Three limit cycles for a first-order exothermic chemical reaction in a continuous stirred-tank reactor *

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We study a first-order exothermic chemical reaction in a continuous stirred-tank reactor modelled by a 3-parameter family of vector fields in $\mathbb{R}^2$. We prove that there exist regions in $\mathbb{R}^3$ which contain points that depend on parameters such that the chemical reaction has 0, 1, 2, or 3 small amplitude limit cycles that surround the origin. We conclude that this model can reach two stable small amplitude limit cycles. Finally, we show that one of these regions contains the point in the parameter space considered by Gurel and Lapidus [6] who proved numerically the existence of one stable limit cycle.

1. Introduction

In many important physical systems, the dynamical system equations are in the form of autonomous coupled nonlinear ordinary differential equations such that

$$\frac{\mathrm{d}x}{\mathrm{d}t} = \dot{x} = f(x, y, \beta),$$
$$\frac{\mathrm{d}y}{\mathrm{d}t} = \dot{y} = g(x, y, \beta),$$

where $\beta = (\beta_1, \ldots, \beta_n)$ denotes the physical parameters of the system.

It is often desirable to obtain conditions under which all orbits are attracted to a single equilibrium point. The possibility of a cyclic behaviour, which is reflected in the existence of periodic solutions, is also of considerable interest (see, e.g., [8]).

There are many parameters that influence the behaviour of the industrial processes such as the chemical composition, temperatures of the input and output fluxes, the presence of chemical reactions, and others. Nevertheless, processes are designed to

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be stable, that is, they must show a stable behaviour when the parameters of the system change. In many cases, the variation of the parameters can be very significant and, eventually, the process becomes unstable. In order to avoid this instability, it is necessary to add a control system to the process that must be capable to ensure the stability of the process.

We study in this article an open-stirred tank cooled by a water stream which has a chemical reaction. The model has been deduced from the mass and energy conservation principles, the former taking into account that the flux entering the tank has a concentration equal to c_0 and the flux that exists from the tank has a concentration c in the reactive. Both fluxes were assumed the same in order to satisfy the hypothesis of zero accumulation. The occurrence of a chemical reaction was also assumed where a reactive A transforms into a product B according to an n-order kinetics that can be described by $A^n \to B$. The reaction rate may be represented as $\mathrm{d}c/\mathrm{d}t = kc^n$, where $k = k_0 \mathrm{e}^{-E/RT}$ is a quantity that increases with temperature, k_0 a constant, E the activation energy for the reaction, R the universal gas constant, and T the absolute temperature. The energy balance assumes an exothermic reaction with an enthalpy change ΔH. The inner cooling water has a temperature T_1 and the outer water a temperature T_2. The heat exchange between the water and the reactor occurs over an area A_c of the wall being this transfer characterised through the heat transfer coefficient h. Then the heat flux has a value $hA_\mathrm{c}(T - \overline{T})$, where $\overline{T} = (T_1 + T_2)/2$. The example analysed includes a proportional control $q_\mathrm{c} = K(T - T_\mathrm{s})$ that is externally applied to the inner cooling water and acts through a heat flow.

We let c_s and T_s denote the equilibrium values that correspond to no control, that is, $q_\mathrm{c} = 0$.

With all these assumptions, it is possible to write the mass conservation and the energy balance as follows (see figure 1):

$$\begin{aligned} V\frac{\mathrm{d}c}{\mathrm{d}t} &= Q(c - c_0) - kVc^n, \\ V\rho C_p \frac{\mathrm{d}T}{\mathrm{d}t} &= Q\rho C_p(T_0 - T) + \Delta H kVc^n - (hA + q_\mathrm{c})\big(T - \overline{T}\,\big), \end{aligned} \tag{1}$$

where $\overline{T}$ corresponds to the average temperature of the cooling water. It was assumed that no changes occur in the density along the process, that is, the densities of the inner and outer streams are similar.

After the following change of variables and normalisation,

$$x = \frac{c_\mathrm{s} - c}{c_\mathrm{s}}, \qquad y = \frac{T_\mathrm{s} - T}{T_\mathrm{s}}, \qquad t = \frac{t^* Q}{V}\,\frac{t^*}{\tau}, \qquad \beta_1 = \tau c_\mathrm{s}^{n-1} \mathrm{e}^{m-\theta},$$

$$\beta_2 = n, \qquad \beta_3 = \frac{E}{RT_\mathrm{s}}, \qquad \beta_4 = 1 + \frac{\tau h A}{V\rho C_p}, \qquad \beta_5 = \frac{\Delta H \tau c_\mathrm{s}^n}{\rho C_p T_\mathrm{s}} \mathrm{e}^{m-\theta},$$

$$\beta_6 = \frac{\tau K T_\mathrm{s}}{V\rho C_p}, \qquad \beta_7 = 1 - \frac{\overline{T}}{T_\mathrm{s}}$$

