

Dynamics of Linear and Nonlinear Autocatalysis and Competition

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Abstract

The aim of this contribution is to analyse some conventional kinetic equations of reaction mechanisms which include autocatalytic steps. In case of first-order or linear autocatalysis represented by the elementary step



the dynamics is very simple: we find a uniquely defined asymptotically stable steady state under almost all conditions. Higher order autocatalytic processes, e.g. the trimolecular elementary step



which has been used in several investigations of chemical non-equilibrium phenomena (SCHLÖGL [1,2]; PRIGOGINE and LEFEVER [3]) lead to much richer dynamics. In higher dimensional systems we encounter limit cycles and chaotic attractors.

1. Introduction

Competition, selection and permanence [4] are particularly important notions in the theory of biological evolution. Recently, experimental investigations were undertaken [5] which allow to study these phenomena in molecular systems far from equilibrium. Polynucleotides, in particular RNA and enzymes of simple bacteriophages are used in these experiments. Polynucleotides have an intrinsic capability to act as autocatalysts built into their molecular structures. It is of particular interest, therefore, to find model systems which allow to control the distance from the equilibrium state and which are simple enough for extensive qualitative analysis. A combination of an autocatalytic reaction step, a degradation reaction and a recycling process was found to represent an appropriate mechanism for this purpose. For reasons which will

become clear soon we distinguish between linear (1) and second-order (2) autocatalysis. Although trimolecular reactions like (2) are commonly excluded in chemistry as acceptable elementary steps, they may appear in overall kinetics and particularly in biochemical reactions involving biological macromolecules.

2. Linear Autocatalysis

The single-step reactions (1) and (2) are well understood and easy to analyze - they lead essentially to the logistic or to the hypologistic equation respectively. As indicated above we embed the two elementary steps in many-step mechanisms which keep the autocatalytic reaction off equilibrium in a controllable manner. The autocatalytic reaction is coupled to a first-order degradation process and a recycling reaction driven by external energy. We consider the open system



As indicated in (3c) the rate constant of the recycling reaction $g(E)$ is determined by an external energy source E . An example of such a process is a photochemical reaction using a light source.

The dynamics of the mechanism (3) is described by the differential equations

$$\dot{a} = gb + f'x^2 - fax \quad (4a)$$

$$\dot{b} = dx - (d'+g)b \quad (4b)$$

$$\dot{x} = fax + d'b - f'x^2 - dx \quad (4c)$$

wherein we denote the concentrations of A, B and I by a , b and x respectively. Obviously, the total concentration of all substances is constant:

$$a + b + x = c_0 = \text{const.} \quad (5)$$

Hence, we are left with a two-dimensional system, defined on the state space

$$S = \{(a, b, x) \in \mathbb{R}_+^3 : a+b+x = c_0\}$$

which is positively invariant under the flow of (4). The fixed points

of (4) are the solutions of

$$d\bar{x} - (d'+g)\bar{b} = 0 \tag{6a}$$

$$f(c_0 - \bar{b} - \bar{x})\bar{x} + d'\bar{b} - f'\bar{x}^2 - d\bar{x} = 0. \tag{6b}$$

They are given by

$$P_0 = (\bar{b}_0, \bar{x}_0) = (0, 0) \tag{7a}$$

$$\text{and } P_1 = (\bar{b}_1, \bar{x}_1) = \frac{fc_0(d'+g) - dg}{(f+f')(d'+g) + fd} \left(\frac{d}{d'+g}, 1 \right). \tag{7b}$$

If $c_0 < c_{\text{crit}} = \frac{dg}{f(d'+g)}$, P_1 is outside of the physically relevant state space S ; at $c_0 = c_{\text{crit}}$, P_1 coincides with P_0 and enters S for $c_0 > c_{\text{crit}}$. Local stability analysis performed by computing the linearization of (4) at P_0 and P_1 shows that P_0 is a sink for $c_0 < c_{\text{crit}}$, and P_1 is asymptotically stable whereas P_0 is a saddle for $c_0 > c_{\text{crit}}$.

