PERGAMON

Applied Mathematics Letters 14 (2001) 405–411

Applied Mathematics Letters

www.elsevier.nl/locate/aml

A Simple Method of Parameter Space Determination for Diffusion-Driven Instability with Three Species

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(Received and accepted May 2000)

Abstract—A very simple, practical, necessary and sufficient condition for diffusion-driven linear instability and parameter space determination in nonlinear reaction systems with three species is presented. With respect to the stability matrix $\boldsymbol{A}$ from linearization near a stable steady-state of a reaction system, two necessary conditions are given:

(i) $\boldsymbol{A}$ is stable but not negative definite; and
(ii) either the largest diagonal elements of $\boldsymbol{A}$ is positive or the smallest diagonal cofactors of $\boldsymbol{A}$ is negative.

Condition (i) can be generalized to any number of species but (ii) is a stronger condition which, in fact, is shown to be sufficient for diffusion-driven instability. As an example, the result is applied to the three-species Oregonator, the model for the Belousov-Zhabotinsky reaction.

Keywords—Chemical oscillation, Linear analysis, Oregonator, Pattern formation, Reaction-diffusion equation.

1. INTRODUCTION

Diffusion-driven instability is considered one of the mechanisms implicated in the spatial organization in morphogenesis [1]. The interplay between diffusion and nonlinear reaction kinetics giving rise to spatially inhomogeneous patterns comes under the general heading of reaction-diffusion systems. In the case of chemical reactions with only two species, it is well known that a sufficient and necessary condition for a linearly stable homogeneous steady state to be stable against diffusion-driven instability is $a_{11}a_{22} > 0$, where

$$\boldsymbol{A} = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \tag{1}$$

is the matrix characterizing the linear stability of the steady state in the absence of the diffusion. Linear stability requires $\boldsymbol{A}$ to have two eigenvalues with negative real parts [2]. Condition $a_{11}a_{22} > 0$ further guarantees it to be stable against diffusion.

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We thank Professor R. J. Field for helpful discussions.

0893-9659/01/$ - see front matter Typeset by AMS-TEX
PII: S0893-9659(00)00169-5

Here we derive a similarly simple and practical rule for chemical reaction systems with three species. Although it is clear what is required, the derivation of conditions in the case of three species by standard methods are convoluted and impractical to use in determining the parameter space for diffusion-driven instability. We first introduce two necessary conditions for diffusion-driven instability in reaction systems with three species. The first condition can be easily generalized to systems with more than three species. However, the second one is a stronger necessary condition which we show is also sufficient.

We consider the reaction-diffusion system

$$\begin{aligned}\frac{\partial u}{\partial t} &= f(u,v,w) + d_1\nabla^2 u,\\ \frac{\partial v}{\partial t} &= g(u,v,w) + d_2\nabla^2 u,\\ \frac{\partial w}{\partial t} &= h(u,v,w) + d_3\nabla^2 w,\end{aligned} \tag{2}$$

where the f, g, and h represent nonlinear kinetics, and d_1, d_2, and d_3 are the respective diffusion coefficients of u, v, and w. A linear analysis of (2) in the neighborhood of a stable steady-state of its corresponding nonlinear ordinary differential equations yields [2]:

$$\frac{\partial \boldsymbol{y}}{\partial t} = \left(\boldsymbol{A} - k^2\boldsymbol{D}\right)\boldsymbol{y}, \tag{3}$$

where

$$\boldsymbol{y} = \begin{pmatrix}\tilde{u}\\ \tilde{v}\\ \tilde{w}\end{pmatrix}, \qquad \boldsymbol{A} = \begin{pmatrix} f_u & f_v & f_w\\ g_u & g_v & g_w\\ h_u & h_v & h_w\end{pmatrix}, \qquad \boldsymbol{D} = \begin{pmatrix} d_1 & 0 & 0\\ 0 & d_2 & 0\\ 0 & 0 & d_3\end{pmatrix},$$

and k is the wavenumber from Fourier transform $u(x,t) = \int \tilde{u}(k,t)e^{-ikx}\,dx$. All $d_i > 0$. We assume that, without the diffusion term, the steady-state is stable, that is all the eigenvalues of the stability matrix $\boldsymbol{A}$ having negative real parts. Diffusion-driven instability is the problem of finding the conditions on $\boldsymbol{A}$, k, and $\boldsymbol{D}$ such that with the presence of diffusion (i.e., nonzero ds), the matrix $\boldsymbol{A}(k) = \boldsymbol{A} - k^2\boldsymbol{D}$ has an eigenvalue with positive real part.