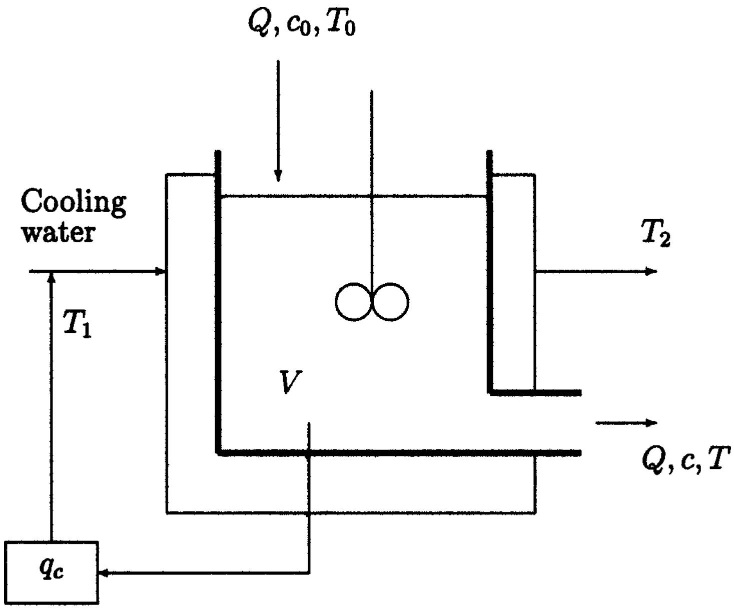

Figure 1. Continuous stirred-tank reactor chemical reaction $A^n \to B$.

the set of equations that models the above reaction is

$$\begin{aligned} \dot{x} &= -x + \beta_1 \left[(1-x)^{\beta_2} \exp\left(\frac{\beta_3 y}{y-1} \right) - 1 \right], \\ \dot{y} &= -\beta_4 y - \beta_5 \left[(1-x)^{\beta_2} \exp\left(\frac{\beta_3 y}{y-1} \right) - 1 \right] - \beta_6 y(\beta_7 - y), \end{aligned} \tag{2}$$

where the β_i, for $i = 1, \ldots, 7$, denote the physical parameters of the system.

Both Aris and Amundson [1] and Gurel and Lapidus [6] have shown that, for a first-order exothermic chemical reaction in a continuous stirred-tank reactor, the introduction of a proportional control can lead to a stable limit cycle where temperature and concentration oscillate continuously. For the particular values $(\beta_1, \beta_2, \beta_3, \beta_4, \beta_5, \beta_6, \beta_7) = (1.0, 1.0, 25.0, 2.0, 0.25, 16.0, 0.125)$, using numerical arguments, Gurel and Lapidus [6] showed the existence of a stable limit cycle.

We prove that there exist four regions in the 3-parameter space such that if we take a point in each region, system (2) has, respectively, 0, 1, 2, or 3 small amplitude limit cycles around the origin.

In the particular case of above, we consider the numerical values β_i for $i = 1, 2, 3, 7$. Let us discuss the dynamical behaviour of (2) in the parameter space $\lambda = (\beta_4, \beta_5, \beta_6) \in \mathbb{R}^3_+$, with $\beta_5 > 4/25$. Let X_λ be the respective C^∞-vector field of (2).

We have that

$$DX_\lambda(0,0) = \begin{pmatrix} -2 & -25 \\ \beta_5 & -\beta_4 + 25\beta_5 - \dfrac{\beta_6}{8} \end{pmatrix}$$

and that

$$\det DX_\lambda(0,0) = 2\beta_4 - 25\beta_5 + \frac{\beta_6}{4},$$
$$\operatorname{tr} DX_\lambda(0,0) = -\beta_4 + 25\beta_5 - \frac{\beta_6}{8} - 2.$$

Now if

$$\Sigma_0^+ = \{\lambda \in \mathbb{R}_+^3 \mid \det DX_\lambda(0,0) > 0\}, \quad \text{and}$$
$$\Sigma_1^{\mathrm{sg}(\iota)} = \{\lambda \in \mathbb{R}_+^3 \mid \mathrm{sg}(\operatorname{tr} DX_\lambda(0,0)) = \mathrm{sg}(\iota)\}, \quad \text{with } \iota \in \{-1,0,1\},$$

then $\operatorname{Div} X_\lambda(0,0) \equiv 0$ if $\lambda \in \Sigma_1^0$.

If $\lambda \in \Sigma_0^+ \cap \Sigma_1^0$, then the singularity at the origin of X_λ is a weak focus of at least order one.

To compute the Liapunov quantities on the nonhyperbolic focus at the origin and give a simple description of the bifurcations diagram in the parameter space, it is necessary to reduce (2) to a normal form (see, e.g., Blows and Lloyd [3], Dumortier [4]).