In order to globalize these results we use the Bendixson-Dulac test to exclude the existence of limit cycles (see ANDRONOV et al. [6]: we apply the Dulac-function x^{-1} to the vector field (4) and obtain

$$(\dot{b}) = d - (d'+g) \frac{b}{x} \tag{8a}$$

$$\text{and } (\dot{x}) = f(c_0 - b - x) + d' \frac{b}{x} - f'x - d \tag{8b}.$$

Equation (8) obviously has a strictly negative divergence on S . Therefore the flow (8) is area contracting, and periodic orbits are impossible. Then, the Poincaré-Bendixson theorem implies that the stable stationary solutions P_0 or P_1 respectively are indeed globally stable: every solution starting in S converges to the stable fixed point (see figs. 1a and 1b).

For $d'=0$, global stability of P , can be proved also by means of the Lyapunov function

$$V = f(b - \bar{b}_1)^2 + 2d(x - \bar{x}_1 \log x), \text{ where}$$

(\bar{b}_1, \bar{x}_1) are the coordinates of P_1 given by (7b)

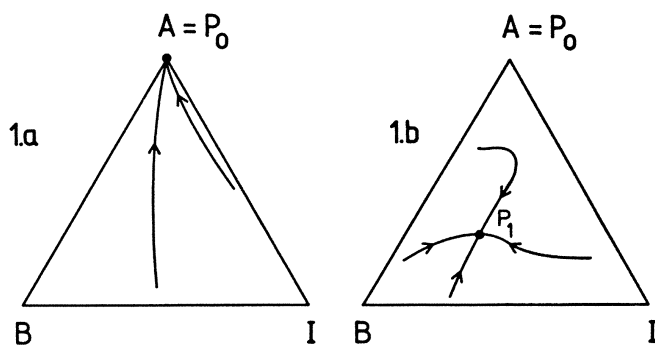


Figure 1. Phase portrait of equation (4) (a: $c_0 < c_{\text{crit}}$ and b: $c_0 > c_{\text{crit}}$).

The reaction mechanism (3) thus admits only two qualitatively different types of dynamical behaviour. The plane of external parameters (g, c_0) is split into two regions corresponding to the stability of either P_0 or P_1 (see Fig.2).

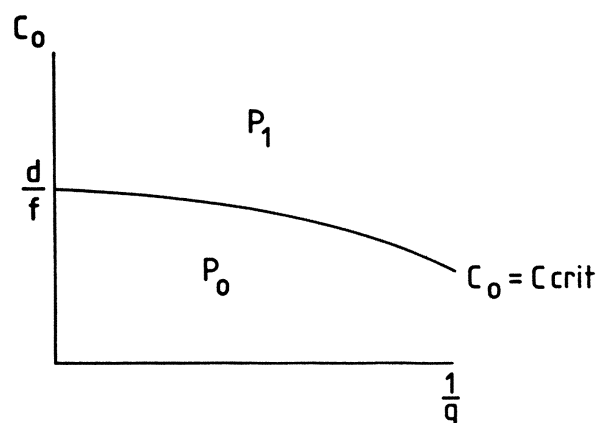
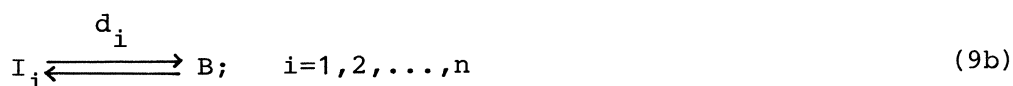
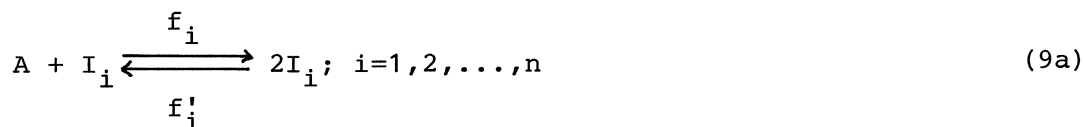


Figure 2. The plane of external parameters (g, c_0) of equation (4) is split into two regions corresponding to the stability of either P_0 or P_1 . The two regions are separated by the curve $c_0 = c_{crit}$.

If all reactions are irreversible ($f' = d' = 0$), a condition which is approximately true for most biochemical and biological systems, the concentration of A at the stationary solution P_1 and the critical value c_{crit} coincide and are independent of g : $\bar{a} = c_{crit} = \frac{d}{f}$. The irreversible autocatalytic processes consume the reactant A to the ultimate limit: P_1 would become unstable if the concentration of A went below c_{crit} .

