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Stability analysis of dissipative structures in a nonlinear diffusion-reaction problem

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Topological-degree and Lyapunov-potential techniques are used to study the stability of nonlinear dissipative structures found in a problem involving autocatalysis and the Langmuir-Hinshelwood adsorption law.

I. INTRODUCTION

Recently there has been an extraordinary impetus in the study of nonlinear nonequilibrium phenomena (Refs. 1, 2, and for a discussion of physical and nonphysical problems, see, for instance, Ref. 3). Most of the works refer to the onset of new phases, usually with a higher degree of ordering and synergesis; these nonequilibrium phases arise or bifurcate from some primary, rather disordered, basic states that become unstable at well-defined critical values of certain parameters. Very little is known, however, about the stability of the ordered branches that exhibit either time periodic (limit cycle) behavior, nonuniform distribution of concentrations, temperature, etc., or ordered convective motions (Bénard cells, Taylor vortices, etc.). In a recent review Normand, Pomeau, and Velarde[2] have discussed the useful perturbative approach originally due to Landau and Hopf for hydrodynamic convective motions (see also Refs. 4 and 5). In the present paper we use more refined, and nonperturbative mathematical techniques to dicsuss the stability of ordered phases (dissipative structures) in diffusion-reaction systems. These techniques (topological degree of a mapping, and Lyapunov's "potential" method) seems to us of useful and wide applicability in nonlinear problems that at present belong to general physics.

If one wishes to consider a realistic model problem containing a minimal set of the most relevant physicochemical components, a tentative case is one with the following items: (i) autocatalysis, of which a typical step involving binary collisions only is $X+Y\rightarrow 2X$, in which X and Y denote concentrations of species X and Y; (ii) a saturation-adsorption law such as the Langmuir-Hinshelwood law in heterogeneous catalysis,[6] which mathematically is just the functional law called Holling's law in ecology, and in enzyme-controlled biophysical-chemistry, the Michaelis-Menten law. In the simplest case it corresponds to a reaction rate like $X/(1+qX)$, where q denotes some characteristic parameter that yields the strength of the saturation; and (iii) diffusion of matter, should energy dissipation and convective motions be disregarded. Lastly, for an isothermal process with two species participating the simplest case corresponds to one with large separation in the diffusion constants.

Under the circumstances just described above the model problem is contained in the following physicochemical scheme:

$$A\rightarrow Y,\quad X+Y\rightarrow 2X,\quad X\xrightarrow{S}P\ ,$$

in which A and P also denote concentrations of products that we keep constant in the simplest case. S accounts for the saturation law. Various aspects of the mathematical analysis of such a physicochemical scheme have been described in Refs. 7 and 8, and under the approximations (i)–(iii) the most simple case is a one-dimensional line process. It leads to an evolution and eigenvalue equation which in dimensionless form is

$$\frac{\partial X}{\partial t}=D\frac{\partial^2 X}{\partial r^2}+Y_s X-\frac{X}{1+qX}\ , \tag{1.1a}$$

$$X(0)=X(1)=\text{const} \tag{1.1b}$$

on the interval $[0,1]$. D is the mass diffusion constant of X (we have taken $D_x\ll D_y$, $D_y\rightarrow\infty$). The term Y_sX comes from the autocatalytic step with Y_s being a fixed value ($D_y\rightarrow\infty$). We shall take this constant equal to the fixed value at the steady state when there is no mass diffusion: $Y_s=1-qA$. On the other hand, to this value Y_s corresponds a steady-state value to $X_s=A/(1-qA)$ that we take as the boundary value in Eq. (1.1b) (for details, see, Refs. 7 and 8). The change of variables

$$X=A/(1-qA)+u \tag{1.2}$$

transforms Eqs. (1.1a) and (1.1b) into the following Schrödinger-like equation:

$$\frac{1}{D}\frac{\partial}{\partial t}u=\Delta u+f(u)\ , \tag{1.3a}$$

$$f(u)=\frac{1}{D}\left(A+(1-qA)u-\frac{A+(1-qA)u}{1+q(1-qA)u}\right)\ , \tag{1.3b}$$

$$u(0)=u(1)=0 \ , \qquad (1.3c)$$

in which Δ denotes the Laplacian here in dimension 1.