2. STABILITY AND NEGATIVE DEFINITENESS

We shall call a real matrix, $\boldsymbol{A}$, *stable* if all its eigenvalues have negative real parts, and call it *negative definite* if the quadratic form $\boldsymbol{y}^\top \boldsymbol{A}\boldsymbol{y}$ is negative for all real $\boldsymbol{y} \neq \boldsymbol{0}$.

A stable matrix $\boldsymbol{A}$ does not imply negative definite. A simple example is

$$\begin{pmatrix} -3 & -4.1\\ -2 & -3\end{pmatrix}.$$

The negative definiteness, however, is a sufficient condition for stability. To show this, let nonzero $\{\boldsymbol{z} : z_k = \mu_k + i\omega_k,\ k = 1,2,3\}$ be a complex eigenvector of matrix $\boldsymbol{A}$ associated with complex eigenvalue λ: $\boldsymbol{A}\boldsymbol{z} = \lambda\boldsymbol{z}$. Then,

$$\begin{aligned}\left(\lambda + \overline{\lambda}\right)\sum_{\ell=1}^{3}\|z_\ell\|^2 &= \sum_{\ell,k=1}^{3}\left(\overline{z_\ell}a_{\ell k}z_k + z_\ell\overline{a_{\ell k}z_k}\right)\\ &= 2\sum_{\ell,k=1}^{3}\left(a_{\ell k}\mu_\ell\mu_k + a_{\ell k}\omega_\ell\omega_k\right)\\ &= 2\left(\boldsymbol{\mu}^\top \boldsymbol{A}\boldsymbol{\mu} + \boldsymbol{\omega}^\top\boldsymbol{A}\boldsymbol{\omega}\right)\\ &< 0,\end{aligned}$$

which implies $\mathrm{Re}[\lambda] < 0$.

If matrix $A(0)$ is negative definite, then it is easy to show that $A(k)$ is also negative definite

$$yA(k)y = yA(0)y - k^2 \sum_{i=1}^{3} d_i x_i^2 < yA(0)y < 0,$$

for all real $y \neq 0$. Therefore, a simple, necessary condition for diffusion-driven instability is that A is stable but not negative definite. In practice, it is easy to verify the negative definiteness of a matrix A, which is equivalent to the symmetric matrix $A + A^\top$ being negative definite, with a sufficient and necessary condition [3]

$$a_{11} < 0, \qquad \begin{vmatrix} 2a_{11} & a_{12} + a_{21} \\ a_{21} + a_{12} & 2a_{22} \end{vmatrix} > 0, \qquad \det\left[A + A^\top\right] < 0.$$

Results in this section can be generalized to reaction diffusion systems with any number of species $n > 3$.

3. MATRIX A AND ITS COFACTORS

The condition presented above is simple, but often not tight enough. The stability of nonsingular, 3×3, matrix

$$A = \begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix},$$

can be determined in the usual way using the Routh-Hurwitz criteria on the characteristic equation. This is best understood in terms of the invariants of the matrix and of its inverse matrix

$$A^{-1} = \frac{1}{\det[A]} \begin{pmatrix} M_{11} & M_{21} & M_{31} \\ M_{12} & M_{22} & M_{32} \\ M_{13} & M_{23} & M_{33} \end{pmatrix},$$

where M_{ij} is the cofactor of a_{ij} and matrix $[M_{ij}]$ is the adjunct of A. If matrix A is stable, so is its inverse A^{-1}. The characteristic equation for A is

$$\lambda^3 + p_2\lambda^2 + p_1\lambda + p_0 = 0,$$

where

$$p_2 = -\operatorname{tr}[A], \qquad p_1 = M_{11} + M_{22} + M_{33}, \qquad p_0 = -\det[A].$$