3. Competition between First-Order Autocatalytic Reactions

Now, we consider a network of $2n+1$ reactions: n first-order autocatalytic reaction steps followed by n first-order degradation processes which are coupled to a common irreversible recycling reaction which again is controlled from outside and which allows to drive the system far off equilibrium.



This reaction scheme may be considered as competition between n autocatalysts for the common source of material A for synthesis. In the case of reversibility of reactions (9a) and (9b) there is a

thermodynamic restriction of the choice of rate constants because of the uniqueness of the thermodynamic equilibrium (by \bar{b} and \bar{a} we denote equilibrium concentrations):

$$\frac{\bar{b}}{\bar{a}} = \frac{f_i d_i}{f_i' d_i'}; \quad i=1,2,\dots,n.$$

The dynamics of mechanism (9) is described by the differential equation (concentrations of I_i are denoted by x_i):

$$\dot{a} = gb + \sum_{i=1}^n f_i' x_i^2 - \sum_{i=1}^n f_i a x_i \quad (10a)$$

$$\dot{b} = \sum_{i=1}^n d_i x_i - (g + \sum_{i=1}^n d_i') b \quad (10b)$$

$$\dot{x}_i = x_i (f_i a - f_i' x_i - d_i) + d_i' b. \quad (10c)$$

Equations (10) imply the conservation law

$$a + b + \sum_{i=1}^n x_i = c_0 = \text{const.}$$

Since the analysis is rather involved we consider first the case of irreversible degradation ($d_i'=0$). Then, the fixed points are readily computed. There are 2^n fixed points of equation (10). The most convenient notation uses sets of indices which correspond to the non-vanishing components: P_i is the fixed point with $\bar{x}_i \neq 0$ and $\bar{x}_k = 0$ for $k \neq i$; for P_{ij} we have $\bar{x}_i \neq 0$, $\bar{x}_j \neq 0$ and $\bar{x}_k = 0$ for $k \neq i, j$, etc.

Without losing generality we arrange indices such that

$$\frac{d_1}{f_1} < \frac{d_2}{f_2} < \dots < \frac{d_n}{f_n}.$$

Then, it is straightforward to show from (10) that the presence of species I_i at a stable stationary point implies also the presence of species I_j with $j < i$. Therefore all fixed points different from P_0 , P_1 , P_{12} , P_{123} , ..., $P_{12\dots n}$ are unstable irrespective of the values of the external parameters g and c_0 . In fact only one of the fixed points listed above is stable depending on the values of g and c_0 : if $f_i' > 0$, we obtain a sequence of double point bifurcations of the type $P_{12\dots i} \rightarrow P_{12\dots i, i+1}$ with increasing c_0 . At the point of coincidence a change in stability takes place (Fig.3) and a new species is introduced into the system at the stable stationary state. The regions of stability in the (g, c_0) plane are shown in Fig.4. The larger the environment, sustaining a larger total concentration c_0 , the more

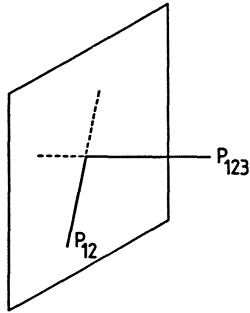


Figure 3. Double point bifurcation introducing a new species into the stable solutions of equations (10).

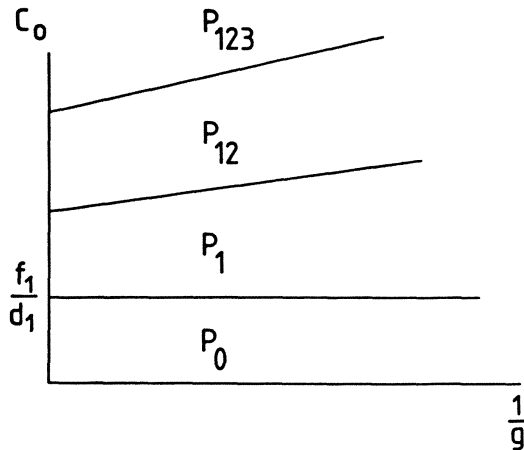


Figure 4. The (g, c_0) plane is split into $n+1$ regions of stability of the fixed points $P_0, P_1, P_{12}, \dots, P_{12\dots n}$ of equations (10) with $f_i' > 0$ ($i = 1, 2, \dots, n$).

species can coexist. We mention that a similar phenomenon has been found for the competition of self-replicating macromolecules the reproduction of which is based on Michaelis-Menten type kinetics [7,8] and in Lotka-Volterra food chains [9,10].