In the parameter space, we consider the function $\varphi : \mathbb{R}^2 \to \mathbb{R}^3$ given by

$$\varphi(\alpha, \beta_6) = \left(\alpha^2 - \frac{\beta_6}{8} + 2, \frac{\alpha^2 + 4}{25}, \beta_6\right) = \lambda,$$

where α is a new real parameter such that $\alpha^2 = 25\beta_5 - 4$ and that $(\alpha, \beta_6) \in \varphi^{-1}(\Sigma_1^0)$. Consider the change of coordinates $\psi : \mathbb{R}^2 \times \mathbb{R} \to \mathbb{R}^2 \times \mathbb{R}$ which includes a rescaling of the time and is given by

$$\psi(u, v, \tau) = \left(-2u - \alpha v, \frac{\alpha^2 + 4}{25} v, \frac{\tau}{|\alpha|}\right) = (x, y, t).$$

The C^∞-vector field obtained is $Y_\xi = (D\psi)^{-1} X_\xi \psi$, with $\xi = \varphi^{-1}(\lambda)$, and the linear part is

$$DY_\xi(0,0) = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}.$$

It is well known that the order of the fine focus at the origin depends on the coefficients of the Taylor series of the vector field at the origin. Therefore, we consider

$$Y_\xi = P_\xi \frac{\partial}{\partial u} + Q_\xi \frac{\partial}{\partial v}, \tag{3}$$

where

$$\begin{aligned} P_\xi(u,v) &= -v + \sum_{i,j=1}^{7} A_{ij} u^i v^j + \text{higher order terms}, \\ Q_\xi(u,v) &= u + \sum_{i,j=1}^{7} B_{ij} u^i v^j + \text{higher order terms}. \end{aligned} \tag{4}$$

Table 1
Polynomial expressions for $l_2,\ l_3$ and l_4.

$l_2(\alpha,\beta_6)$	$-64-951\alpha+368\alpha^2+481\alpha^3-529\alpha^4-32\beta_6+92\alpha^2\beta_6-4\beta_6^2$
$l_3(\alpha,\beta_6)$	$4532608+53874210\alpha+33772392\alpha^2-63845626\alpha^3$
	$-88551694\alpha^4+139203387\alpha^5+38225080\alpha^6-104000871\alpha^7$
	$+36099489\alpha^8+2530496\beta_6+8978880\alpha\beta_6+309404\alpha^2\beta_6$
	$-26117448\alpha^3\beta_6+2602444\alpha^4\beta_6+24276408\alpha^5\beta_6-12556344\alpha^6\beta_6$
	$+448408\beta_6^2+969516\alpha\beta_6^2-895112\alpha^2\beta_6^2-1862796\alpha^3\beta_6^2$
	$+1637784\alpha^4\beta_6^2+33024\beta_6^3+46800\alpha\beta_6^3-94944\alpha^2\beta_6^3+2064\beta_6^4$
$l_4(\alpha,\beta_6)$	$-429062556416-5167843081924\alpha-8969331235664\alpha^2$
	$+5084299738179\alpha^3+26702365686584\alpha^4-25269607477799\alpha^5$
	$-13948744494121\alpha^6+3993289637399\alpha^7+23286367661444\alpha^8$
	$+935337872833\alpha^9-26565150500845\alpha^{10}+16819040721012\alpha^{11}$
	$-2862718021482\alpha^{12}-283530691968\beta_6-1784447071296\alpha\beta_6$
	$-1891575047504\alpha^2\beta_6+5293618404864\alpha^3\beta_6+2965147558100\alpha^4\beta_6$
	$-3469439233792\alpha^5\beta_6-6954642315368\alpha^6\beta_6+3054804891876\alpha^7\beta_6$
	$+8216198839132\alpha^8\beta_6-7248578533752\alpha^9\beta_6+1493592011208\alpha^{10}\beta_6$
	$-71208378544\beta_6^2-309009271104\alpha\beta_6^2-41839325204\alpha^2\beta_6^2$
	$+639287074920\alpha^3\beta_6^2+491828355872\alpha^4\beta_6^2-783402414888\alpha^5\beta_6^2$
	$-880494239604\alpha^6\beta_6^2+1249483594272\alpha^7\beta_6^2-324693915480\alpha^8\beta_6^2$
	$-9575428096\beta_6^3-27595625536\alpha\beta_6^3+4989543008\alpha^2\beta_6^3$
	$+62190757808\alpha^3\beta_6^3+32690458048\alpha^4\beta_6^3-107682159552\alpha^5\beta_6^3$
	$+37645671360\alpha^6\beta_6^3-796485376\beta_6^4-1543767552\alpha\beta_6^2$
	$+273326336\alpha^2\beta_6^4+4639720512\alpha^3\beta_6^4-2455152480\alpha^4\beta_6^4$
	$-29703168\beta_6^5-79958400\alpha\beta_6^5+85396608\alpha^2\beta_6^5-1237632\beta_6^6$

