Biophysical Chemistry 3 (1975) 345–354

DISSIPATIVE STRUCTURES IN TWO DIMENSIONS

T. ERNEUX and M. HERSCHKOWITZ-KAUFMAN
Faculté des Sciences, Université Libre de Bruxelles, Belgium

Received 13 May 1975
Revised manuscript received 11 August 1975

Some further results on dissipative structures for one- and, more particularly, for two-dimensional bounded systems are presented. A model chemical network involving reactions and diffusion is investigated. The influence of the boundary conditions (no fluxes or fixed concentrations), the geometrical shape of the limits (circular or rectangular) and the size of the system on the variety of possible patterns is demonstrated. A comparison of the numerical results with bifurcation theory is outlined. Finally, the problem of multiplicity of stable ordered solutions which turns out to increase sharply from one to two dimensions, is discussed.

1. Introduction

Theoretical studies of open chemical networks governed by non linear rate equations have shown that beyond a critical distance from thermodynamic equilibrium, the steady state corresponding to the extrapolation of the close-to-equilibrium behavior may become unstable [1].

Such systems then evolve to new stable regimes which may exhibit temporal and spatial organization [2–4]. These configurations have been called "dissipative structures" by Prigogine et al. because they require continuous dissipative processes for their appearance and maintenance.

The generality of this type of behavior in non linear systems seems to be confirmed by several laboratory experiments [5] for example on the Belousov–Zhabotinskii system [6–8] where reproductible chemical wave propagation phenomena are observed. In the field of biological regulatory processes a typical example is provided by the glycolitic system [9] which exhibits both uniform and space dependent time oscillations. More recently, spatially ordered regimes in the pH distribution across artificial enzyme membranes have been observed [10].

On the other hand, several aspects of complex biological order phenomena such as development or the electrical activity of the brain have also been investigated in the framework of dissipative structures [11–13,24].

In agreement with the observations, mathematical models based on the kinetic data available were elaborated for systems involving both chemical reactions and diffusion processes. Further simplified models have also been conceived in order to better understand the mathematical aspects related to the occurence of the dissipative structures. In this context, one of the most studied models is the simple trimolecular scheme [14–18]:

$$A \xrightarrow{k_1} X, \quad B + X \xrightarrow{k_2} Y + D,$$
$$2X + Y \xrightarrow{k_3} 3X, \quad X \xrightarrow{k_4} E, \tag{1}$$

involving only two intermediates X and Y. The reaction steps are all irreversible: system I operates thus automatically at infinite distance of thermodynamic equilibrium. Notice the presence of the autocatalytic step in the third reaction which introduces in the model the necessary non linear element. Model I does not represent any known chemical reaction chain but, rather, is a minimal model capable of exhibiting cooperative behavior.

The object of this paper is to study the possibilities of appearance of time periodic structures for *bounded* one-dimensional systems and to investigate symmetry breaking situations for two-dimensional systems corresponding to different geometries such as a rectangle or a circle. The motivation for investigating two-dimensional systems is twofold. First many bio-physical processes take place in two-dimensional media like membranes. And second, additional symmetry features of the spatial domain considered introduce a highly interesting interplay between form and dimensions, and enhance in this way the variety of possible patterns.

In section 2 we briefly recall the principal results obtained in one dimension for model I. We also investigate the occurrence of time periodic behavior for two kinds of boundary conditions: fixed concentrations and no fluxes. Section 3 is devoted to the linear stability analysis in two dimensions of the unique uniform steady state of the model for the same boundary conditions as in section 2.

In their general features the instability conditions on the physico-chemical parameters are the same as those encountered in a one-dimensional analysis; however the wave-number, k, which characterizes the influence of diffusion on the steady-state stability is differently defined. Moreover, the spectrum of permitted values of k appears to be directly related to the size, the shape of the system, and the boundary conditions.

Particular attention was focused on the variety of the new stable solutions that could be obtained numerically and their relation to the results of the stability analysis of the uniform steady state.

In the last section, we discuss the problem of successive instabilities which is related to the existence of multiple solutions and may become more common for two-dimensional systems.

2. Some further results in one-dimensional systems

The system is considered open to products A, B, D and E whose concentrations are maintained time and space independent. The kinetic equations describing the evolution of the intermediate products are:

$$\partial_t X = k_1 A + k_3 X^2 Y - k_2 BX - k_4 X + D_X \Delta_s X,$$

$$\partial_t Y = k_2 BX - k_3 X^2 Y + D_Y \Delta_s Y, \qquad 0 \leqslant S \leqslant S_0, \quad (1)$$

where ∂_t and Δ_s represent respectively $\partial/\partial t$ and $\partial^2/\partial S^2$.

