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The irreversible Brusselator: the influence of degenerate singularities in a closed system

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Abstract. The appearance of apparently chaotic behaviour in numerical treatment of this two-dimensional system is examined from an analytical point of view. The original two-parameter model exhibiting chaotic-like solutions is unfolded to a three-parameter model. This enlarged model is shown to have a codimension-two degenerate Hopf bifurcation, the unfolding of which contains phase portraits with three concentric limit cycles.

Some segments of these limit cycles are so close to each other that numerical integration causes transitions across the unstable limit cycle, thus giving the appearance of chaotic behaviour. The region in parameter space where this occurs is quite significant and it includes part of the plane of the original two-parameter model.

1. Introduction

The 'Brusselator' is a model reaction scheme introduced by Prigogine and Lefever [1] as a nonlinear chemical oscillator. Its viability as a legitimate chemical model has been questioned by e.g. Gray and Morley-Buchanan [2], King [3], Gray, Merkin and Scott [4] amongst others, and defended by Nicolis, Lefever and Borckmans [5]. The present work is completely independent of this controversy, simply looking at the mathematical behaviour of the original model, where new and at first puzzling results are obtained. These are understood in terms of an extended model (from two parameters to three) in a way which has no bearing on the questions of chemical viability. A similar phenomenon shown by another simple chemical oscillator, the cubic autocatalator, has been discussed recently by Gray and Thuraisingham [6]. In that case unfolding the model by inclusion of an extra parameter gave a complete explanation of the numerical behaviour. The present case is parallel to the earlier one, so we will present rather less detail than in [6].

The differential equations describing the irreversible Brusselator are

$$\frac{dx}{dt} = \mu - (1 + \alpha)x + x^2 y , \tag{1}$$

$$\frac{dy}{dt} = x - x^2 y . \tag{2}$$

This dimensionless form is as used in [4]. The nonlinearity $x^2 y$ also occurs in the related 'cubic autocatalator' [6] described in dimensionless form by

$$\frac{dx}{dt} = x^2 y - x + ry , \tag{3}$$

$$\frac{dy}{dt} = \mu - x^2 y - ry . \tag{4}$$

In [6] numerical solution of (3) and (4) gave chaos-like behaviour for very small ranges of μ in the region $1/8 > r > 0$. This computer-generated artifact was understood by analysing the most degenerate Hopf bifurcation of the 'unfolded system'

$$\frac{dx}{dt} = x^2 y - x + ry \,, \tag{5}$$

$$\frac{dy}{dt} = \mu - x^2 y - (\alpha + r)y \,. \tag{6}$$

This system shows an $H3_2$ singularity (see Gray and Roberts [7]) and the phase portrait containing three concentric limit cycles implied by this is shown to occur on the axis $\alpha = 0$ in parameter space. Segments of these limit cycles are so close together that numerical instability results regardless of the precision and computational parameters used. We apply similar techniques here to the model (1) and (2) and obtain a similar explanation of the chaos-like behaviour shown by this model. The latter has not been reported previously although the model has now been in the literature for over twenty years.

2. The unfolded model

We generalise the original model to a three-parameter one. In this case the following differential equations serve our purpose:

$$\frac{dx}{dt} = \mu - (1 + \alpha)x + x^2 y \,, \tag{7}$$

$$\frac{dy}{dt} = \mu - x^2 y - \beta y \,, \tag{8}$$

where the new term is $-\beta y$. If we use the methods outlined in Gray and Roberts [7] for two-dimensional three-parameter o.d.e.'s, we can obtain a complete qualitative map of the behaviour of (7) and (8). The unfolded model differs qualitatively from the original one in that for $\beta > 0$ it can exhibit saddle-node bifurcations, i.e. it exhibits multistability. In this sense, the original model is structurally unstable with respect to the term $-\beta y$, $\beta > 0$, however small β may be.

The loci of saddle-node bifurcations are given by

$$\mu = \frac{2x^3\beta}{x^4 + 2\beta x^2 + \beta^2} \,, \tag{9}$$

$$\alpha = \frac{\beta(x^2 - \beta)}{x^4 + 2\beta x^2 + \beta^2} \,, \tag{10}$$

where the parameter $x > \sqrt{\beta}$ for physical applicability (i.e. the nonnegative quadrant in parameter space). This system also shows a hysteresis degeneracy which is

$$\alpha = 1/8 \tag{11}$$

in the codimension-one (α, β) plane [7].