Consider $\xi\in\varphi^{-1}(\Sigma_1^0)$. The coefficients of (4) are functions of the parameters α and β_6, that is,

$$A_{ij}=A_{ij}(\alpha,\beta_6)\quad\text{and}\quad B_{ij}=B_{ij}(\alpha,\beta_6).$$

Using the software for symbolic calculus of Mathematica [10], if L denotes the Liapunov quantities of the singularity at the origin of (3) (see, e.g., Blows and Lloyd [3], Lloyd et al. [7], Guiñez et al. [5]), then we obtain:

$$L_1=0,\quad L_2=\frac{(4+\alpha^2)^2l_2}{10^4\alpha^2},\quad L_3=\frac{(4+\alpha^2)^4l_3}{9\cdot 10^8\alpha^4}\quad\text{and}\quad L_4=\frac{(4+\alpha^2)^6l_4}{108\cdot 10^{12}\alpha^6},$$

where the polynomial expressions for $l_2,\ l_3$ and l_4 are shown in table 1.

2. Main results

Let Ω_1 and Ω_2 be defined by

$$\Omega_1=\left\{(\alpha,\beta_6)\,\middle|\,\alpha\geqslant\sqrt{\frac{951}{481}},\ \beta_6>0\right\},$$
$$\Omega_2=\left\{(\alpha,\beta_6)\,\middle|\,-\sqrt{\frac{951}{481}}\leqslant\alpha\leqslant 0,\ \beta_6>0\right\}.$$

Lemma 1. There exist five points F_i, with $i = 1, \dots, 5$, in the parameter space $\varphi^{-1}(\Sigma_0^+ \cap \Sigma_1^0)$ such that:

- if
$$F_3 \in \Omega_1 \cap l_2^{-1}(0) \cap l_3^{-1}(0) \quad \text{or} \quad F_5 \in \Omega_2 \cap l_2^{-1}(0) \cap l_3^{-1}(0),$$
then the singularity at the origin of (3) is an attracting fine focus of order three;
- if
$$F_1, F_2 \in \Omega_1 \cap l_2^{-1}(0) \cap l_3^{-1}(0) \quad \text{or} \quad F_4 \in \Omega_2 \cap l_2^{-1}(0) \cap l_3^{-1}(0),$$
then the singularity at the origin of (3) is a repulsive fine focus of order three.

Lemma 2. In the parameter space $\varphi^{-1}(\Sigma_0^+ \cap \Sigma_1^0)$, for each point F_i with $i = 1, \dots, 5$, there exist open sets $\mathcal{N}_{i_j}$, for $j = 0, 1, 2$, such that if $\xi \in \mathcal{N}_{i_j}$, then the vector field Y_ξ has j small amplitude limit cycles surrounding the origin.

Theorem 1. There exists an open set $\mathcal{R}_\lambda$ in the parameter space Σ_0^+ such that if $\lambda \in \mathcal{R}_\lambda$, then the vector field X_λ has at least three concentric small amplitude limit cycles surrounding the origin and the system can reach two stable small amplitude limit cycles.

Theorem 2. There exists a bifurcation surface $\mathcal{S}_\lambda \subset \Sigma_0^+$ that divides the parameter space into regions such that if $\lambda \in \mathcal{S}_\lambda$, then the vector field X_λ has one semistable limit cycle or two limit cycles, being one of them a semistable limit cycle and the other a hyperbolic limit cycle. All of the limit cycles surround the origin.

3. Proof of the main results

Proof of lemma 1

Since $l_2(\alpha, \beta_6) = -64 - 951\alpha + 368\alpha^2 + 481\alpha^3 - 529\alpha^4 - 32\beta_6 + 92\alpha^2\beta_6 - 4\beta_6^2$ is a quadratic polynomial in the parameter β_6, the discriminant is $\Delta(\alpha) = 16\alpha(-951 + 481\alpha^2)$. Now if $\Delta(\alpha) < 0$, then there are no real zeros of l_2, namely, when $\alpha < -\sqrt{951/481}$ or $0 < \alpha < \sqrt{951/481}$.

If $\alpha \geqslant \sqrt{951/481}$, then the zeros of the polynomial l_2 and the straight line $\alpha = \sqrt{951/481}$ are tangent at the point
$$\left(\sqrt{\frac{951}{481}}, \frac{18025}{962}\right)$$
and
$$l_2(\alpha, 0) = -64 - 951\alpha + 368\alpha^2 + 481\alpha^3 - 529\alpha^4 < 0.$$
Therefore, there exists a branch of $l_2^{-1}(0)$ in Ω_1.