Diffusion is governed by Fick's law and the diffusion coefficients D_X, D_Y of X and Y are supposed to be constant. The boundary conditions are chosen to be either fixed concentrations:

$$X(0) = X(S_0) = \text{const.}, \qquad Y(0) = Y(S_0) = \text{const.}.$$

or zero fluxes:

$$\left(\frac{\partial X}{\partial S}\right)_0 = \left(\frac{\partial X}{\partial S}\right)_{S_0} = 0, \quad \left(\frac{\partial Y}{\partial S}\right)_0 = \left(\frac{\partial Y}{\partial S}\right)_{S_0} = 0.$$

Without loss of generality we may introduce the following two modifications of system (1):

1. By simple scale transformations of time and concentration of each product, all forward kinetic constants are put equal to one. Thus setting:

$$t' = k_4 t,$$

$$X' = (k_3/k_4)^{1/2} X, \qquad Y' = (k_3/k_4)^{1/2} Y,$$

$$A' = (k_1^2 k_3/k_4^3)^{1/2} A, \qquad B' = (k_2/k_4) B.$$

System (1) becomes in the new variables t', X', Y', A' and B':

$$\partial_{t'} X' = A' + X'^2 Y' - B'X' - X' + D'_X \Delta_s X', \qquad (2)$$

$$\partial_{t'} Y' = B'X' - X'^2 Y' + D'_Y \Delta_s Y', \qquad 0 \leqslant S \leqslant S_0,$$

where we defined

$$D'_X = D_X/k_4, \qquad D'_Y = D_Y/k_4.$$

2. We introduce in system (2) the dimensionless parameter l defined as $l = S/S_0^*$ where S is the real space dimension and S_0^* an arbitrary chosen length ($0 \leqslant l \leqslant l_0$; $l_0 = S_0/S_0^*$). In this way, the reaction-diffusion equations to be studied take the form:

$$\partial_t X = A + X^2 Y - (B+1) X + D_X \Delta_l X,$$

$$\partial_t Y = BX - X^2 Y + D_Y \Delta_l Y, \qquad 0 \leqslant l \leqslant l_0. \qquad (3)$$

Note that, thanks to the scale transformation defined above, one may consider quite realistic values of the rate constants and the diffusion coefficients which are compatible with eq. (3).

For suitable boundary conditions there exists a single uniform steady state solution of system (3) given by:

$$X_0 = A, \qquad Y_0 = B/A. \qquad (4)$$

The stability properties of this steady state are determined by the characteristic equation which is in our case [14]:

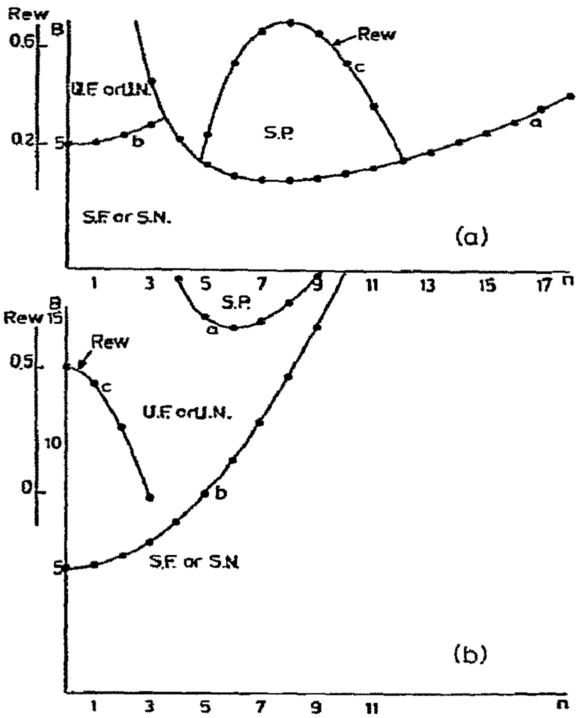

Fig. 1. Stability diagram $B = B(n)$ for a one-dimensional system of length $l = 1$. Curve a, $B = 1 + A^2 D_X/D_Y + A^2/(D_Y k_i^2) + D_X k_i^2$, delimits the region where state (4) is a saddle point (SP) always unstable and curve b, $B = 1 + A^2 + (D_X + D_Y) k_i^2$, the region corresponding to an unstable focus or unstable node (UF, UN). The permitted values of k_i are: $k_i = n\pi/l$ with $n = 0, 1, 2\ldots$ except for fixed boundary conditions where $n = 0$ is excluded. The black dots give the allowed states of the system. Curve c shows the real part of the positive eigenvalues $\omega(n)$, for a given value of B. The conditions upon the parameters are:

(a) $D_X = 1.6 \times 10^{-3}$, $D_Y = 8 \times 10^{-3}$, $A = 2$; Re ω is plotted for $B = 4.6$. The first bifurcation corresponds to Re $\omega_i = 0$, Im $\omega_i = 0$ and $k_{ic} \neq 0$.

(b) $D_X = 8 \times 10^{-3}$, $D_Y = 4 \times 10^{-3}$, $A = 2$; Re ω is plotted for $B = 6$. The first bifurcation corresponds to Re $\omega_i = 0$, Im $\omega_i \neq 0$ with either $k_{ic} = 0$ (zero fluxes at the boundaries) or $k_{ic} \neq 0$ (fixed boundary conditions).

$D_X = 1.6 \times 10^{-3}$, $D_Y = 8 \times 10^{-3}$, $A = 2$; Re is plotted for $B = 4.6$. (a) The first bifurcation corresponds to Re $\omega_i = 0$.

$$\omega_i^2 - [B - 1 - A^2 - (D_X + D_Y) k_i^2]\ \omega_i + A^2(1 + k_i^2 D_X) + D_Y k_i^2 (1 - B) + D_X D_Y k_i^4 = 0. \tag{5}$$

The discrete spectrum of values of $k_i = k_i(n)$ is found by determining the eigenvalues, $-k_i^2$, of the laplacian operator knowing the boundary conditions, the spatial extension of the system and the geometry of the boundaries:

$$\Delta \phi_i(r) = -k_i^2 \phi_i(r).$$

There exist two ways to destabilize state (4): through real (Re $\omega_i > 0$, Im $\omega_i = 0$) or complex (Re $\omega_i > 0$, Im $\omega_i \neq 0$) eigenvalues. Which type of instability occurs first when B increases from zero depends on the values of the physico-chemical parameters like e.g. the ratio of the diffusion coefficients D_X/D_Y (see fig. 1).

