Now, for the sake of clarity let us suppose that $\bar{x} = \bar{y} = \tilde{C} = 1$. Then the models we have obtained are

$$\dot{x} = 2xy(1-y), \qquad \dot{y} = y(xy-1) \tag{36'''}$$

$$\dot{x} = 2x^2(1-y), \qquad \dot{y} = y(xy-1) \tag{39'''}$$

$$\dot{x} = x(1-y^2), \qquad \dot{y} = y(xy-1) \tag{42'''}$$

$$\dot{x} = 2x(1-y), \qquad \dot{y} = y(xy-1). \tag{44'''}$$

A possible formal chemical reaction giving rise to (42''') is

$$X + 2Y \to 3Y, \qquad A + X \to 2X, \qquad Y \to B, \tag{60}$$

which is the two-dimensional (or reduced) explodator.

4. Discussion

We have shown that the Lotka–Volterra model is the simplest unique model among all those models having the same linearized form. We have also shown that only three models are distinct from the two-dimensional explodator, if all of these models are to have the same linearized form. These models are therefore worth investigating by methods of the qualitative theory of differential equations since the two-dimensional explodator [4], as well as the original [12] explodator, have already been studied by such methods. However, it seemed to us to be a relatively easy task to formulate a common scheme, in the language of mixed-integer programming, for the construction of formal chemical reactions possessing prescribed properties. This enabled the construction of formal chemical reactions having desired features (e.g. multiple stationary states with prescribed linear parts, local controllability, etc.) algorithmically by computer [9].

Acknowledgements. We would like to thank Henrik Farkas and Zoltán Noszticzius for their helpful discussions, and János Hoffer for his calculations revealing a mistake. Valuable remarks by John J. Tyson and by the referee on preliminary versions of this paper were also taken into consideration.

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Theor. Chim. Acta (1986) 70: 143–150

Theoretica Chimica Acta

Specification of oscillating chemical models starting from a given linearized form

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(Received May 6, 1985, revised April 15/Accepted April 23, 1986)

The Lotka-Volterra model is shown to be the simplest unique model among those models having the same linearized form. We also show that the two-dimensional explodator model is not unique in its own class: there are four models possessing the same linearized form. Finally, we propose a method for the construction of formal chemical models having prescribed properties (i.e. having prescribed a type of one or more equilibrium points).

Key words: Lotka-Volterra model — Explodator — Linearized part — Uniqueness — Design of periodic reactions

1. Introduction

The investigations of Hanusse [1], Tyson and Light [2] and Póta [3] have shown that in two-component bimolecular systems there is only one oscillator: the Lotka–Volterra model. The following question has now arisen; is it also true that the Lotka–Volterra model is the simplest unique one among all those models having the same linearized form (around their own stationary state)? From both chemical and mathematical points of view this is a very different question.

From the mathematical point of view, we provide a new proof that the Lotka–Volterra model is the only possible two-component bimolecular oscillator. (Strictly speaking, this statement is proved under a slightly different set of assumptions.) It is also shown that the reduced explodator model by Farkas et al. [4] is almost just as unique in its own class. However, the chemical approach seems to be more useful: The present investigation bears on the area of *designing periodic reactions*. Experimental work in this field has been reviewed by Epstein [5], while Escher [6], Schnakenberg [7], and Császár et al. [8] have discussed the theory. Our results may be obtained as results of a mixed-integer programming problem too, and this formulation has led us to construct a promising method for the design of chemical models with prescribed irregularities, e.g. with periodic or exploding solutions, with multistationarity, or with local controllability [9].

The present paper is an exposition of a part of our lecture [10] presented in Bordeaux.

2. Uniqueness of the Lotka–Volterra model

Exactly formulated, the question is as follows: Let us consider the linearized form

$$\dot{\xi} = -\eta, \qquad \dot{\eta} = \xi \tag{1}$$

of the Lotka–Volterra model

$$\dot{x} = x - xy, \qquad \dot{y} = xy - y. \tag{2}$$

What is the simplest two-dimensional kinetic system that leads to (1) when linearized around one of its own stationary points?

By "kinetic" we mean that the system of differential equations we are looking for is a polynomial system which does not contain negative cross-effects [11], i.e. it does not contain terms expressing the decrease of one of the components without the participation of that component in a polynomial term. Equation (1) is an example of a non-kinetic equation since $-\eta$ expresses the decrease of ξ in a process in which ξ does not take part.