In the following we shall denote by $u(x, t, \phi)$ the solution to Eqs. (1.3a)–(1.3c) with initial condition $u(x, 0, \phi)=\phi(x)$. The steady-state solution $u_s(x)$ to Eqs. (1.3a)–(1.3c) originate in the eigenvalue problem

$$\Delta u_s + f(u_s) = 0 \ , \qquad (1.4a)$$

$$u_s(0)=u_s(1)=0 \ . \qquad (1.4b)$$

The functions $u(\cdot, t, \phi)$ and $u_s(\cdot)$ belong to a space B of functions defined on $[0,1]$ with continuous differentials, that vanish on the boundaries of the interval.

Let $u(x, v_0)$ be a formal solution to Eq. (1.4a) obeying the initial data $u(0, v_0)=0$ and $\nabla u(0, v_0)=v_0$. Then there is an obvious first integral to (1.4a)

$$E=\tfrac{1}{2}\,[\nabla u(x, v_0)]^2 + F[u(x, v_0)] \ , \qquad (1.5a)$$

with

$$F(u)=\int_0^u f(\xi)\,d\xi = \frac{1}{D}\left(\frac{(1-qA)u^2}{2} - \frac{u(1-qA)}{q} + \frac{1}{q^2}\ln|1+q(1-qA)u|\right) . \qquad (1.5b)$$

It appears that for a given $k \geqslant 0$, $u(x, v_0)$ will be a solution to Eqs. (1.4a) and (1.4b) if any of the following conditions are satisfied:

$$\text{(i)}\ (k+1)T_1 + kT_2 = 1 \ , \qquad (1.6a)$$

$$\text{(ii)}\ kT_1 + (k+1)T_2 = 1 \ , \qquad (1.6b)$$

$$\text{(iii)}\ (1+k)(T_1+T_2) = 1 \ , \qquad (1.6c)$$

in which

$$T_{1,2}=\sqrt{2}\int_0^{\xi_{1,2}} \{E[u(x, v_0)] - F(\xi)\}^{-1/2}\,d\xi \ , \qquad (1.7a)$$

$$F(\xi_{1,2})=\tfrac{1}{2}v_0^2 \ , \qquad (1.7b)$$

and k is an integer: $0, 1, 2, \ldots$. Figures 1 and 2 provide a qualitative sketch of the multiple steady-state solutions for the values $q=2.0$, $D=0.002$, and $A \in (0, q^{-1})$.

The solutions $u_s(x)=u(x, v_0)$, $X \in [0,1]$ depend on the parameter v_0, and $v_0=0$ yields the homogeneous steady-state solution.

II. STABILITY OF DISSIPATIVE STRUCTURES

The linear stability of steady states $u_s(x)$ is related to the eigenvalue problem

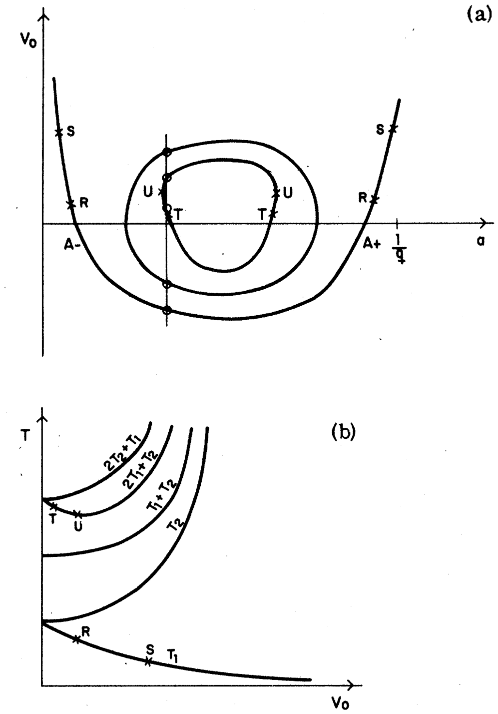

FIG. 1. (a) Qualitative sketch of the steady-state solutions parametrized by $v_0=v_0(A)$; $q=2.0$, $D_X=0.002$. (b) Qualitative sketch of T as a function of v_0 for the same values of q and D_X. The branches RS and TU in both cases are in correspondence with each other.

$$\left(\Delta + \frac{\partial f}{\partial u}[u_s(x)]\right)\psi_\eta = \lambda_\eta \psi_\eta \ . \qquad (2.1)$$

The origin is asymptotically stable for

$$A \in (0, A_-) \cup (A_+, 1/q) \ , \qquad (2.2a)$$

$$A_\pm = 0.5 \pm 0.5(1-4\pi^2 D)^{1/2} \ , \qquad (2.2b)$$

and it is unstable for $A \in (A_-, A_+)$. In the case $D > \frac{1}{4}$ the origin is asymptotically stable for $A \in (0, 1/q)$ and no other steady state exists.