If $-\sqrt{951/481} \leqslant \alpha \leqslant 0$, then the graph of the equation $l_2 = 0$ with the straight lines $\alpha = 0$ and $\alpha = -\sqrt{951/481}$ are tangent at the points

$$(0, -4) \quad \text{and} \quad \left(-\sqrt{\frac{951}{481}}, \frac{18025}{962}\right),$$

respectively. It is easy to see that there exists another branch of $l_2^{-1}(0)$ that is a closed curve at Ω_2.

Then

$$\Omega_1 \cap l_2^{-1}(0) \cap l_3^{-1}(0) = \{F_1, F_2, F_3\} \quad \text{and} \quad \Omega_2 \cap l_2^{-1}(0) \cap l_3^{-1}(0) = \{F_4, F_5\},$$

where the F_i, with $i = 1, \ldots, 5$, are given by

$$\begin{aligned} F_1 &\approx (1.52029147891315, 30.39571075638284), \\ F_2 &\approx (2.211371456535958, 24.40477641046841), \\ F_3 &\approx (4.497343282263827, 327.9436638653552), \\ F_4 &\approx (-0.5749216533159115, 10.47056163965918), \\ F_5 &\approx (-0.921320620660345, 16.94203529778311). \end{aligned}$$

Figure 2 shows, qualitatively, the position of the above points F_i, with $i = 1, \ldots, 5$, on the two connected components of $l_2(\alpha, \beta_6)$ in Ω_1 and Ω_2, respectively.

Furthermore,

$$L_4(F_1) > 0, \quad L_4(F_2) > 0, \quad L_4(F_3) < 0, \quad L_4(F_4) > 0 \quad \text{and} \quad L_4(F_5) < 0,$$

thus, the singularity at the origin of system (2) is a repulsive weak focus of order three at the points F_1, F_2 and F_4 and an attracting weak focus of order three at the points F_3 and F_5. The proof is now complete.

Proof of lemma 2

Lemma 1 stated that, for the points F_3 and F_5, the origin is an attracting fine focus of order three and that, for the points F_1, F_2, and F_4, the origin is a repulsive fine focus of order three. Hence, we only prove the lemma for F_5, since the proofs for the other cases are essentially the same. The bifurcation diagrams are shown in figures 3–5.

Let $\varepsilon > 0$ be sufficiently small. We consider the point $F_5 = (\alpha, \beta_6)$ as in figure 2.

We then have that:

(a) When $(\alpha - \varepsilon, \beta_6(\varepsilon)) \in \Omega_2 \cap l_2^{-1}(0)$, we obtain a Hopf bifurcation; that is, the unique hyperbolic attracting small amplitude limit cycle bifurcates and the singularity at the origin is a fine focus of order two.

(b) When $(\alpha + \varepsilon, \beta_6(\varepsilon)) \in \Omega_2 \cap l_2^{-1}(0)$, the stability at the origin of (3) does not change. Since $L_3(\alpha + \varepsilon, \beta_6(\varepsilon)) < 0$, the singularity at the origin is an attracting fine focus of order two.

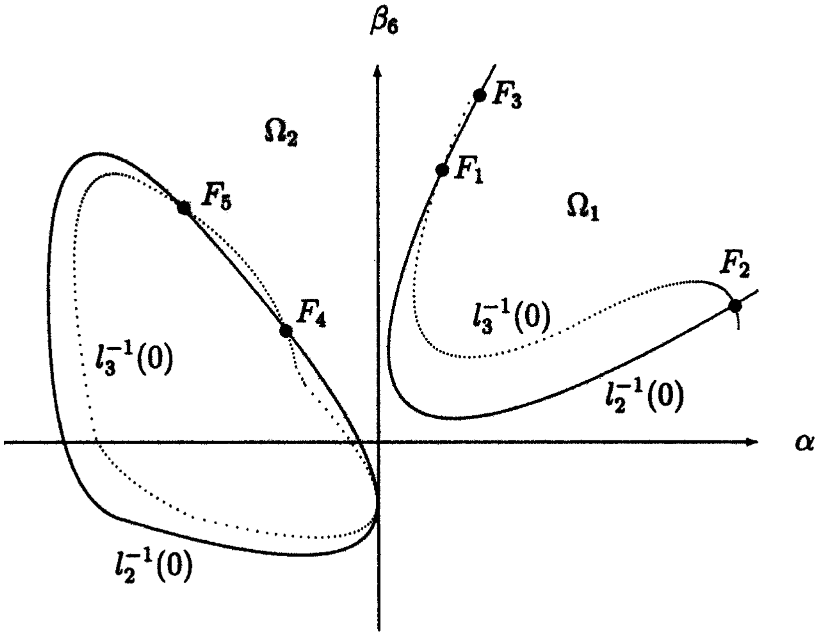

Figure 2. The position of the points F_i, with $i = 1, \ldots, 5$, in the parameter space such that the origin of (2) is a fine focus.