In order the define "simplest" we first remark that no linear kinetic system may lead to (1) through linearization since the linearized form of a linear system is itself, and (1) is non-kinetic.

Thus, let us look for the system to be linearized among systems of the second degree, i.e. among those systems of the form

$$\begin{aligned} \dot{x} &= ax^2 + bxy + cy^2 + dx + ey + f \\ \dot{y} &= Ax^2 + Bxy + Cy^2 + Dx + Ey + F \end{aligned} \tag{3}$$

and let us suppose that the system is kinetic. This means that

$$c, e, f, A, D, F \geq 0. \tag{4}$$

Let us suppose too that there are no genuine elementary reactions of the third order. This implies that

$$a, C \leq 0 \tag{5}$$

and that at most one of b and B is strictly positive. (The arguments leading to these inequalities can be found in [1], p. 1247, or in [2], Sect. II.) Finally, let (3) be the simplest possible equation in the sense that at most two of the coefficients on the right-hand sides of (3) differ from zero. (The reader may wish to check that no kinetic solution to the problem exists when one requires that only one coefficient be nonzero. On the other hand, two nonzero coefficients will suffice.)

Denoting one of the positive stationary points of (3) by $(\bar{x}, \bar{y})$ [around which the linearized form should be the same as the linearized form of the Lotka–Volterra model around $(\bar{x}, \bar{y})$], the above requirements can be expressed as follows. The fact that $(\bar{x}, \bar{y})$ is a stationary point means that

$$a\bar{x}^2 + b\bar{x}\bar{y} + c\bar{y}^2 + d\bar{x} + e\bar{y} + f = 0 \tag{6}$$

$$A\bar{x}^2 + B\bar{x}\bar{y} + c\bar{y}^2 + D\bar{x} + E\bar{y} + F = 0. \tag{7}$$

Equation (3), when linearized around $(\bar{x}, \bar{y})$, gives (1), which implies

$$2a\bar{x} + b\bar{y} + d = 0 \qquad b\bar{x} + 2c\bar{y} + e = -1 \tag{8}$$

$$2A\bar{x} + B\bar{y} + D = 1 \qquad B\bar{x} + 2C\bar{y} + E = 0. \tag{9}$$

First, let us concentrate on the coefficients of the first equation of (3). We have to determine those solutions of the system (6), (8a, b) that have at most two components different from zero. It is clear that at least one of b, c and e must be negative because of (8b). On the other hand, neither c nor e may be negative because of (4). Thus, b must be less than zero. If it is, at least one of a and d must be positive because of (8a). On the other hand, a may not be positive because of (5). We now show that it is enough to suppose that only b and d differ from zero, namely $b<0$ and $d<0$. Assuming that all of the other coefficients are zero, system (6), (8a, b) reduces to

$$b\bar{x}\bar{y} + d\bar{x} = 0 \qquad b\bar{y} + d = 0 \qquad b\bar{x} = -1. \tag{10}$$

This system of linear equations has a unique solution:

$$b = -1/\bar{x}, \qquad d = \bar{y}/\bar{x}. \tag{11}$$

The form of (3a) in this case is:

$$\dot{x} = (x/\bar{x})(\bar{y} - y).$$

Let us turn to the coefficients of the second equation of (3). We have to determine those solutions of the system (7), (9a, b) that have at most two components different from zero. It is obvious that at least one of A, B and D must be positive because of (9a). Two cases may occur: in the first, at least one of A, B and D is negative, but this can only be B because of (4). In the second, none of A, B and D is negative. In this case, (7) implies that at least one of C, E and F is negative, but this can only be either C or E because of (4). Thus, the set of nonzero coefficients may be either

$$A > 0, \qquad B < 0 \tag{12}$$

$$D > 0, \qquad B < 0 \tag{13}$$

or

$$A>0, \qquad C<0 \tag{14}$$

$$A>0, \qquad E<0 \tag{15}$$

$$B>0, \qquad C<0 \tag{16}$$

$$B>0, \qquad E<0 \tag{17}$$

$$D>0, \qquad C<0 \tag{18}$$

$$D>0, \qquad E<0. \tag{19}$$

Equation (9b) implies that either $B=C=E=0$ or at least two of B, C and E differ from zero. Only conditions (16) and (17) represent cases that are not excluded by this argument. System (7), (9a, b) is contradictory under condition (16). Thus, the only admissible case is (17) and we find