In case the autocatalytic reactions are irreversible ($f_i' = 0$), a condition usually met by ecological systems, and if the values of the quotients d_i/f_i are distinct, only P_0 and the one species fixed points, P_1, P_2, \dots, P_n , exist at finite values of c_0 , and P_1 , the steady state corresponding to the lowest value of d_i/f_i , is stable for $c_0 > d_1/f_1$. In this case we observe selection of the fittest species, this is the one which is characterized by the smallest quotient of degradation rate over replication rate.

The following theorems give some information on the global behaviour of the dynamical systems (10) with $d_i' = 0$, $i = 1, 2, \dots, n$.

Theorem 1: If there is no stationary point in the interior of the state space, i.e. $P_{12\dots n}$ does not exist, then at least one species dies out.

Proof: If the system of linear equations

$$\frac{\dot{x}_i}{x_i} = f_i a_i - f_i' x_i - d_i = 0; \quad i = 1, 2, \dots, n \quad (11a)$$

$$\dot{b} = \sum_{i=1}^n d_i x_i - gb = 0 \quad (11b)$$

has no solution $(\bar{a}, \bar{b}, \bar{x}_1, \dots, \bar{x}_n)$ with positive entries then a well known convexity argument ensures the existence of real numbers q_i ($i=0, 1, \dots, n$) such that

$$q_0 \left(\sum_{i=1}^n d_i x_i - gb \right) + \sum_{i=1}^n q_i (f_i a - f_i x_i' - d_i) > 0 \quad \text{for all } a, b, x_i > 0, i=1, 2, \dots, n.$$

But then the function $V = q_0 b + \sum_{i=1}^n q_i \log x_i$, defined for $x_i > 0$, satisfies

$$\dot{V} = q_0 \dot{b} + \sum_{i=1}^n q_i \frac{\dot{x}_i}{x_i} > 0$$

and therefore is a Lyapunov function. Hence, all orbits converge to points where at least one of the concentrations x_i vanishes.

Conversely, if there is an interior fixed point $P_{12\dots n}$, then it is stable. We were not able to prove its global stability. But we can provide a proof for the somewhat weaker statement.

Theorem 2: If the interior fixed point $P_{12\dots n}$ exists then the system is permanent [4]. The time averages of every solution starting with positive concentrations $x_i > 0, i=1, 2, \dots, n$, converge to the stationary solution $P_{12\dots n}$.

Proof: Firstly, it is easy to derive that if $P_{12\dots n}$ exists, then all 2^n fixed points P_J (with J any subset of $\{1, 2, \dots, n\}$) exist too and furthermore that the relation

$$\bar{a}(P_J) > \bar{a}(P_{12\dots n}) > \frac{d_i}{f_i} \quad (12)$$

holds for all i .

Therefore, we may assume (by induction on the number n of competitors) that on every boundary face $\{x_i = 0 \text{ for } i \notin J\}$ the restricted system (10) is permanent and time averages converge to P_J . But then we can verify the condition of the theorem derived in section 4 of [4] with

$$P = \prod_{i=1}^n x_i$$

as average Lyapunov function:

$$\begin{aligned} \frac{1}{T} \int_0^T \psi(x(t)) dt &= \frac{1}{T} \int_0^T \sum_{i=1}^n \frac{\dot{x}_i}{x_i} dt = \\ &= \sum_{i=1}^n \frac{1}{T} \int_0^T [f_i a(t) - f_i' x_i(t) - d_i] dt \rightarrow \sum_{i=1}^n [f_i \bar{a}(P_J) - f_i' \bar{x}_i(P_J) - d_i] \end{aligned}$$

for $T \rightarrow \infty$, and

$$\sum_{i=1}^n [f_i \bar{a}(P_J) - f_i' \bar{x}_i(P_J) - d_i] = \sum_{i \in J} [f_i \bar{a}(P_J) - d_i] > 0$$

from (12). Hence (10) is permanent. But then the convergence of the time averages is obtained immediately by integration of (11) using similar arguments as in [11].

