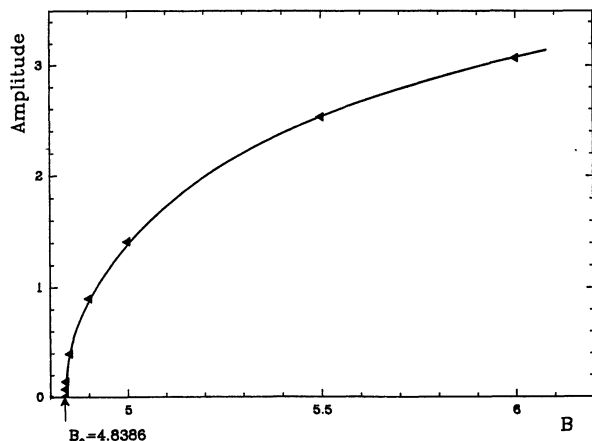


Fig. 6a. — Amplitude of stable spatial mode over bifurcation parameter $B(\ell)$.

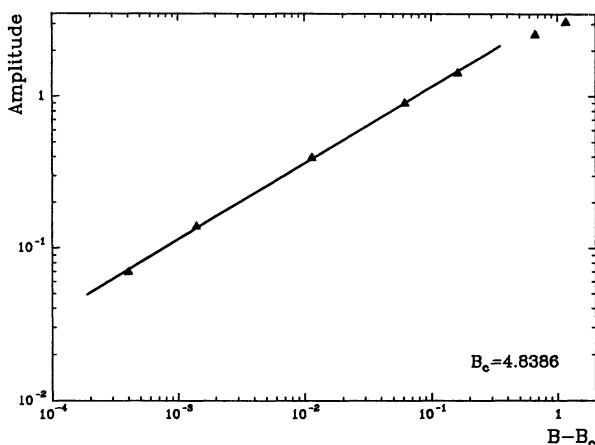


Fig. 6b. — Amplitude of stable spatial mode over $B - B_c$ (Log-Log plot). The straight line has slope 0.5 and corresponds to the classical exponent.

Conclusion.

The results of our numerical simulations support the conclusions obtained on the basis of heuristic arguments. We have shown that the symmetry breaking induced by the Turing instability can be revealed by the self-organization of a stationary reaction front where no convective or other thermal processes are involved, and yields a periodic structure. A practical experimental set up in agreement with the scheme of figure 1 should be easy to build. The only role of the permeable membrane is the separation of the reacting medium from the turbulent or convective motions in the feed. The membrane can be made of thin

fritted glass or paper filter. Since the reactor is a thin film of negligible volume, only moderate flows (or buffered well stirred reservoirs) are necessary on the boundaries. This solves the crucial problem of the continuous feed of the system to maintain non equilibrium conditions. The periodic boundary condition are well approximated either in a thin film formed by the gap between two close concentric cylinders or in a flat fluid ring limited by two large circular walls. No flux boundary conditions (rigid impermeable wall parallel to Oy) could be used as well. The expected structure, localized far from the walls, should be quite insensitive to small perturbations to local feed inhomogeneities on the boundaries. These perturbations are damped by molecular diffusion before they reach the instability region of the front. This point has been confirmed on the model by computer tests.

A common objection to the experimental observation of Turing structures is the need for different diffusion coefficients : this requirement should be difficult to achieve in dilute aqueous solution where most of small molecules coefficients range only from $0.5 \text{ cm}^2 \text{ s}^{-1}$ to $2 \text{ cm}^2 \text{ s}^{-1}$ but in gels or porous films the differences between the effective coefficients can be much larger. Use of such media would also avoid parasitic convective effects.

Finally, analogous experiments performed in a thin circular flat film of gel with the B. Z. reaction have just been reported (16). They confirm the formation of a reaction front visualized in this case with ferroin as a redox indicator. The observed deformation kinks propagating along the front clearly correspond to excitations of the same nature as the travelling waves in the currently observed target patterns. Though the propagation is driven by diffusion, the excitation do not result from a reaction-diffusion instability and these waves are not relevant of the spatial structures discussed in our paper. Nevertheless, the possibility of propagating excitations localized within a reaction front is very stimulating in the perspective to bring experimental evidence of a Turing instability through a steady structured front. Experimental research is currently in progress.

Acknowledgments.

I am specially indebted to P. De Kepper and A. Arnéodo for stimulating discussions. I also thank H. Swinney for careful reading of the manuscript. This work was supported by the BP Venture Research Unit.

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