A different matter is the stability of nonuniform solutions, and we shall use two methods to study the problem: (i) method of topological degree, and (ii) Lyapunov's potential method (direct method).

III. APPLICATION OF THE TOPOLOGICAL-DEGREE METHOD

The solution $u_s(x)$ to Eqs. (1.4) can be written

$$u_s(x) = -\int_0^1 G(x, s) f[u_s(\xi)]\,d\xi \equiv H(A, q, D)u_s(x) \ , \qquad (3.1)$$

where H denotes a nonlinear operator on B, and $G(x, \xi)$ is the Green's function corresponding to

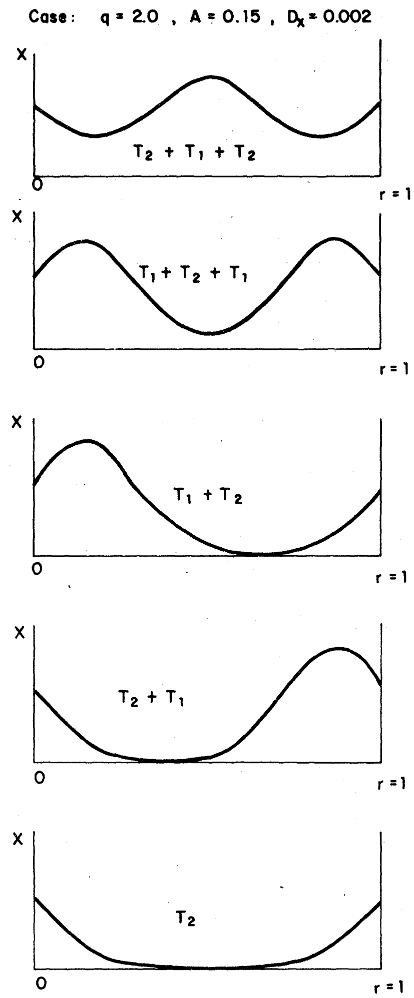

FIG. 2. Five steady-state solutions that appear at $q=2.0$, $D_X=0.002$, and $A=0.15$. They correspond to the open circles shown in Fig. 1(a).

(1.4). The steady states are the fixed points of H. We shall associate an index γ related to the rotation of H on an infinitesimal sphere around $u_s(x)$ on B [for details, see, Refs. 2 (Appendix), 12 (Appendix), 13.

Let L be the linearization of H around $u_s(x)$. Then $\gamma=(-)^\beta$, where β is the sum of multiplicities of the eigenvalues μ_η to the problem

$$L\phi_\eta=\mu_\eta^{-1}\phi_\eta \ , \qquad (3.2)$$

in the segment $[0,1]$. Thus, in our case we have

$$\Delta\phi_\eta=-\frac{\mu_\eta}{D}\left((1-qA)-\frac{(1-qA)^2}{[1+q(1-qA)u_s(x)]^2}\right)\phi_\eta \ , \qquad (3.3a)$$

$$\phi_\eta(0)=\phi_\eta(1)=0 \ . \qquad (3.3b)$$

For the trivial fixed point, $u_s\equiv 0$, it is easily found that for given q and D, γ has the following values:

$$\gamma=+1 \quad \text{if } A_k^{\text{even}}<A<A_k^{\text{odd}} \ ,$$

$$\gamma=-1 \quad \text{if } A_k^{\text{odd}}<A<A_{k+2}^{\text{even}} \ ,$$

where $0<A<1/q$, and

$$qA_k^{\text{even}}(1-qA_k^{\text{even}})=D\pi^2k^2 \quad , k=0,2,4,\ldots$$

$$qA_k^{\text{odd}}(1-qA_k^{\text{odd}})=D\pi^2(k+1)^2 \quad , k=0,2,4,\ldots .$$

Thus, the origin changes its topological degree at the bifurcation points.