A final remark is to be made on the reversible case, $d_i' > 0$. Here, only the first double point bifurcation $P_0 \rightarrow P_1$ occurs and at P_1 all competitors I_i , $i=1,2,\dots,n$, are present with positive concentrations. The further bifurcations do not take place since their branches do not intersect (the points $P_{12}, \dots, P_{12 \dots n}$ are always outside the state space). At small values of d_i' the branches, nevertheless, may come very close and we are dealing with "imperfect" bifurcations (see [12]).

4. Second-Order Autocatalysis

In order to provide some feeling for the enormous richness of the dynamics of higher order catalytic systems we study now the trimolecular reaction (2) coupled to degradation and recycling in analogy to the reaction scheme (3):



The dynamics of this mechanism is described by the differential equation

$$\dot{a} = gb + f'x^3 - fax^2 \quad (14a)$$

$$\dot{b} = dx - (d'+g)b \quad (14b)$$

$$\dot{x} = fax^2 + d'b - f'x^3 - dx. \quad (14c)$$

Again the total concentration is conserved:

$$a+b+x = c_0 = \text{const.}$$

To simplify the calculations we consider only the case $d'=0$ here, although the results obtained are valid also for systems with $d'>0$. The first basic difference compared to equation (4) one observes in analyzing (14) is the fact that the fixed point $P_0(\bar{a}=c_0, \bar{b}=\bar{x}=0)$ is

always stable. The positions of the other two fixed points P and Q are given by

$$\bar{x} = \frac{c_0 f \pm \left[c_0^2 f^2 - 4d \left(f + f' + \frac{fd}{g} \right) \right]^{1/2}}{2 \left(f + f' + \frac{fd}{g} \right)}$$

Let $p > q$ denote the two values of x at the corresponding interior fixed points P and Q. Note that p grows and q shrinks with increasing c_0 . It is then easy to derive that Q is always a saddle and P either a sink or a source.

So for $c_0 < c_{SN} = \frac{1}{f} \left[4d \left(f + f' + \frac{fd}{g} \right) \right]^{1/2}$, P_0 is the only fixed point and as c_0 crosses c_{SN} , by a saddle-node bifurcation P and Q emerge into the interior of the state space S.

In order to determine the stability of P we evaluate the trace of the Jacobian (A) at P:

$$\text{trace } A(x=p) = d - g - p^2 (f + f') \quad (16)$$

which is a decreasing function of c_0 .

If expression (16) is negative at the birth of P ($c_0 = c_{SN}$) then P is stable for all $c_0 > c_{SN}$. This corresponds to a direct transition from region I into region V in fig.5 and it happens in particular always if $d \leq g$. In this special case a similar argument as applied in section 2, using now x^{-2} as a Dulac function, excludes the existence of periodic orbits so that all solutions have to converge to one of the fixed points P_0 , P or eventually Q.

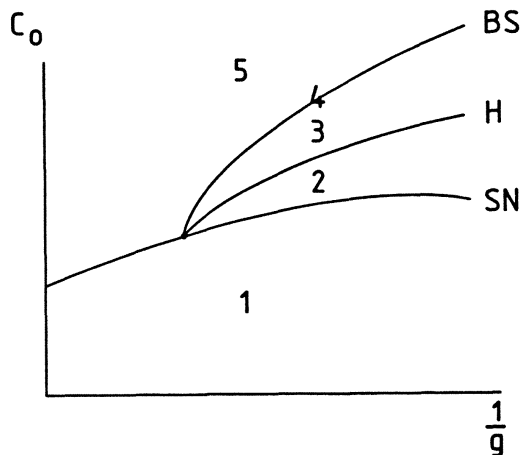


Figure 5. The regions of different dynamical behaviour of equation (14) in the (g, c_0) plane. We observe three types of bifurcations: SN = saddle-node bifurcation, H = Hopf bifurcation and BS = blue sky bifurcation. The regions denoted by 1, 2, ..., 5 refer to different phase portraits shown in Fig.6.