$$B=1/\bar{y}, \qquad E=-\bar{x}/\bar{y}. \tag{20}$$

The form of (3b) in this case is

$$\dot{y}=(y/\bar{y})(x-\bar{x}). \tag{21}$$

3. Non-uniqueness of the two-dimensional explodator

We now consider the linearized form

$$\dot{\xi}=-2\eta, \qquad \dot{\eta}=\tilde{C}\xi+\tilde{C}\eta \qquad (\tilde{C}>0) \tag{22}$$

of the two-dimensional explodator [4]

$$\dot{x}=x(1-y^2), \qquad \dot{y}=\tilde{C}y(xy-1). \tag{23}$$

It is obvious that no linear kinetic system may lead to (22) through linearization since (22) is non-kinetic.

The reader will find no difficulty in showing by arguments similar to those of Sect. 2 that no kinetic system of the form (3), having only two nonzero coefficients in both of the equations, may have (22) as its linearized form.

Thus, let us look for the system to be linearized to (22) from among systems of the third degree, i.e. among systems of the form

$$\begin{aligned}\dot{x}&=ax^3+bx^2y+cxy^2+dy^3+ex^2+fxy+gy^2+hx+iy+j\\ \dot{y}&=Ax^3+Bx^2y+Cxy^2+Dy^3+Ex^2+Fxy+Gy^2+Hx+Iy+J\end{aligned} \tag{24}$$

and let us suppose that the system is kinetic. This implies, in the same way as

in Sect. 3 (cf. [1] and [2]), that

$$d,\, g,\, i,\, j\geq 0, \qquad A,\, E,\, H,\, J\geq 0. \tag{25}$$

Let us suppose that there are no genuine elementary reactions of the fourth order. This implies that

$$a\leq 0, \qquad D\leq 0 \tag{26}$$

and that b and B (and c and C as well) together can only be nonnegative if both of them are zero. Finally, let (24) be the simplest possible equation in the sense that at most two of the coefficients of the right hand sides differ from zero.

Denoting one of the positive stationary points of (24) by $(\bar{x}, \bar{y})$ [around which the linearized form should be (22)], the above requirements can be expressed as follows. The fact that $(\bar{x}, \bar{y})$ is a stationary point means that

$$a\bar{x}^3+b\bar{x}^2\bar{y}+c\bar{x}\bar{y}^2+d\bar{y}^3+e\bar{x}^2+f\bar{x}\bar{y}+g\bar{y}^2+h\bar{x}+i\bar{y}+j=0 \tag{27}$$

$$A\bar{x}^3+B\bar{x}^2\bar{y}+C\bar{x}\bar{y}^2+D\bar{y}^3+E\bar{x}^2+F\bar{x}\bar{y}+G\bar{y}^2+H\bar{x}+I\bar{x}+J=0. \tag{28}$$

Equation (24), when linearized around $(\bar{x}, \bar{y})$, gives (22), which implies that

$$3a\bar{x}^2+2b\bar{x}\bar{y}+c\bar{y}^2+2e\bar{x}+f\bar{y}+h=0 \tag{29}$$

$$b\bar{x}^2+2c\bar{x}\bar{y}+3d\bar{y}^2+f\bar{x}+2g\bar{y}+i=-2 \tag{30}$$

$$3A\bar{x}^2+2B\bar{x}\bar{y}+C\bar{y}^2+2E\bar{x}+F\bar{y}+H=\tilde{C} \tag{31}$$

$$B\bar{x}^2+2C\bar{x}\bar{y}+3D\bar{y}^2+F\bar{x}+2G\bar{y}+I=\tilde{C}. \tag{32}$$

First, let us concentrate on the coefficients of the first relation of (25). We have to determine those solutions of the system (27), (29), (30) that have at most two components different from zero. It is obvious that at least one of b, c, and f must be negative because of (25a) and (30). Two cases may occur: In the first, at least one of b, c and f is positive. In the second, none of b, c and f is positive. In the latter case (29) implies that at least one of a, e and h is positive but this can only be either e or h because of (26a). Thus, the set of nonzero coefficients may be either