The degrees of the remaining stationary solutions follow from the fact that the sum of all degrees of the steady states to the left of a bifurcation point equals the corresponding sum for those to the right, i.e., for given q and D the operators $H(A,q,D)$ and $H(A',q,D)$ are homotopically equivalent on the sphere that encloses all steady states arising between a and a'.

Shown in Fig. 3 are the topological degrees of the several branches found with variable A, and given $q=2.0$ and $D=0.002$.

Luss and Amudson (Refs. 10, 11, and see also 12, p. 90) have shown that a necessary and sufficient condition for the eigenvalues λ_η of Eq. (2.1) to be negative is that Eq. (3.2) would not have eigenvalues in the closed interval $[0,1]$. Thus, all solutions $u_s(x)$ with (-1) are unstable, though not all $u_s(x)$ with degree $(+1)$ are asymptotically stable.

We speak of stability or instability with respect to the norm of the Banach space B. Given $h\in B$, $\|h\|=\sup\{|\nabla h(x)|\}$ for $x\in[0,1]$, thus $u_s(x)$ is said to be asymptotically stable if for given $\epsilon>0$ there exists $\delta>0$ such that if

$$\|\phi-u_s(x)\|=\sup[\nabla\phi-\nabla u_s(x)]<\delta$$

for $x\in[0,1]$ it follows that

$$\|u(x,t,\phi)-u_s(x)\|=\sup\{|\nabla u(x,t,\phi)-\nabla u_s(x)|\}\to 0$$

for $x\in[0,1]$ and $t\to\infty$. For any function $\phi\in B$

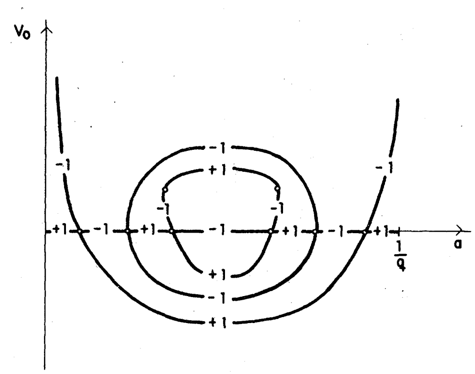

FIG. 3. Topological degree or index corresponding to the various steady-state solutions for $q=2.0$ and $D_X=0.002$.

$$\sup\{|\phi(x)|\} \leq C \sup\{|\phi'(x)|\}$$

for $x \in [0,1]$, in which C is a constant that does not depend on the uniform convergence of $|\nabla u(x,t,\phi) - \nabla u_s(x)|$, ensures the pointwise convergence of the functions themselves.

IV. APPLICATION OF LYAPUNOV'S DIRECT METHOD

From Sec. III it remains to study the stability of solutions with degree +1. This can be achieved with the help of a Lyapunov functional[13] on B

$$V(\phi) = \int_0^1 \left(\frac{1}{2}(\nabla\phi)^2 - F(\phi)\right) dx\ , \qquad (4.1)$$

with F defined by Eq. (1.5b). The functional V has the following properties: (i) For a solution $u(x,t,\phi)$ to Eq. (1.3)

$$\frac{d}{dt} V[u(x,t,\phi)] \leq 0\ . \qquad (4.2)$$

(ii) The equal sign in Eq. (4.2) corresponds to the case of a stationary solution only.

$$\text{(iii)}\ V[u_s(x)+h] - V[u_s(x)] = W(u_s,h) + (\|\|h\|\|^2), \qquad (4.3)$$

where

$$W(u_s,h) = \frac{1}{2}\int_0^1 \left[(\nabla h)^2 - \left(\frac{d}{du} f(u_s(x))\right)h^2\right] dx \qquad (4.4)$$

and

$$\|\|h\|\| = \left(\int_0^1 (\nabla h)^2\, dx\right)^{1/2}\ . \qquad (4.5)$$

Then the stationary solution $u_s(x)$ is asymptotically stable if V has a local minimum at $u_s(x)$ and unstable if V has a local maximum.