The Routh-Hurwitz conditions are

(I) $\det[A] = -p_0 < 0, \iff \det[A^{-1}] < 0$;
(II) $\operatorname{tr}[A] = -p_2 < 0$;
(II′) $\operatorname{tr}[A^{-1}] < 0$, that is, $p_1 = M_{11} + M_{22} + M_{33} > 0$;
(III) $q = \operatorname{tr}[A] \cdot (M_{11} + M_{22} + M_{33}) - \det[A] = -p_1p_2 + p_0 < 0$, that is, $\operatorname{tr}[A]\operatorname{tr}[A^{-1}] > 1$.

One sees that these conditions possess a symmetry with respect to A and A^{-1}. Contradiction of any one of the above conditions implies the existence of an eigenvalue with positive real part, hence, instability.

The $-k^2 D$ term in (3) never leads to a contradiction with (II).

We now show that if all $a_{ii} < 0$ and $M_{ii} > 0$, then (I) will never be contradicted. We have

$$d\left(\det[A]\right) = M_{11}da_{11} + M_{22}da_{22} + M_{33}da_{33},$$

in which $da_{ii} < 0$. If both a_{22} and $a_{33} < 0$, then $M_{11}(k) > M_{11}(0) > 0$. Similarly, $M_{ii}(k) > M_{ii}(0) > 0$, $(i = 1, 2, 3)$. Hence, $d(\det[A]) < 0 \Longrightarrow \det[A(k)] < \det[A(0)] < 0$.

For (III), we note

$$\begin{aligned} dq = &\left\{M_{22} + M_{33} + \mathrm{tr}^2[\boldsymbol{A}] - a_{11}\,\mathrm{tr}[\boldsymbol{A}]\right\} da_{11} \\ &+ \left\{M_{11} + M_{33} + \mathrm{tr}^2[\boldsymbol{A}] - a_{22}\,\mathrm{tr}[\boldsymbol{A}]\right\} da_{22} \\ &+ \left\{M_{11} + M_{22} + \mathrm{tr}^2[\boldsymbol{A}] - a_{33}\,\mathrm{tr}[\boldsymbol{A}]\right\} da_{33}, \end{aligned}$$

in which all the coefficients of the da_{ii} are positive $\forall\, k$ and $d_i > 0$. Hence, $q(k) < 0$.

Thus, we have shown that $a_{ii} < 0$ and $M_{ii} > 0$, $(i = 1, 2, 3)$ are sufficient conditions for $\boldsymbol{A}(k)$ being stable. We, therefore, have the following new criteria for reaction systems with three species: *A necessary condition for diffusion-driven instability is that either the largest diagonal elements of* $\boldsymbol{A}$ *is greater than zero; or the smallest diagonal cofactors of* $\boldsymbol{A}$ *is less than zero.*

This condition is clearly stronger than that of the above section. A matrix $\boldsymbol{A}$ satisfying this condition is necessarily not negative definite, since a negative definite matrix has all its diagonal elements negative and diagonal cofactors positive.

4. EXPLICIT CONDITIONS FOR d_i AND k: THE SUFFICIENT CONDITION

In the above section, a necessary condition is obtained for matrix $\boldsymbol{A}$ irrespective of k and $\boldsymbol{D}$. We now show that the condition is, in fact, also sufficient. We demonstrate this by finding appropriate k and d_i.