Ordinary Hopf bifurcations occur in this system on the locus given by

$$\alpha = \frac{x^2 - x^4 - (\beta + 2x^2\beta + \beta^2)}{x^2 + \beta}, \tag{12}$$

$$\mu = \frac{x(\beta^2 + 2\beta) + x^3(2\beta - 1) + x^4}{x^2 + \beta}, \tag{13}$$

$$\det J > 0, \tag{14}$$

where J is the Jacobian matrix for the system.

A degenerate Hopf bifurcation of type $H2_1$ (see [7]) can occur in this system, and is defined by equations (12), (13) and (14) plus

$$\frac{d}{d\mu} \operatorname{trace} J = 0, \tag{15}$$

where we have now chosen μ as the bifurcation parameter. After some algebra we obtain this locus as

$$\alpha = 1 - 2\sqrt{2}\beta, \qquad \det J > 0. \tag{16}$$

The second type of codimension-one Hopf bifurcation ($H3_1$) has to be investigated numerically. We use the condition of Gobber and Willamowski [8] to calculate these points numerically.

Codimension-two bifurcations, the most degenerate ones possible in this system, if they exist, will occur on the codimension-one curves, i.e. as discrete points on such curves. We are particularly interested in the occurrence of an $H3_2$ degeneracy on the $H3_1$ loci. These are located by calculating the point(s) $P_6 = 0$ along the $H3_1$ ($P_4 = 0$) curve, using the notation of Gobber and Willamowski. In this particular system, i.e. (7) and (8), we find the presence of one such point.

It finally remains to investigate the occurrence of a double zero eigenvalue condition (DZE) where

$$\operatorname{trace} J = 0, \qquad \det J = 0, \tag{17}$$

which is a codimension-one degeneracy. This corresponds to the intersection of the saddle-node and nondegenerate Hopf loci, and it will be a curve in three-dimensional parameter space. This can be obtained explicitly as

$$\beta = \frac{3\alpha}{4} \pm \frac{\alpha}{4}(1 - 8\alpha)^{1/2}. \tag{18}$$

it is also possible that this system can exhibit one or more degenerate DZE points, for example where the $H2_1$ locus (16) intersects the DZE locus (18) at the roots of

$$(\alpha - 1)^4 - 4\alpha^2(1 - 8\alpha) = 0. \tag{19}$$

The simplest possible unfolding of a point of this type is given in Gray and Roberts [7].

Other possible degenerate DZE's for this system will be discussed in another paper, our prime concern here being to relate the $H3_2$ degeneracy to the numerical results.

3. Quantitative results

In Figs 1 and 2 the chaos-like behaviour is shown for two different sets of parameter values. In Fig. 3 a phase-plane plot is shown of similar mixed oscillations, showing the two stable limit cycles. Between them, in an unknown position will be an unstable limit cycle.

In Fig. 4 the loci of codimension-one degeneracies are shown in the (α, β) plane. There are two branches of the $H3_1$ curve and a single $H3_2$ point at $\alpha = 0.112$, $\beta = 0.063$. Note that the classical model has $\beta = 0$. Two non-local curves, the hysteresis of limit-cycle locus and Hopf plus period orbit bifurcations (see [6]) are included in order to give path consistency in the (α, β) planes around the $H3_2$ point. The region between the HL and H + PO curves is relevant to our computed results since it contains phase portraits with three concentric limit cycles. The presence of the mixed oscillations for the classical model indicates that this region intersects the $\beta = 0$ plane in parameter space. Here two curves as shown in Fig. 4 have to be computed. For example, for $\beta = 10^{-4}$, the mixed oscillations occur for $0.95 > \alpha > 0.01$ (albeit for a small range of μ, see Fig. 6).

The bifurcation loci in the (μ, α) plane for small $\beta(= 10^{-4})$ are shown in Fig. 5. Again we have not included all the codimension-zero bifurcation loci (i.e. the saddle-node loci), only those relevant to the present discussion, i.e. those on which the number of limit cycles changes. Note that Fig. 5 for $\beta = 10^{-4}$ is qualitatively different from the classical diagram Fig. 6, to which we have added the new region F where the phase portrait contains three limit cycles.