In order to understand the implications of the instability conditions, the evolution of the system was studied numerically beyond the different transitions points. This analysis revealed the following features:

1. When the unstable steady state is a "saddle point", stationary non-uniform stable structures have been obtained. Quite recently, an analytical approach was carried out by Nicolis, Auchmuty [19] and Herschkowitz-Kaufman [20] for the stationary solutions around the first bifurcation point. It appears clearly, from this analysis, that the critical value k_i describes the wavelength of the emerging dissipative structure since the corresponding eigenfunction ϕ_i represents the first approximation to it.

2. When the uniform steady state becomes an "unstable focus", a stable time periodic regime sets up. However for the same values of the parameters, two different behaviors are observed depending on the nature of the boundary conditions.

For fixed concentrations at the boundaries, we obtain a spatio-temporally ordered regime (fig. 2) as expected from the linear stability analysis where the possibility of uniform solutions are ruled out ($k_i \neq 0$, and the corresponding ϕ_i's are never space independent). An analytical analysis of the first bifurcation in this case is also developed by Nicolis and Auchmuty [21].

In the second case – zero fluxes at the boundaries – when the initial conditions are not uniform, we observe a sharp concentration front propagating from one boundary to another; but, this periodical repeating propagation stage dissapears gradually and seems to tend to uniform sustained oscillations. This agrees with the predictions of the linear stability analysis according to which the state at $k_i = 0$ corresponds to the first bifurcation.

These two different cases underline the importance of considering *finite systems* which allows to distinguish between transient phases (such as in case 2) and asymptotic states (such as in case 1). However, the

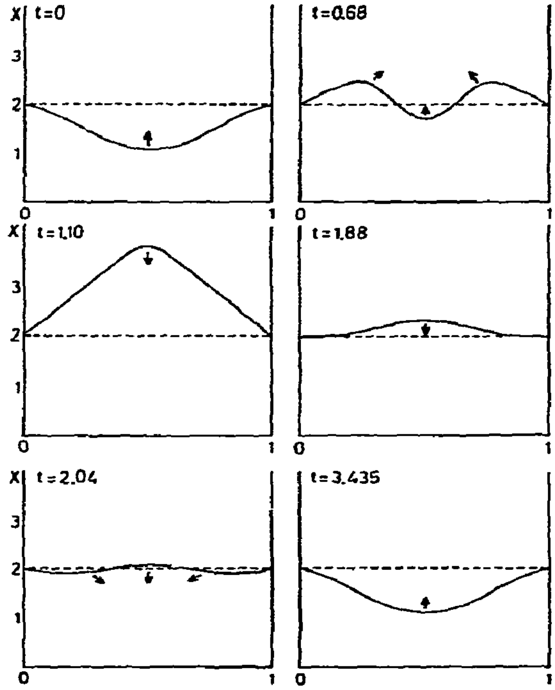

Fig. 2. One-dimensional spatio-temporal regime for fixed concentrations at the boundaries. Dashed line: unstable steady state; Full line: stable periodic regime. $l = 1$; $A = 2$, $B = 5.45$; $D_X = 8 \times 10^{-3}$; $D_Y = 4 \times 10^{-3}$.

possibility of "wave-like solutions" in reaction-diffusion systems subjected to zero fluxes at the boundaries is conceivable as a result of a new instability corresponding to further bifurcations. On the other hand, it has been established that systems involving more than 2 variables can lead to time-periodic space-dependent solutions even for the first bifurcation [21].

3. Two-dimensional systems

In this section, we describe the properties of model I for a two-dimensional medium; we choose two types of boundary geometries: the rectangle and the circle. For these two cases, the general features of the stability analysis of the uniform steady state (4) are the same as those encountered in one dimension (see caption fig. 1). Only the explicit form of k_i, $\phi_i(r)$, the eigenvalues and eigenfunctions of the laplacian operator satisfying the boundary conditions are changed.

Note that as in the preceding section, we use dimensionless variables which are now defined by

$$x = S_x/S_0^*, \qquad 0 \leqslant x \leqslant l_x; \; l_x = S_{x0}/S_0^*,$$

$$y = S_y/S_0^*, \qquad 0 \leqslant y \leqslant l_y; \; l_y = S_{y0}/S_0^*,$$

for the rectangle of dimensions $S_{x0} \times S_{y0}$; and by

$$r = R/S_0^*, \qquad 0 \leqslant r \leqslant r_0; \; r_0 = R_0/S_0^*,$$

for the circle of radius R_0.