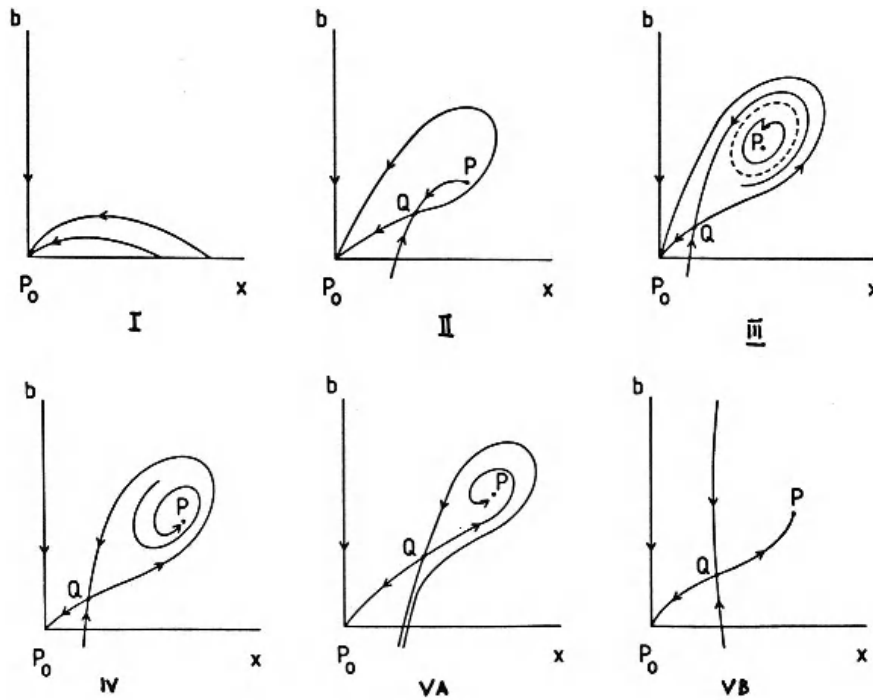


Figure 6. Phase portraits of the regions of different dynamical behaviour of equation (14) according to Fig.5. I: $c_0 < c_{SN}$; P_0 is the only fixed point in S . II: $c_{SN} < c_0 < c_H$; Q is a saddle, P a source. III: $c_H < c_0 < c_{BS}$; P is a sink whose basin of attraction is bounded by an unstable periodic orbit. IV: $c_0 = c_{BS}$; a homoclinic orbit including Q . V: A: $c_0 \gtrsim c_{BS}$ the basin of P contains only a small strip of the x axis. B: $c_0 \gg c_{BS}$; P has a large basin of attraction.

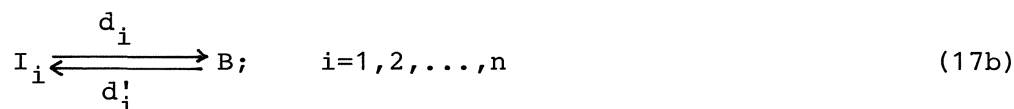
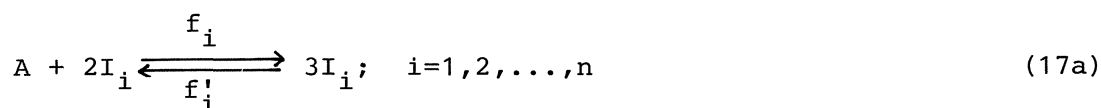
If, however, the numerical value of (16) at $c_0 = c_{SN}$ is positive, the fixed point P is first an unstable node (fig.6,II). With increasing c_0 the eigenvalues become complex conjugate. Since (16) is decreasing the eigenvalues have to cross the imaginary axis at a certain value $c_0 = c_H$ at which a Hopf bifurcation occurs. The value of c_H may be obtained from (16) by setting $\text{trace } A(x=p) = 0$. A lengthy calculation based on the treatment given by MARS DEN and MC CRACKEN [13] shows that this Hopf bifurcation is always subcritical. This means that the bifurcating periodic orbit is always unstable and occurs for $c_0 > c_H$ when P is stable. In fact, it separates the basins of attraction for the two stable fixed points P_0 and P (fig.6,III). Further increase of c_0 leads to growth of the periodic orbit until it includes the saddle Q at the next critical value $c_0 = c_{BS}$. Accordingly, the periodic orbit changes into a homoclinic orbit (fig.6,IV) and disappears for $c_0 > c_{BS}$. This phenomenon is sometimes called "blue sky bifurcation" [14]. The numerical value of c_0 at which the blue sky bifurcation occurs (c_{BS}) can be computed approximately only.