The characteristic equation for matrix $\boldsymbol{A}(k)$ is

$$\lambda^3 + p_2\left(k^2\right)\lambda^2 + p_1\left(k^2\right)\lambda + p_0\left(k^2\right) = 0,$$

where

$$\begin{aligned} p_0\left(k^2\right) &= d_1 d_2 d_3 k^6 - \left(a_{11} d_2 d_3 + a_{22} d_1 d_3 + a_{33} d_1 d_2\right) k^4 \\ &\quad + \left(d_1 M_{11} + d_2 M_{22} + d_3 M_{33}\right) k^2 - \det[\boldsymbol{A}], \\ p_1\left(k^2\right) &= \left(d_1 d_2 + d_2 d_3 + d_1 d_3\right) k^4 - \left[d_1\left(a_{22} + a_{33}\right) + d_2\left(a_{11} + a_{33}\right) + d_3\left(a_{11} + a_{22}\right)\right] k^2 \\ &\quad + \left(M_{11} + M_{22} + M_{33}\right), \\ p_2\left(k^2\right) &= \left(d_1 + d_2 + d_3\right) k^2 - \mathrm{tr}[\boldsymbol{A}]. \end{aligned}$$

Let us focus on $p_0(k^2)$. According to Routh-Hurwitz criteria, $p_0(k^2) < 0$ is a sufficient condition for matrix $\boldsymbol{A}(k)$ being unstable. Let us assume that $M_{33} < 0$. If we choose $d_1 = d_2 = 0$ and $k^2 d_3 > (\det[\boldsymbol{A}])/(M_{33}) > 0$, then

$$p_0\left(k^2\right) = d_3 M_{33} k^2 - \det[\boldsymbol{A}] < 0.$$

In practical terms, this indicates that to generate a pattern, one needs the diffusion coefficients for species u and v significantly smaller than that of w.

If all diagonal cofactors are positive but $a_{11} > 0$, then we choose $d_2 = d_3 = 1$ and $d_1 = 0$. Therefore,

$$p_0\left(k^2\right) = -a_{11} k^4 + \left(M_{22} + M_{33}\right) k^2 - \det[\boldsymbol{A}],$$

which is negative for sufficiently large k. In this case, one needs the diffusion coefficients for species u significantly smaller than that of v and w.

We, therefore, also have the following criteria for reaction systems with three species: *A sufficient condition for diffusion-driven instability is that either*

(i) *the largest diagonal elements of* $\boldsymbol{A}$ *is greater than zero with corresponding diffusion coefficient very small; or*

(ii) *the smallest diagonal cofactors of* $\boldsymbol{A}$ *is less than zero with corresponding diffusion coefficient very large.*

In the case of activator-inhibitor type reactions with two species, it is generally accepted that the instability requires the diffusion coefficient of the inhibitor being larger than that of the activator. The above sufficient condition can be viewed as a natural generalization to three species with two scenarios:

(i) two inhibitors and one activator and
(ii) one inhibitor and two activators.

Meeting the condition for diffusion-driven instability, however, is not a guarantee for stationary pattern formation. The behavior of the reaction-diffusion system cannot be completely determined from the simple linear analysis for instability near the homogeneous steady-state. The global spatiotemporal behavior of a reaction diffusion system, including traveling front, depends on other factors, namely, the nonlinearity in the equation, the boundary conditions, and the geometry in the case of multidimensional space. These questions have to be addressed with a nonlinear analysis.

5. AN EXAMPLE OF APPLICATIONS

Let us consider the three-species reaction system known as Oregonator [4] which models the widely studied Belousov-Zhabotinsky reaction which has been considered a pedagogical paradigm for biological pattern formation:

$$\begin{gathered} A+Y \xrightarrow{k_1} X+P, \qquad X+Y \xrightarrow{k_2} 2P, \qquad A+X \xrightarrow{k_3} 2X+2Z, \\ 2X \xrightarrow{k_4} A+P, \qquad Z \xrightarrow{k_5} fY. \end{gathered} \tag{4}$$

The kinetic equations for X, Y, and Z after appropriate nondimensionalization [5]:

$$\epsilon\frac{dx}{dt} = qy - xy + x(1-x), \tag{5}$$

$$\delta\frac{dy}{dt} = -qy - xy + 2fz, \tag{6}$$

$$\frac{dz}{dt} = x - z, \tag{7}$$

where $\epsilon = k_5/(k_3[A])$, $\delta = k_4k_5/(k_2k_3[A])$, $q = k_1k_4/(k_2k_3)$, and f are all positive constants.