3.1. Linear stability analysis

3.1.1. Rectangle of sides $l_x \times l_y$

According to the linear stability analysis, we are looking for the perturbations u and v solutions of the linearized equations of motion

$$u = |X - X_0|, \qquad u/X_0 \ll 1,$$

$$v = |Y - Y_0|, \qquad v/Y_0 \ll 1.$$

One can show that the general expressions for u and v are infinite series of the eigenfunctions $\exp(\omega_i t)\phi_i(r)$; these eigenfunctions take the following explicit form.

a) For fixed concentrations at the boundaries (equal to X_0 and Y_0):

$$\begin{pmatrix} u_i \\ v_i \end{pmatrix} = \begin{pmatrix} c_i \\ d_i \end{pmatrix} \exp(\omega_i t) \sin(k_{xi} x) \sin(k_{yi} y), \tag{8a}$$

with

$$k_{xi} = n_{xi}\pi/l_x \quad \text{and} \quad n_{xi} = 1, 2, \ldots,$$

$$k_{yi} = n_{yi}\pi/l_y \quad \text{and} \quad n_{yi} = 1, 2, \ldots. \tag{9a}$$

b) For zero fluxes at the boundaries:

$$\begin{pmatrix} u_i \\ v_i \end{pmatrix} = \begin{pmatrix} c_i \\ d_i \end{pmatrix} \exp(\omega_i t) \cos(k_{xi} x) \cos(k_{yi} y), \tag{8b}$$

with

$$k_{xi} = n_{xi}\pi/l_x \quad \text{and} \quad n_{xi} = 0, 1, 2, \ldots,$$

$$k_{yi} = n_{yi}\pi/l_y \quad \text{and} \quad n_{yi} = 0, 1, 2, \ldots. \tag{9b}$$

The permitted values of k_i are related to k_{xi}, k_{yi}, by

$$k_i = (k_{xi}^2 + k_{yi}^2)^{1/2} = \pi(n_{xi}^2/l_x^2 + n_{yi}^2/l_y^2)^{1/2}, \tag{10}$$

where n_{xi} and n_{yi} are given either by (9a) or (9b).

ω_i is solution of the characteristic eq. (5) and c_i, d_i are two arbitrary constants related by

$$(D_X k_i^2 + \omega_i + 1 - B)\, c_i = A^2 d_i. \tag{11}$$

3.1.2. Circle of radius r_0

Applying the same considerations as above, we see, in terms of polar coordinates (r, θ) that the eigenfunctions are now of the form:

$$n = 0: \begin{pmatrix} u_i \\ v_i \end{pmatrix} = \begin{pmatrix} c_i \\ d_i \end{pmatrix} \exp(\omega_i t) J_0(k_i r), \quad (12)$$

$$n \neq 0: \begin{pmatrix} u_i \\ v_i \end{pmatrix}_\pm = \begin{pmatrix} c_i \\ d_i \end{pmatrix}_\pm \exp(\omega_i t)(e^{in\theta} \pm e^{-in\theta}) J_n(k_i r),$$

where $J_n(k_i r)$ is the Bessel function of integer order n. The eigenvalues k_i corresponding to the $J_n(k_i r)$ are given by:

a) For fixed concentrations (X_0, Y_0) at the boundaries:

$k_i r_0 = j_{n,s}$ (sth zero of the nth Bessel function $J_n(\zeta)$) with

$$n = 0, \quad s = 1, 2 \ldots; \quad n \neq 0, \quad s = 2, 3 \ldots. \quad (13a)$$

b) For zero fluxes of X and Y at the boundaries:

$$k_i r_0 = j'_{n,s}$$

(sth zero of the first derivative of the nth Bessel function) with

$$n = 0, \quad s = 1, 2 \ldots; \quad n \neq 0, \quad s = 1, 2 \ldots. \quad (13b)$$

Like for the rectangle, ω_i is solution of the characteristic eq. (5) and c_i, d_i are related by (11).

We conclude that for two-dimensional systems, the instability conditions are the same as in one dimension (same characteristic equation). Though the permitted values of k_i are changed, one has for the first bifurcation:

$k_{ic} = 0$, for an unstable focus with zero fluxes at the boundaries;

$k_{ic} \neq 0$, for an unstable focus with fixed concentration at the boundaries;

$k_{ic} \neq 0$, for a saddle point.

3.1.3. Degenerate cases

In two dimensions, depending on the symmetry of the spatial domain, there exists the possibility for the first instability to be the origin of multiple bifurcations; indeed, it can now happen that more than one eigenfunction $\phi_i(r)$ corresponds to the same eigenvalue $-k_i^2$ (and thus to the same value of ω_i). When $n \neq 0$ this is always the case for the circle which presents naturally for each k_i two independent eigenfunctions $\phi_i(r)$:

$$\phi_{i,1} = c \sin n\theta \, J_n(k_i r),$$

$$\phi_{i,2} = c \cos n\theta \, J_n(k_i r), \qquad (n \neq 0).$$

The same situation exists for the square when $n_{xi} \neq n_{yi}$.

An example on the square is considered in the next section. Besides these examples involving high symmetries, degenerate eigenvalues of the laplacian operator can also appear for the rectangle. These situations require however very particular relations between the lengths of the sides.

It should also be pointed out that if the first bifurcation does not lead to a case of pure degeneracy, it can happen, both in one and two dimensions, that two different bifurcations appear for identical values of the bifurcating parameter as represented on the diagram below. This case of "accidental" degeneracy has been analyzed analytically for a one-dimensional system by Boa [22]. The degeneracy specific to the square (pure degeneracy) has been investigated by Shiffman [23].

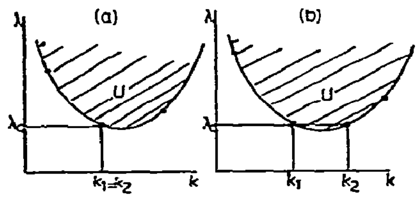

Diagram 1. U denotes the unstable uniform steady state. λ is the bifurcating parameter. (a) Pure degenerate case, (b) "accidental" degenerate case.