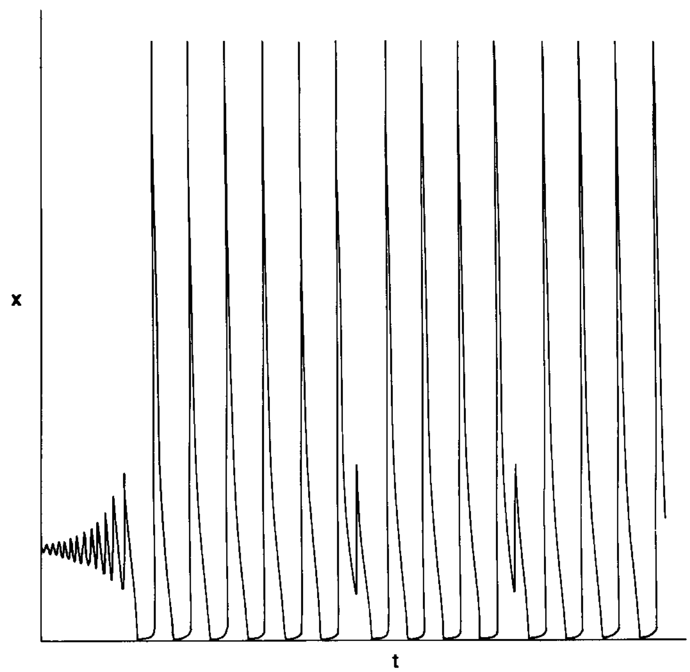

Fig. 1. Plot of x as a function of time for $\mu = 0.048$, $\alpha = 0.05$.

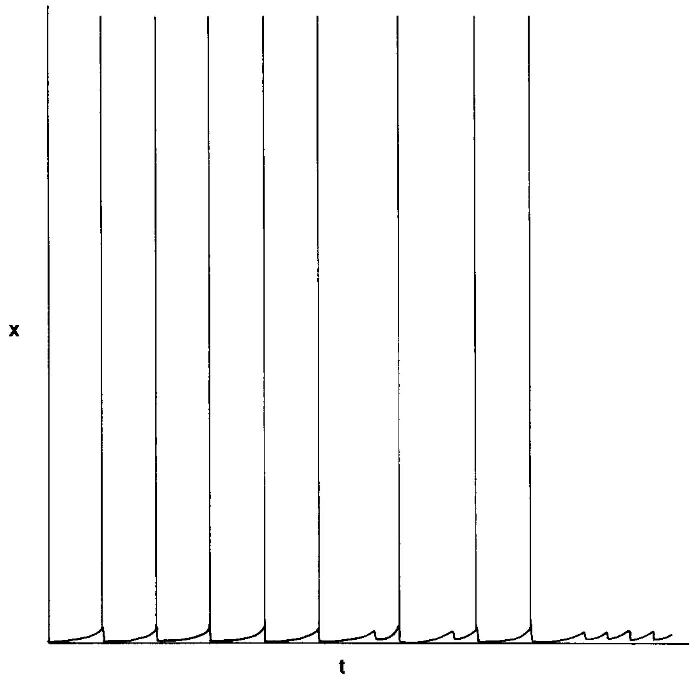

Fig. 2. Plot of x as a function of time for $\mu = 0.112$, $\alpha = 0.98$.

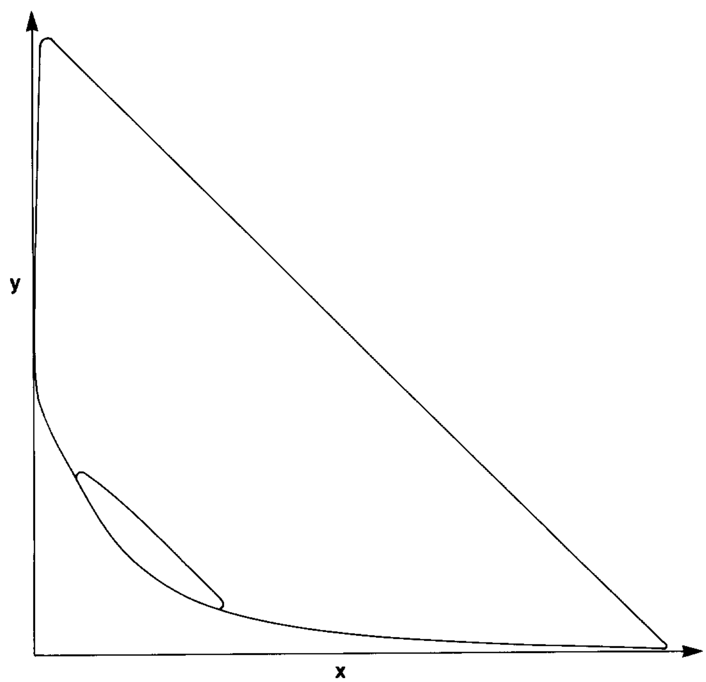

Fig. 3. Phase portrait y vs. x for $\mu = 0.048$, $\alpha = 0.05$.