(c) When $(\alpha - \varepsilon, \beta_6(\varepsilon) + \varepsilon) \in \Omega_2 - l_2^{-1}(0)$ and $(\alpha - \varepsilon, \beta_6(\varepsilon))$ is as in (a) of above, we have that $L_2(\alpha - \varepsilon, \beta_6(\varepsilon) + \varepsilon) < 0$, hence, the stability at the origin of (3) is reversed. Thus, another Hopf bifurcation occurs. Therefore, there exists an open set $\mathcal{N}_{5_2}$ such that if $\xi \in \mathcal{N}_{5_2}$, then the vector field has two hyperbolic small amplitude limit cycles, namely, a repelling limit cycle in the interior and an attracting limit cycle in the exterior: the singularity at the origin is an attracting fine focus of order one.

(d) When $(\alpha + \varepsilon, \beta_6(\varepsilon) + \varepsilon) \in \Omega_2 - l_2^{-1}(0)$ and $(\alpha + \varepsilon, \beta_6(\varepsilon))$ is as in (b) of above, we have that $L_2(\alpha + \varepsilon, \beta_6(\varepsilon) + \varepsilon) < 0$, hence, the stability at the origin of (3) does not change and the singularity at the origin is an attracting fine focus of order one. Then there exists an open set $\mathcal{N}_{5_0}$ such that if $\xi \in \mathcal{N}_{5_0}$, then the vector field has no small amplitude limit cycle.

(e) When $(\alpha - \varepsilon, \beta_6(\varepsilon) - \varepsilon) \in \Omega_2 - l_2^{-1}(0)$ and $(\alpha - \varepsilon, \beta_6(\varepsilon))$ is as in (a) of above, we have that $L_2(\alpha - \varepsilon, \beta_6(\varepsilon) - \varepsilon) > 0$, hence, the stability at the origin of (3) does not change, that is, the singularity at the origin is a repelling fine focus of order one. The limit cycle obtained in (a) of above persists, since it is hyperbolic. Then there exists an open set $\mathcal{N}_{5_1}$ such that if $\xi \in \mathcal{N}_{5_1}$, then the vector field has one small amplitude limit cycle.

(f) When $(\alpha + \varepsilon, \beta_6(\varepsilon) - \varepsilon) \in \Omega_2 - l_2^{-1}(0)$ and $(\alpha + \varepsilon, \beta_6(\varepsilon))$ is as in (b) of above, we have that $L_2(\alpha + \varepsilon, \beta_6(\varepsilon) - \varepsilon) > 0$, hence, the stability at the origin of (3) is reversed. Thus, we have a Hopf bifurcation; that is, the unique hyperbolic attracting small amplitude limit cycle bifurcates and the singularity at the origin is a fine focus of order one. Therefore, there exists an another open set $\mathcal{N}_{5_1}$ such that if $\xi \in \mathcal{N}_{5_1}$, then the vector field has one small amplitude limit cycle, which completes the proof for F_5.

The proof of the lemma now follows.

Since at the point F_3 there is an attracting fine focus of order three, and since at the points F_1, F_2 and F_4 there are repelling fine foci of order three, the proof is essentially the same as that of above and the bifurcation diagrams are shown in figures 3–5.

Proof of theorem 1

By the proof of lemma 2 part (c), in the parameter space $\varphi^{-1}(\Sigma_0^+ \cap \Sigma_1^0)$ and given ε small enough, there exists $\mathcal{N}_{5_2}$ such that

$$\xi_\varepsilon = (\alpha - \varepsilon, \beta_6(\varepsilon) + \varepsilon) \in \mathcal{N}_{5_2}.$$

Then the vector field Y_{ξ_ε} has two concentric small amplitude limit cycles surrounding the origin.