3.2. Numerical simulations

In this section, we present different examples of solutions obtained numerically. The conditions upon the physico-chemical parameters A, B, D_X, D_Y, (l_x, l_y) or r_0 have been chosen to give simple dissipative structures. However, this first approach gives a good idea of the complexity of the solutions which can be obtained in two-dimensional systems. Like for one dimension, we have focused our attention on the two different instability conditions $B = B(k_i)$ (see caption fig. 1) leading either to time periodic or to stationary solutions. The first case leads globally to the same conclusions as we encountered in one dimension, for example: a continuously repeated concentration progression to the center for a circle with fixed uniform boundary conditions $X = X_0$, $Y = Y_0$ and the transient periodic non uniform phenomena when zero fluxes of X and Y are prescribed at the boundaries.

The second case has been more thouroughly investigated because it clearly reflects the influence of both the applied conditions on the concentrations at the boundaries and the different shapes. Figs. 3, 5, 7 and 9 give stationary structures, obtained for intermediate X when the unstable steady state X_0 is slightly perturbed (in most cases, the position of the initial perturbation applied to the uniform unstable solution is indicated). In each of those figures the resulting pattern reflects the geometrical properties of one of the unstable eigenfunctions $\phi_i(r)$. We characterize each of them by two integer numbers which are defined in the linear stability analysis [see (9a), (9b), (13a), (13b)]: n_x, n_y for the rectangle and n, s for the circle. The critical eigenfunctions ϕ_i which correspond to a ω_i with a positive real part are indicated in a stability diagram $B = B(k_i)$ for the value of B considered (see figs, 4, 6, 8 and 10). In fig. 6, we also present the values of the positive real part of ω_i. This gives some idea of the relative amplifications of the different unstable eigenfunctions. We see that the stationary dissipative structure obtained can be related to one of those modes which have an appreciable positive real part of ω_i.

Fig. 3 presents the circle with no fluxes at the

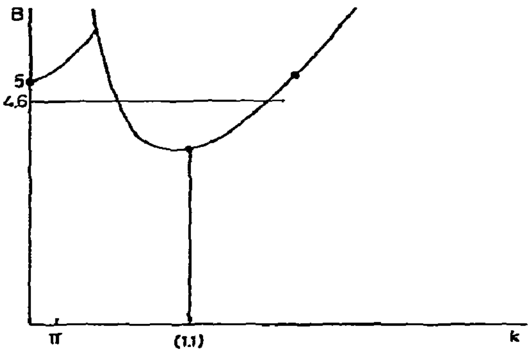

Fig. 4. Stability diagram $B = B(k)$ corresponding to the conditions of fig. 3. The dots indicate the permitted states of the system, which are characterized by the two numbers (n, s) defined in the linear stability analysis.

boundaries; clearly, the stationary structure has the properties of the unstable eigenfunction $n = 1, s = 1$ with *its characteristic polarity*. Notice that in one dimension polarity corresponds to spontaneous onset of a macroscopic concentration gradient along the system resulting from different values of X or Y at the boundaries $l = 0$ and $l = l_0$ [11]. The organized pattern which appears in fig. 3 is not a trivial extension of one-dimensional polar structures to the circle; in fact we observe that the circular limits permit us to have in the same structure, a region of space which is greatly organized (around $\theta \approx 0$) and another quasi-uniform region (around $\theta \approx \pi/2$).

In fig. 5, we considered the circle for fixed concen-

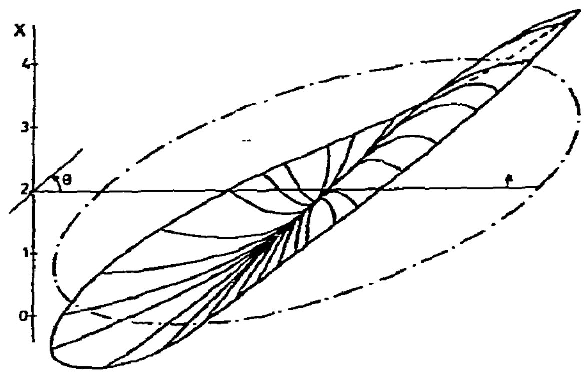

Fig. 3. Polar stationary structure for the circle with zero fluxes at the boundaries. Broken dotted line: unstable steady state. $R = 0.1$; $A = 2$; $B = 4.6$; $D_X = 3.25 \times 10^{-3}$; $D_Y = 1.62 \times 10^{-2}$.

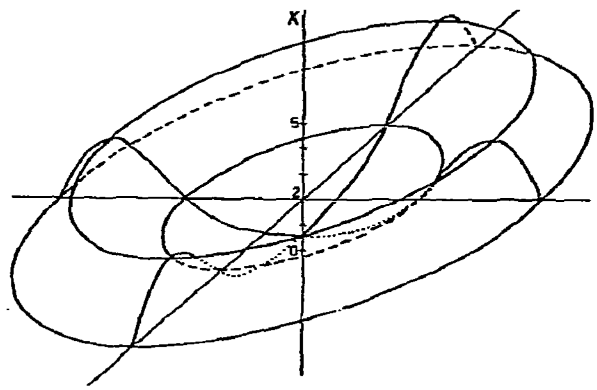

Fig. 5. Stationary structure for the circle with fixed concentrations at the boundaries. $R = 0.2$; $A = 2$; $B = 4.6$; $D_X = 1.6 \times 10^{-3}$; $D_Y = 8 \times 10^{-3}$.