At total concentrations $c_o > c_{BS}$ also orbits starting from the boundary of S have a chance to converge to P. At values close to c_{BS} the admissible range of initial concentrations $x(o)$ for final survival of the autocatalyst is very small (fig.6,VA): if $x(o)$ is too small or if it is too large the orbit will converge to P_o and the autocatalyst will die out. For large values of c_o , of course, the basin of attraction of P becomes almost the entire state space S, since the saddle Q tends towards P_o for $c_o \rightarrow \infty$ (fig.6,VB).

Finally, we mention that the existence of two stable solutions on S as found in regions III, IV and V (fig.5,6) is a common and well-known feature of higher order autocatalytic reactions. One of the most striking properties of reaction networks including higher order autocatalytic processes is the enormous sensitivity on initial conditions as illustrated in fig. 6 (VA).

5. Competition Between Higher Order Autocatalytic Reactions

We conclude this contribution with some remarks on the second order analogue of mechanism (9). By second order we refer to the trimolecular autocatalytic reaction (2):



For the sake of simplicity we restrict ourselves to two competitors ($n=2$) and irreversible degradation ($d'_i=0$). Then, the dynamics is described by the differential equation

$$\dot{b} = d_1 x_1 + d_2 x_2 - gb \quad (18a)$$

$$\dot{x}_1 = x_1 (f_1 a x_1 - f'_1 x_1^2 - d_1) \quad (18b)$$

$$\dot{x}_2 = x_2 (f_2 a x_2 - f'_2 x_2^2 - d_2) \quad (18c)$$

with $a = c_o - x_1 - x_2 - b$.

Again the fixed point $P_o (\bar{x}_1 = \bar{x}_2 = \bar{b} = 0)$ is always stable. Moreover, it is obvious that any x_i which is very small is characterized by $\dot{x}_i < 0$ and hence will tend towards zero. In contrast to the competition

between "linear" autocatalytic processes (9) all invariant surfaces of S , coordinate planes where some x_i are zero, are attracting. On both planes, $x_1=0$ and $x_2=0$, we observe all the dynamical behaviour described in the previous section. In particular, we obtain two stable fixed points $P_1 (\bar{x}_1>0, \bar{x}_2=0)$ and $P_2 (\bar{x}_1=0, \bar{x}_2>0)$ after having passed through the regions of transition shown in fig.6.

Increasing c_0 further in the case $f'_i>0$ we observe creation of two pairs of fixed points in the interior of the state space. If c_0 is large enough one of the four fixed points, $P_{12}(\bar{x}_1>0, \bar{x}_2>0)$, will be stable and finally, we end up with four stable and five unstable fixed points. The various bifurcations occurring with increasing c_0 are difficult to analyze and one might expect some chaotic behaviour near the Hopf and homoclinic bifurcations. The methods developed by Couillet [15] will be applied to this problem in the near future.

In the case $f'_i=0$ only one pair of fixed points emerges in the interior of S . These are always a source and a saddle. There is no stable fixed point in the interior not even for large values of c_0 . This indicates that in the irreversible case all orbits converge to one of the planes $x_1=0$ or $x_2=0$. At least one of the competitors has to die out. As we found also for the "linear" case (9) there is no region of coexistence of the two competitors when the autocatalytic processes are irreversible.

6. Conclusion

In this contribution we tried to present an impressive illustration for the difference between linear and higher order autocatalytic processes. In case of linear autocatalysis we were able to derive global results, i.e. results for a given set of external parameters which are valid in the entire phase space S . This is not the case for higher order autocatalytic processes: in general, we find more than one attractor and hence, only results of local validity can be obtained. No global fitness functions or global section criteria exist in these systems.

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