Now, since

$$X_{\varphi(\xi_\varepsilon)} = (D\psi)Y_{\xi_\varepsilon}\psi^{-1},$$

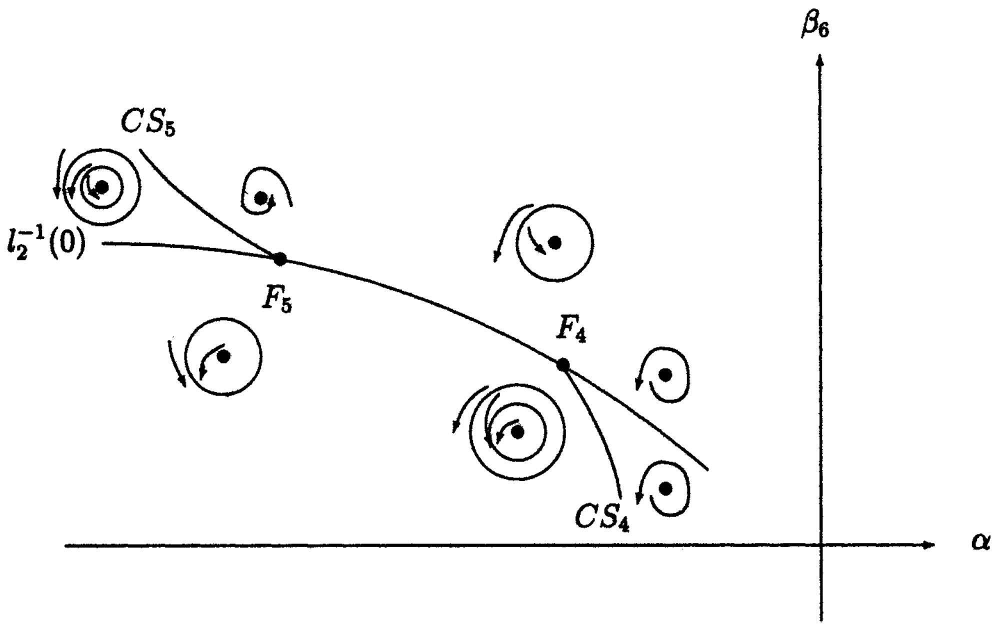

Figure 3. Bifurcation diagram of the origin of (2) in a neighborhood of F_4 and F_5 in Ω_2.

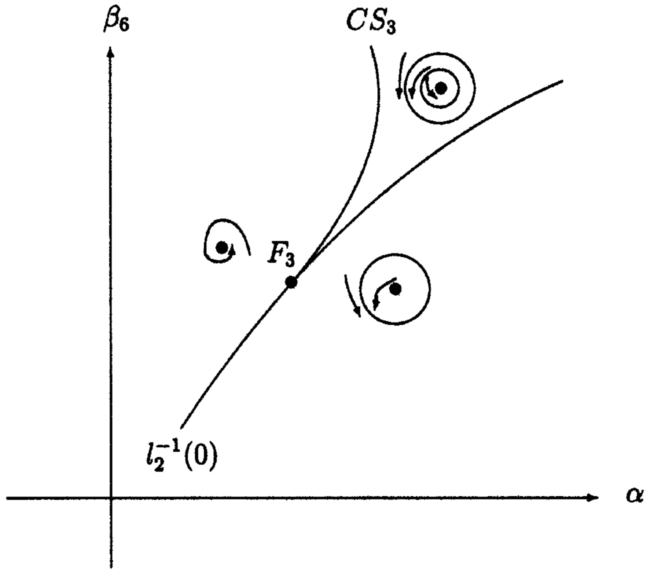

Figure 4. Bifurcation diagram of the origin of (2) in a neighborhood of F_3 in Ω_1.

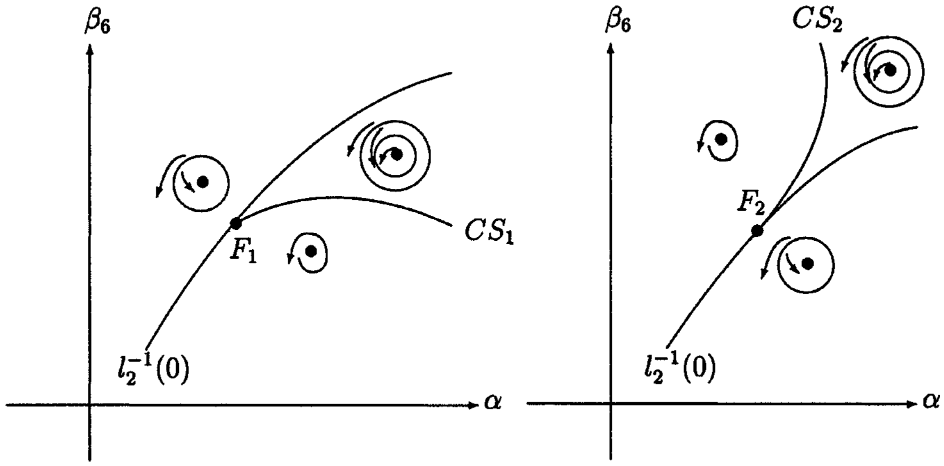

Figure 5. Bifurcation diagrams of the origin of (2) in a neighborhood of F_1 and F_2 in Ω_1, respectively.

the vector field $X_{\varphi(\xi_\varepsilon)}$ has two concentric small amplitude limit cycles around the origin where $\varphi(\xi_\varepsilon) = \lambda_\varepsilon \in \Sigma_1^0$. Furthermore, the singularity at the origin is an attracting fine focus of order one.