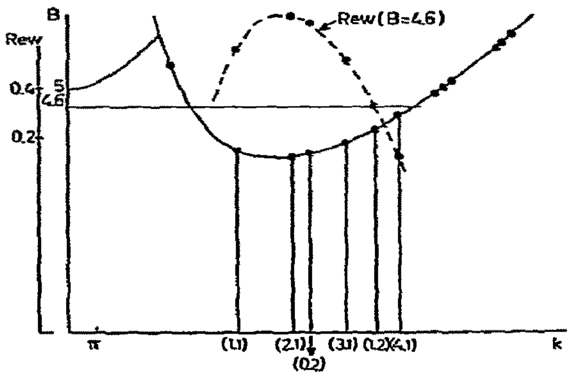

Fig. 6. Stability diagram $B = B(k)$ corresponding to the conditions of fig. 5.

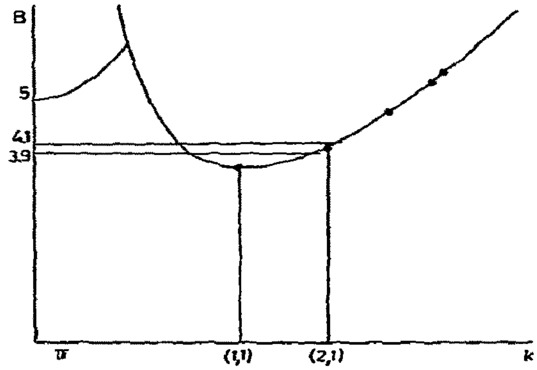

Fig. 8. Stability diagram $B = B(k)$ corresponding to the conditions of fig. 7. The states which are accessible for the system are represented by black dots and characterized by two numbers (n_x, n_y).

trations at the boundaries: the structure is phase independent, and corresponds to the eigenfunction $n = 0$, $s = 2$.

Fig. 7a shows the rectangle for the same boundary conditions and corresponds to the eigenfunction $n_x = 1$, $n_y = 1$. This case was also considered for a greater value of B; the resulting pattern (fig. 7b) does not longer, as in fig. 7a, totally reflect the symmetry properties of the eigenfunction $\phi(n_x = 1, n_y = 1)$ but undergoes a grooving in its central region which increases with B. This shows the influence on the bifurcating solution of subharmonical terms [19] which modify the geometrical aspects of the first approximation $[\phi(n_x = 1, n_y = 1)]$ for increasing values of the bifurcating parameter (here: $B - B_c$).

Fig. 9 presents the square with no fluxes at the

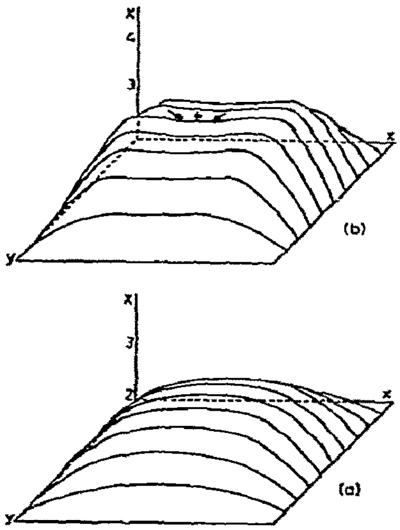

Fig. 7. Stationary structures for a rectangle with fixed concentrations at the boundaries. $l_x = 0.231$; $l_y = 0.165$; $A = 2$; $D_X = 1.6 \times 10^{-3}$; $D_Y = 8 \times 10^{-3}$. (a) $B = 3.9$, (b) $B = 4.1$.

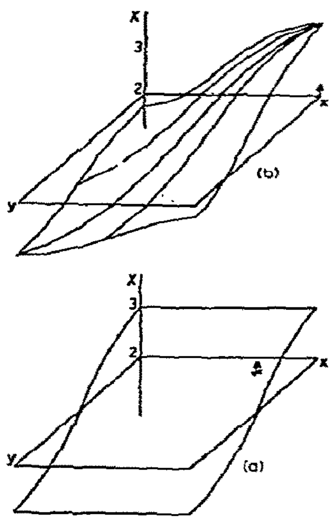

Fig. 9. Stationary structures for a square subjected to zero fluxes at the boundaries. $l_x = l_y = 0.132873$; $A = 2$; $B = 4$; $D_X = 1.6 \times 10^{-3}$; $D_Y = 8 \times 10^{-3}$. (a) Stable structure, (b) unstable structure.

boundaries, corresponding to a degenerate case. Two different initial conditions were imposed: fig. 9a presents the first case where we slightly perturbed the unstable steady state at one point ($\delta X = 0.02$): this leads to the simple structure of fig. 9a which can be characterized by $r_x = 0, n_y = 1$; fig. 9b presents the second case where we have perturbed at another point ($\delta X = 0.02$): the emerging structure is the degenerate solution which corresponds to $\phi(n_x = 0, n_y = 1) + \phi(n_x = 1, n_y = 0)$. But this structure is not stable: upon a slight perturbation it tends either to the structure corresponding to the shapes of eigenfunctions $\phi(n_x = 0, n_y = 1)$ such as in fig. 9a, or to the structure characterized by $\phi(n_x = 1, n_y = 0)$.