Chafee and Infante[9] have proved that for systems defined by Eqs. (1.4) one has the following: (i) if a positive constant $c > 0$ exists such that

$$W(u_s,h) \geq c\,\|\|h\|\|^2 \qquad (4.6)$$

for every $h \in B$, then the solution $u_s(x)$ is asymptotically stable (with the definition given in Sec. III). (ii) $u_s(x)$ is unstable if there exists $h_1 \in B$ such that $W(u_s(x), h_1) < 0$.

The Jacobi equation corresponding to the Lyapunov functional V and solution $u_s(x)$ is[14]

$$Z'' + \frac{\partial f}{\partial u}[u_s(x)]Z = 0\ . \qquad (4.7)$$

Then $\xi \in (0,1]$ is a *conjugate point* when the solution to (4.7) for initial conditions $Z(0)=0$ and $Z'(0)=1$ is such that $Z(\xi)=0$. The utility of Jacobi's equation to study second variations of V around $u_s(x)$ and consequently the stability of $u_s(x)$ comes from the following results: (i) If Eq. (4.7) possesses a conjugate point $\xi \in (0,1]$, then there exists $h_1 \in B$ such that $W(u_s, h_1) < 0$, and u_s is unstable. (ii) If Eq. (4.7) does not posses a conjugate point in (0, 1), then Eq. (4.6) is satisfied and u_s is asymptotically stable.

Nothing is said about the case of 1 being the conjugate point, in which case there is marginal stability.

The solutions to the Jacobi equation (4.7) are obtained by differentiating $u(x, v_0)$ with respect to the v_0 corresponding to $u_s(x)$. The analysis of Eq. (4.7) follows the pattern presented in Ref. 9. We get (i) for a given triplet (q, A, D_x) if there is a solution corresponding to $T = T_2$, it is asymptotically stable; (ii) all other nonuniform steady-state patterns are unstable in points where the slope, $dT/dv_0 > 0$, is positive; and (iii) for the patterns with negative slope, $dT/dv_0 < 0$, nothing can be said using the work of Chafee and Infante as the function $f(u)$ in Eq. (1.4a) does not satisfy their hypothesis; as a matter of fact it does not comply with the condition $f(u)u^{-1} \to 0$ as $(u) \to \infty$.

However, as they have topological degree (−1) they are unstable.

Thus, for given q, D_x we have (i) for $A \in (0, A_-) \cup (A_+, 1/q)$ with $A_\pm$ given the only asymptotically stable steady-state pattern is the homogeneous solution. If $A_\pm$ is not real, the homogeneous solution is asymptotically stable in the region $A \in (0, 1/q)$; (ii) for $A \in (A_-, A_+)$ the solution corresponding to T_2 is the only asymptotically stable pattern.

Figure 4 provides the diagram of stable branches for $q = 2.0$ and $D_x = 0.002$.

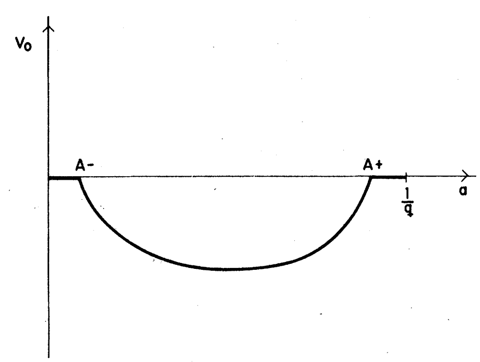

FIG. 4. Plot of the asymptotically stable steady-state branches that appear at different values of A, with $q = 2.0$, and $D_X = 0.002$.

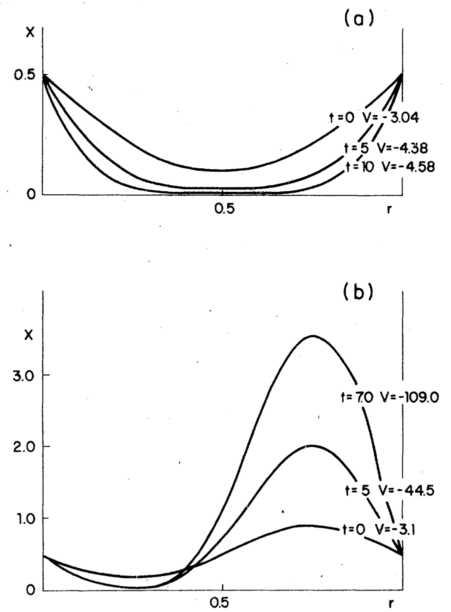

FIG. 5. Spatial nonuniform concentration profiles of reactant X along the line $r(0,1)$. For $A=0.25$, $q=2.0$, and $D_X=2\times10^{-3}$, the only asymptotically stable steady state corresponds to T_2. (a) Gives the time evolution of a concentration profile placed at time $t=0$ in the domain of attraction of T_2. As time goes on the value of the "potential" V decreases to relative minimum $V_m=-4.59$, which corresponds to the steady-state solutions; (b) corresponds to an initial condition outside the domain of attraction of T_2, and the system does not tend to any steady-state solution; rather the potential grows unbounded to $-\infty$.