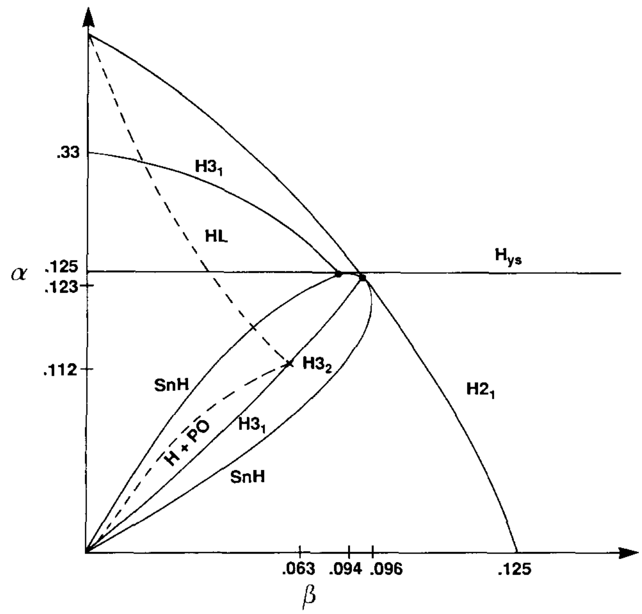

Fig. 4. Codimension (α, β) plot, showing the $H3_2$ point.

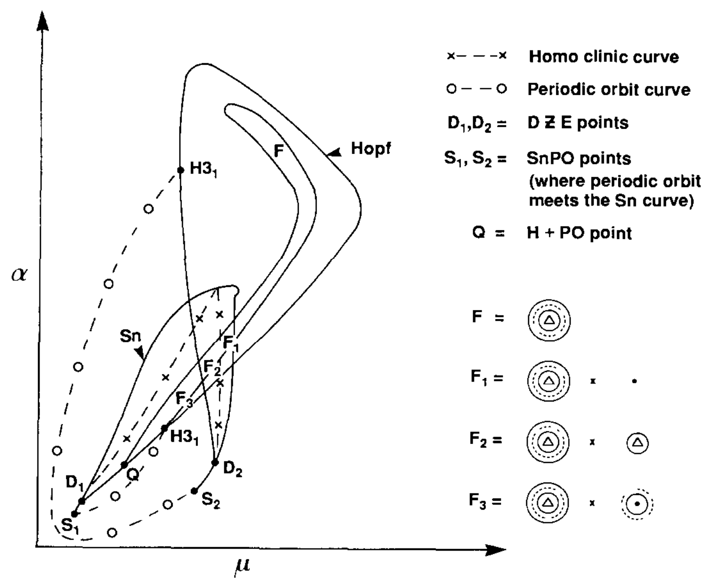

Fig. 5. (α, μ) diagram for $\beta = 10^{-4}$, showing the region of 3 limit cycles.

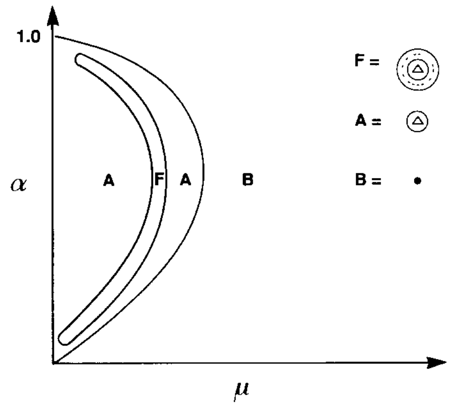

Fig. 6. Modified (α, μ) diagram for $\beta = 0$.

4. Conclusions

The classical irreversible Brusselator model represented by the equations

$$\frac{dx}{dt} = \mu - (1 + \alpha)x + x^2 y ,$$

$$\frac{dy}{dt} = x - x^2 y ,$$

has a hitherto unknown region in parameter space where the phase portrait contains three concentric limit cycles. This behaviour is characteristic of a codimension-two degenerate Hopf bifurcation, requiring a model with at least three parameters. Its presence indicates possible structural instability with respect to further unfolding.

Unfolding of the model to

$$\frac{dx}{dt} = \mu - (1 + \alpha)x + x^2 y ,$$

$$\frac{dy}{dt} = x - x^2 y - \beta y ,$$

confirms the expectation of a nearby $H3_2$ degeneracy which occurs for $\beta = 0.063$. Moreover, the region of its unfolding containing three limit cycles does in fact cross the $\beta = 0$ axis, confirming our interpretation of the computer-generated 'chaos'. For $\beta > 0$ the model also shows multistability in the physical region which is not shown by the classical model with $\beta = 0$. The various bifurcation loci for $\beta \to 0^+$ do *not* uniformly approach the corresponding loci for $\beta = 0$ in the classical model.

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