Let us now consider the possibility of multiplicity of structures: in the preceeding examples, we have analyzed the structure emerging when the uniform unstable steady state was subjected to a slight perturbation. In fact, for the same physico-chemical parameters, many stable structures corresponding to different eigenfunctions ϕ_i may be obtained. One-dimensional studies [20] have already shown that several stable solutions can, indeed, be obtained from different initial conditions. All of them can be related to one of the unstable eigenfunctions generally one which correspond to an important real part for ω_i. This is certainly, also the case in two dimensional systems, where the possibility of new bifurcations increases rapidly with increasing values of the bifurcating parameter. For example, if we compare a one dimensional system of length 1, and a rectangle of sides 1 and 0.5, both with zero fluxes at the boundaries, we have for the eigenvalues k_i corresponding to the different ϕ_i's available:

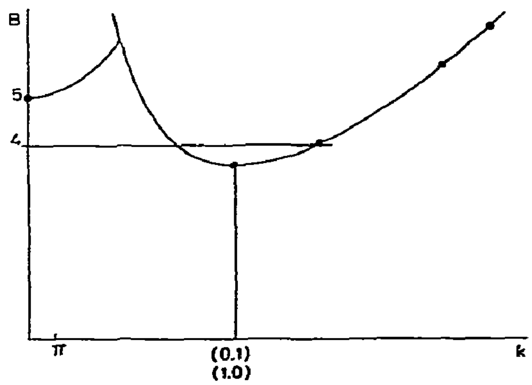

Fig. 10. Stability diagram $B = B(k)$ corresponding to the conditions of fig. 9.

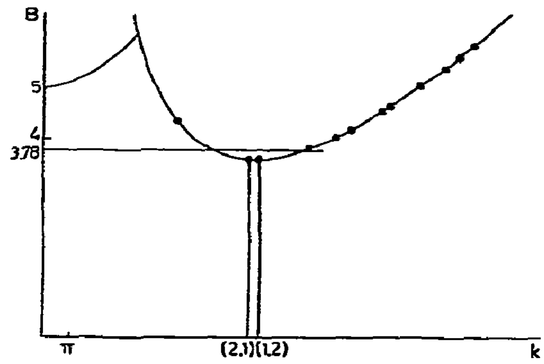

Fig. 11. Stability diagram $B = B(k)$ for the following conditions: $l_x = 0.301486$; $l_y = 0.281353$; $A = 2$; $B = 3.78$; $D_X = 1.6 \times 10^{-3}$; $D_Y = 8 \times 10^{-3}$. Fixed concentrations at the boundaries.

1 dimension: $k_i = n\pi, \quad n = 0, 1, \ldots,$

2 dimensions: $k_i = \pi(n_x^2 + 4n_y^2)^{1/2}$;

$$n_x = 0, 1, 2\ldots, \quad n_y = 0, 1, 2\ldots,$$

If the physico-chemical parameters are chosen such that the permitted values k_j, for which the uniform steady state is unstable obey to: $2\pi \leqslant k_j < 3\pi$, one observes that only $\phi(n = 2)$ satisfies this condition in one dimension whereas in two dimensions, $\phi(n_x = 0, n_y = 1)$, $\phi(n_x = 1, n_y = 1)$ and $\phi(n_x = 2, n_y = 1)$ are to be considered.

This can be illustrated by 2 general bifurcation diagrams (diagram 2): At B^*, only one bifurcation has appeared for one dimension when one increases the value of B from zero. On the contrary, in two-dimensional systems, several bifurcations may occur before B^*. Moreover, as we have already shown, in 2 dimensions multiple solutions can also emerge from the same bifurcation, as a consequence of degenerate eigenvalues in highly symmetrical systems.

A simple example of multiplicity of solutions is presented in fig. 12 for a rectangle of sides (0.301486 × 0.281353) with fixed boundary conditions. Fig. 11 shows the stability diagram for the chosen values of the physico-chemical parameters. We see clearly that the two stable solutions obtained through different initial conditions can be related to the two eigenfunc-

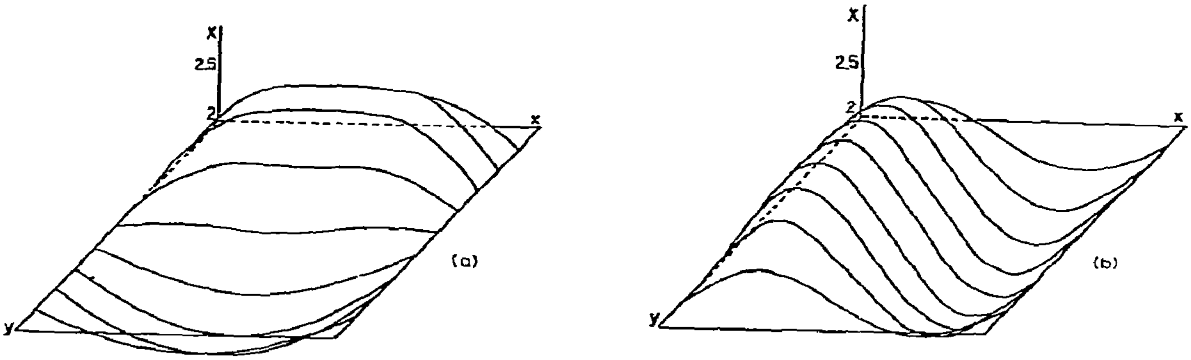

Fig. 12. Two stable solutions obtained through different initial conditions. Physico-chemical parameters as in fig. 11.

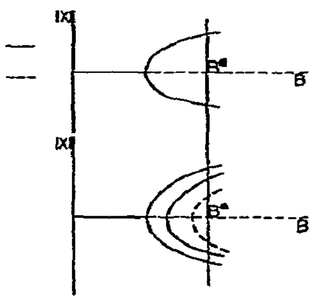

Diagram 2. Full line: stable, dashed line: unstable.

tions which are the only available. Since this example could be conceived as a quasi-degenerate case (the considered rectangle is a slightly distorded square) we investigated the possibility of solutions corresponding to the form $\alpha\phi(1,2) + \beta\phi(2,1)$ were α and β are two constants, but no other stable patterns as those presented in fig. 12a and 12b have been obtained (even as a transient state as it was the case for the square).