V. GLOBAL STABILITY

If for a given triplet (q,A,D_x) we have

$$\min[V(h)]=V[u_s^{as}(x)]\ , \tag{5.1}$$

for all $h\in B$, we say that u_s^{as} is globally stable or stable in the whole.

For the values of (q,A,D_x) where the homogeneous solution is asymptotically stable

$$V(u_s^{as})=0\ . \tag{5.2}$$

Whereas, for those values for which the stable solution corresponds to T_2, it is

$$V(u_s^{as})\geq -k\ , \tag{5.3a}$$

with

$$k=A^2/2(1-qA)+A/q+(1/q^2)\ln(1-qA)>0\ . \tag{5.3b}$$

If we take the family of functions $h_c\in B$,

$$h_c(x)=c\sin\pi x\ , \tag{5.4}$$

it follows that

$$V[h_c(x)]=(c^2/4D_x)[\pi^2D_x-(1-qA)]+O(c)\ . \tag{5.5}$$

Thus, for $(1-qA)>\pi^2D_x$ the V function can be made as small as we please. It suffices to take c as large as we need. Consequently, the corresponding asymptotically stable steady-state solution cannot be stable in the whole (globally stable). To establish this property it suffices to give as an initial condition to Eq. (1.3), a function that yields values of V smaller than $-k$, to prevent the function $u(x,t,\phi)$ from evolving to the solution $u_s^{as}(x)$ for this would contradict relation (4.2). Nothing can be said, however, when $(1-qA)<\pi^2D_x$. Thus, at least for some range of values of the parameters, there is no global stability to the solution. Finally, all that remains is to mention that computer runs confirm all conclusions presented above. Besides, Fig. 5 shows the evolution of the solution $u(x,t,\phi)$ to Eqs. (1.3) with $q=2.0$, $D=0.002$, and $A=0.25$. For these values of the parameters the asymptotically stable steady state T_2 is not globally stable and the solution $u(x,t,\phi)$ goes or does not go to T_2 depending on the initial condition $\phi(x)$; in both cases the Lyapunov function decreases.

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[1]G. Nicolis and I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Interscience, New York, 1977).

[2]Ch. Normand, Y. Pomeau, and M. G. Velarde, Rev. Mod. Phys. 49, 581 (1977).

[3]H. Haken, *Synergetics: An Introduction* (Springer, Berlin, 1977).

[4]J. F. G. Auchmuty and G. Nicolis, Bull. Math. Biol. 37, 323 (1975).

[5]M. Herschkowitz-Kaufman, Bull. Math. Biol. 37, 589 (1975).

[6]R. Aris, *The Mathematical Theory of Diffusion and Reaction in Permeable Catalysts* (Clarendon, Oxford, 1975).

[7]J. L. Ibáñez, V. Fairén, and M. G. Velarde, Phys. Lett. A 58, 364 (1976).

[8]J. L. Ibáñez and M. G. Velarde, J. Math Phys. 19, 151 (1978).

[9]N. Chafee and E. F. Infante, Appl. An. 4, 17 (1974).

[10]D. Luss, N. R. Amundsm, Can. J. Chem. Eng. 45,

341 (1967).

[11]D. Luss, M. E. Ervin, Chem. Eng. Sci. 27, 315 (1972).

[12]G. R. Gavalas, *Nonlinear Differential Equations of Chemically Reacting Systems* (Springer, Berlin, 1968).

[13]M. A. Krasnosel'skii, *Topological Methods in the Theory of Nonlinear Integral Equations* (Pergamon, New York, 1964).

[14]I. M. Gelfand and S. V. Fomin, *Calculus of Variations* (Prentice-Hall, Englewood Cliffs, 1963).