We next perturb the parameters β_4, β_5 and β_6 so that the new λ is given by

$$\lambda_\varepsilon^* \in \Sigma_1^+.$$

We have that the stability at the origin of $X_{\lambda_\varepsilon^*}$ is reversed and the singularity at the origin is a repelling hyperbolic focus, since

$$L_1 = \mathrm{tr}\, DX_{\lambda_\varepsilon^*}(0,0) > 0.$$

Therefore, we have a Hopf bifurcation, that is, the unique hyperbolic attracting limit cycle bifurcates. Note that by the hyperbolicity of the two limit cycles of lemma 2 part (c), they persist. Thus, in the parameter space Σ_1^+, there exists a neighbourhood $\mathcal{V}_{\delta_5}(F_5)$ of the point F_5, with $\delta_5 > 0$ sufficiently small, that contains the open set $\{\lambda_\varepsilon^* \mid 0 < \varepsilon \ll 1\}$ and $X_{\lambda_\varepsilon^*}$ has three hyperbolic small amplitude limit cycles.

Next, at the point F_3, the origin of the vector field is an attracting fine focus of order three, the proofs for the existence of an open set as well as that of the three limit cycles are analogous. Note that two of the limit cycles are stable. Analogously, in the neighbourhood of the points F_1, F_2 and F_4, where the origin of the vector field is a repelling fine focus of order three, it is possible to prove the existence of open sets such that the vector field has three limit cycles which have opposite stabilities. There now exists a set $\mathcal{R}$, which is the union of open sets such that if $\lambda \in \mathbb{R}$, then the vector field X_λ has at least three small amplitude limit cycles, and the proof is now complete.

Proof of theorem 2

We consider the point $F_5 \in \Omega_2 \cap l_2^{-1}(0) \cap l_3^{-1}(0)$. By lemma 1, the singularity at the origin is an attracting fine focus of order three.

Under this hypothesis and after time rescaling, by the theory of the normal forms in a neighbourhood of the origin, the vector field X_λ is equivalent to a vector field given by Takens [9, pp. 488–491] (case $k = 3$) and Arrowsmith and Place [2, pp. 215–217] (Type $(3, +)$). Therefore, there exists a bifurcation surface $\mathcal{S}_\lambda$ in Σ_0^+ that divides the parameter space into regions and if $\lambda \in \mathcal{S}_\lambda$, then two of the three limit cycles of theorem 1 collapse into a semistable limit cycle. (In figure 3 the curve CS_5 is the intersection between $\mathcal{S}_\lambda$ and Σ_1^0 when $(\alpha, \beta_6) \in \varphi^{-1}(\Sigma_1^0)$.)

Moreover, by parts (d) and (e) of the proof of lemma 2, if we perturb the parameters so that $\mathrm{tr}\, DX_\lambda(0,0) < 0$, then we have that, in case (d), the vector field has no small amplitude limit cycles and that, in case (e), it has two small amplitude limit cycles. Therefore, there exists a bifurcation surface $\mathcal{S}_\lambda$ in Σ_0^+ such that if the point λ crosses $\mathcal{S}_\lambda$, then the two limit cycles collapse into a semistable limit cycle and afterwards disappear.

Remark 1. The limit cycle found by Gurel and Lapidus [6] is located close to the point F_5; this limit appears when the parameters are perturbed from $(\alpha, \beta_6) \in \varphi^{-1}(\Sigma_1^0)$ in part (e) of lemma 2 to the sector where $\mathrm{tr}\, DX_\lambda(0,0) > 0$. Hence, the limit cycle is located in the open set $\mathcal{N}_{5_1}$.

Remark 2. We have proved that system (2) has three small amplitude limit cycles (see theorem 1); two of the cycles are stable and the other is unstable. Since systems (1) and (2) are equivalent, under the physical point of view, the process modelled by (1) is a stable one in the sense that, given a realistic initial condition other than the equilibrium

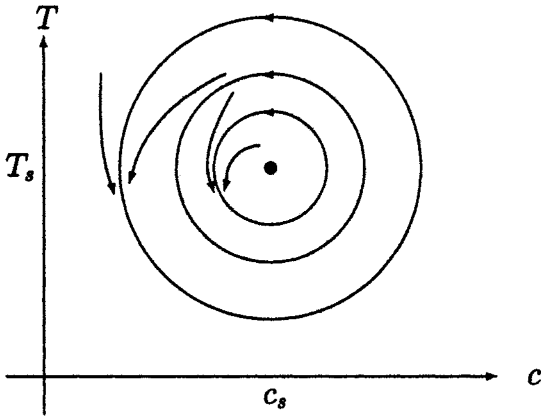

Figure 6. Three limit cycles for a first-order exothermic chemical reaction in continuous stirred-tank reactor.

point (c_s, T_s) and outside the unstable limit cycle, the corresponding solution tends to become stabilised in one of the two stable oscillating behaviours that the system has, as is qualitatively shown in figure 6.

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