$$b<0, \qquad c>0 \tag{33}$$

$$b<0, \qquad f>0 \tag{34}$$

$$c<0, \qquad b>0 \tag{35}$$

$$c<0, \qquad f>0 \tag{36}$$

$$f<0, \qquad b<0 \tag{37}$$

$$f<0, \qquad c<0 \tag{38}$$

or

$$b<0, \qquad e>0 \tag{39}$$

$$b<0, \qquad h>0 \tag{40}$$

$$c<0, \qquad e>0 \tag{41}$$

$$c<0, \qquad h>0 \tag{42}$$

$$f<0, \qquad e>0 \tag{43}$$

$$f<0, \qquad h>0. \tag{44}$$

System (27), (29), (30) is contradictory under conditions (33), (34), (35), (37), (38), (40), (41) and (43). Therefore, the admissible cases are (36), (39), (42) and (44), for which

$$c=-2/\bar{x}\bar{y}, \qquad f=2/\bar{x} \tag{36'}$$

$$b=-2/\bar{x}^2, \qquad e=2\bar{y}/\bar{x}^2 \tag{39'}$$

$$c=-1/\bar{x}\bar{y}, \qquad h=\bar{y}/\bar{x} \tag{42'}$$

$$f=-2/\bar{x}, \qquad h=2\bar{y}/\bar{x}. \tag{44'}$$

The form of (24a) in these cases is, respectively,

$$\dot{x}=(2xy/\bar{x}\bar{y})(\bar{y}-y) \tag{36''}$$

$$\dot{x}=(2x^2/\bar{x}^2)(\bar{y}-y) \tag{39''}$$

$$\dot{x}=(x/\bar{x}\bar{y})(\bar{y}^2-y^2) \tag{42''}$$

$$\dot{x}=(2x/\bar{x})(\bar{y}-y). \tag{44''}$$

Let us turn to the coefficients of the second equation of (24). We have to determine those solutions of the system (28), (31), (32) that have at most two components different from zero.

It is obvious, because of (31), that at least one of A, B, C, E, F and H must be positive. Two cases may occur: in the first one, at least one of these coefficients is negative, but this can only be B, C or F because of (26b); in the second case, none of these coefficients is negative. In this latter case, (28) and (25b) imply that at least one of D, G and I is negative.

Similarly, at least one of B, C, D, F, G and I, must be positive because of (32). On the other hand, D cannot be positive because of (26b). Two cases may again occur: in the first one, at least one of B, C, F, G and I is negative, while in the second one none of them is negative. In this latter case, (28) and (25b) imply that D is negative.

The following 15 possibilities remain as the intersection of the sets of nonzero coefficient pairs defined by the two previous paragraphs:

$$B>0, \qquad C<0 \tag{45}$$

$$B>0, \qquad F<0 \tag{46}$$

$$B>0, \qquad D<0 \tag{47}$$

$$B>0, \qquad G<0 \tag{48}$$

$$B>0, \qquad I<0 \tag{49}$$

$$C>0, \qquad B<0 \tag{50}$$

$$C>0, \qquad F<0 \tag{51}$$

$$C>0, \qquad D<0 \tag{52}$$

$$C>0, \qquad G<0 \tag{53}$$

$$C>0, \qquad I<0 \tag{54}$$

$$F>0, \qquad B<0 \tag{55}$$

$$F>0, \qquad C<0 \tag{56}$$

$$F>0, \qquad D<0 \tag{57}$$

$$F>0, \qquad G<0 \tag{58}$$

$$F>0, \qquad I<0. \tag{59}$$

Under conditions (46), (49), (52), (53), (55), (56) and (59), our system (28), (31), (32) is contradictory. Under conditions (45), (47), (48), (50), (52) (57) and (58), the system can only be solved if an additional condition of the type $\bar{x}/\bar{y}=-1$, $\bar{x}/2\bar{y}=-1$, etc. is fulfilled. However, these conditions are impossible because of the positivity of $\bar{x}$ and $\bar{y}$.

The only condition under which the system (28), (31), (32) has the desired solution is (54), which gives

$$C=\tilde{C}/\bar{x}^2, \qquad I=-\tilde{C} \tag{54'}$$

if and only if the additional condition $\bar{x}=\bar{y}$ is met as well. The form of (24b) in this case is

$$\dot{y}=(\tilde{C}/\bar{x}^2)xy^2-\tilde{C}y=\tilde{C}y[(xy/\bar{x}^2)-1]. \tag{54''}$$

In summary, there are four systems of kinetic differential equations fulfilling the above requirements, namely, (36″), (54″); (39″), (54″); (42″), (54″); and (44″), (54″).

The argument formulated after (26) excludes none of these models. The analogous argument was not needed in the case of the Lotka–Volterra model either.