The stability matrix from linearization about the positive steady-state (x_s, y_s, z_s) is

$$\boldsymbol{A} = \begin{pmatrix} \dfrac{1-2x_s-y_s}{\epsilon} & \dfrac{q-x_s}{\epsilon} & 0 \\ -\dfrac{y_s}{\delta} & -\dfrac{x_s+q}{\delta} & \dfrac{2f}{\delta} \\ 1 & 0 & -1 \end{pmatrix} \tag{8}$$

and its cofactors given by

$$\boldsymbol{A}^{-1} = \frac{1}{\det[\boldsymbol{A}]} \begin{pmatrix} \dfrac{x_s+q}{\delta} & \dfrac{q-x_s}{\epsilon} & \dfrac{2f\,(q-x_s)}{\epsilon\delta} \\ \dfrac{2f-y_s}{\delta} & -\dfrac{1-2x_s-y_s}{\epsilon} & -\dfrac{2f\,(1-2x_s-y_s)}{\epsilon\delta} \\ \dfrac{x_s+q}{\delta} & \dfrac{q-x_s}{\epsilon} & \dfrac{2qy_s-(q+x_s)\,(1-2x_s)}{\epsilon\delta} \end{pmatrix}, \tag{9}$$

in which, with chemically realistic parameter estimates, $(1-2x_s+y_s) < 0$ [2]. Therefore, the condition for diffusion-driven instability of Oregonator is

$$2qy_s - (q+x_s)\,(1-2x_s) < 0,$$

in which

$$x_s = \frac{(1-q-2f) + \sqrt{(1-q-2f)^2 + 4q(1+2f)}}{2},$$

and

$$y_s = \frac{2fx_s}{q+x_s}.$$

With some tedious but elementary algebra, we have

$$\left(1+q-6f+8f^2+8fq\right)x_s + q\left(1+q-2f-8f^2+4fq\right) < 0. \tag{10}$$

Figure 1 shows the (f,q) parameter space in which the reaction will be diffusionally unstable. For small $q \approx 8 \times 10^{-4}$ (a value based on experiment), we have

$$2\left(1+4f-20f^2\right)q - (4f-1)(2f-1)^2 < 0 \tag{11}$$

and

$$4f(1+2f)^2q - \left(1+4f-4f^2\right)(2f-1)^2 < 0, \tag{12}$$

which yield approximately $0.25 < f < 1.207$. Under this condition, a sufficiently large diffusion for Z (i.e., the catalyst cerium Ce^{+++}), with respect to those of X and Y, will lead to diffusion-driven instability. This result has been known by numerical computations from previous work [6]. Beyond the linear analysis, in the Oregonator, both traveling front and stationary patters have been observed [5–7].

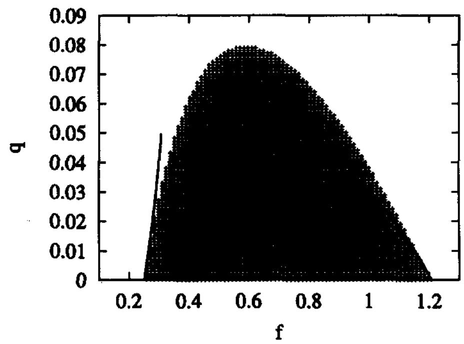

Figure 1. The shaded region is for the parameters (f,q) at which the chemical reaction (4) can exhibit diffusion-driven instability. For small realistic q, $0.25 < f < 1.207$. The lines, given by equations (11) and (12), are asymptotic approximations for the boundary with small q.

It is interesting that the positive steady-state of the Oregonator losses its stability when $1.000 < f < 1.207$ and with sufficient small ϵ and δ [2,4]. Our result, however, indicates that when both ϵ and δ are large enough and the steady-state is stable, it can also loss its stability (bifurcation) due to diffusion. As pointed out in [4], the stoichiometric factor f plays a significant role in oscillation and spatial pattern formation of Belousov-Zhabotinsky reaction [8].

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