4. Discussion

Comparatively to one dimension, two-dimensional systems introduce essentially the influence of the *limit shapes* on the organized patterns which may be obtained in non-linear reaction/diffusion systems. The few examples investigated in the preceding section give us an idea of the variety of solutions available. Like in one dimension, they can be related to the different unstable eigenfunctions of the laplacian operator and considered as bifurcating from the uniform unstable steady state. On the other hand, we see that for given values of the physico-chemical parameters the number of possible bifurcations from the uniform steady state increases very rapidly with the area of the system. Of course, these different bifurcations do not all of them necessary lead to stable branches. The stability of the different solutions may depend on the physico-chemical parameters but also on the boundary properties. On the other hand, recent one dimension simulations seem to confirm that in the case of time periodic solutions, other bifurcations than those we have described, leading to new stable bifurcating branches are not to be excluded.

From the point of view of biology, this high multiplicity of stable, non-trivial solutions of reaction–diffusion equations could have a considerable importance. In particular, the emergence of more and more complex patterns as a result of successive instabilities, either through primary bifurcations or via secondary ones, bears striking analogies with the gradual complexification of morphogenetic patterns in the course of development. This complexification becomes especially spectacular in two dimensions. Further comments on the implication of dissipative structures in development and morphogenesis can be found in ref. [11].

It appears thus that the general outlines of further investigations are the search of analytical expressions of the different bifurcating solutions (a first approach for two dimensional systems is in progress [23]) and the study of their stability: stability of the new branches of solutions appearing through successive bifurcations from the trivial homogeneous steady state solution, but also the stability of the dissipative structure itself.

Acknowledgements

We are greatly indebted to Professor I. Prigogine for his continuous interest in this work. Especial thanks are due to Professor G. Nicolis for many stimulating discussions and critical reading of the manuscript. We also thank Dr. J.W. Turner for his suggestions.

Th. Erneux is supported by l'Institut pour l'Encouragement de la Recherche Scientifique dans l'Industrie et l'Agriculture, I.R.S.I.A., and M. Herschkowitz-Kaufman by the Fonds National de la Recherche Fondamentale Collective.

References

[1] P. Glansdorff and I. Prigogine, Thermodynamic Theory of Structure Stability and Fluctuations (J. Wiley, Interscience, London, 1971).
[2] G. Nicolis and J. Portnow, Chem. Rev. 73 (1973) 365.
[3] A. Goldbeter and R. Lefever, Biophys. J. 12 (1972) 1302;
A. Goldbeter, Proc. Nat. Acad. Sci. (USA) 70 (1973) 3255.
[4] I. Prigogine, R. Lefever, A. Goldbeter and M. Herschkowitz-Kaufman, Nature 223 (1969) 913.
[5] Faraday Symposia, Physical Chemistry of Oscillatory Phenomena, Chem. Soc. (1975) no 9.
[6] A.M. Zhabotinski and A.N. Zaikin, J. Theor. Biol. 40 (1973) 45.
[7] A.T. Winfree, Science 175 (1972) 634.
[8] R. Field and R.M. Noyes, J. Am. Chem. Soc. 96 (1974) 2001.
[9] A. Boiteaux and B. Hess, Faraday Symposium on Physical Chemistry of Oscillatory Phenomena, Chem. Soc. (1975) no 9;
A. Boiteux and B. Hess, unpublished results.
[10] S.R. Caplan, A. Naparstek and N.J. Zabusky, Nature 245 (1973) 364.
[11] A. Babloyantz and J. Hiernaux, Bull. Math. Biol., in press.
[12] H.G. Martinez, J. Theor. Biol. 36 (1972) 479.
[13] W.J. Freeman, Ann. Rev. Biophys. Bioeng. 1 (1972) 225;
W.J. Freeman, in: Proc. IEEE Conf. Autom. Theory, MITRE Corp. Mc Lean, Virginia (1974).
[14] G. Nicolis, Adv. Chem. Phys. 19 (1971) 209.
[15] J. Tyson, J. Chem. Phys. 58 (1973) 3919;
J. Tyson and J.C. Light, J. Chem. Phys. 59 (1973) 4164.
[16] R. Lefever and G. Nicolis, J. Theor. Biol. 30 (1971) 267.
[17] B. Lavenda, G. Nicolis and M. Herschkowitz-Kaufman, J. Theor. Biol. 32 (1971) 283.
[18] M. Herschkowitz-Kaufman and G. Nicolis, J. Chem. Phys. 56 (1972) 1890.
[19] G. Nicolis and J.F.G. Auchmuty, Proc. Nat. Acad. Sci. (USA) 71 (1974) 2748;
J.F.G. Auchmuty and G. Nicolis, Bull. Math. Biol., in press.
[20] M. Herschkowitz-Kaufman, Bull. Math. Biol. in press.
[21] J.F.G. Auchmuty and G. Nicolis, in preparation.
[22] J. Boa, Ph. D. Thesis, Calif. Inst. of Technology (1974);
[23] Y. Shiffmann, Ph. D. Thesis, Université Libre de Bruxelles (1975).
[24] L.A. Segel and J.L. Jackson, J. Th. Biol. 37 (1972) 545.