Dynamics of a Two-Dimensional Model for Electrochemical Corrosion Using Feedback Circuit and Nullcline Analysis

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In this paper we analyze a two-dimensional model of electrochemical corrosion by combining two qualitative approaches: a method based on the feedback circuits found in the Jacobian matrix and a nullcline technique. This leads us to map out regions of the parameter space where the system exhibits sustained oscillatory behavior and/or multistationarity. The results are confirmed by numerical integration.

1. Introduction

Various authors have addressed the role of feedback circuits (or feedback loops) in dynamical systems.[1−5] Here we refer mainly to the methodology and terminologies introduced by Thomas et al. [refs 6−8 and references therein]. Given a differential system, a feedback circuit is a combination of terms of its Jacobian matrix under a circular permutation of the indices. Whenever these terms have fixed signs (positive or negative), the circuit is positive (negative) for an even (odd) number of negative terms.

Previous works have shown that at least one positive circuit is necessary to have multistationarity, whereas at least one negative circuit of at least two elements is necessary to have sustained oscillations.[6,9−12] Even more complex behavior, including multiple periodicity and chaos, might be understood as resulting from the combined effects of several feedback circuits.[13,14] However more systems need to be investigated to fully understand the role of feedback circuits in the generation of complex dynamical behavior.

In section 2 we give the settings and working definitions of our approach, encompassing some earlier works such as refs 9 and 11. Section 3 describes the feedback circuit analysis of a two-dimensional model for electrochemical corrosion. This analysis is complemented by a nullcline-based method. The results are confirmed by numerical integration. The implications of this work are discussed in section 4.

2. Preliminairies

2.1. Definitions. Consider a dynamical system (generally nonlinear) described by

$$\dot{x} = F(x), \quad x = (x_1, ..., x_n) \in \mathrm{R}^n \tag{2-1}$$

with component functions F_i, $i = 1, 2, ..., n$ differentiable in a region U of the variables space, not necessarily a neighborhood of a steady state. The general Jacobian matrix at $x = (x_1, ..., x_n)$ is given by

$$A = \left[\frac{\partial F_i}{\partial x_j}(x)\right] = [a_{ij}], \quad 1 \le i, j \le n \tag{2-2}$$

and in general depends on the state variables. We assume the terms a_{ij} have constant sign ϵ_{ij} in U. For Λ the set of indices 1, ..., n we denote by $I_k = \{i_1, ..., i_k\}$ an ordered subset of k different elements of Λ and by $J_k = \sigma(I_k) = \{j_1, ..., j_k\}$, with σ a permutation of I_k. A nonzero product

$$P_k = P(I_k, J_k) = \prod_{l=1}^{l=k} a_{i_l\sigma(i_l)} = a_{i_1\sigma(i_1)}a_{i_2\sigma(i_2)}...a_{i_k\sigma(i_k)} \tag{2-3}$$

is called a *circuit product.* The sign ϵ_{ij} determines the nature of the influence of element x_i on element x_j (on its own subsequent evolution for $i = j$).

If the product P_k has a constant positive (negative) sign, then the corresponding k-element feedback circuit L_k is positive (negative). This is the case for an even (odd) number of negative factors in P_k.

A k-dimensional feedback circuit L_k is represented by an interaction graph G_k of k vertices and edges (x_i, x_j, ϵ_{ij}). We derive from A a *qualitative Jacobian* A_q containing exclusively the signs ϵ_{ij}. The feedback circuits method is based on the determination and analysis of this qualitative Jacobian matrix, anywhere in the phase space, not only around the steady states (if any). (See, for example, ref 6).

2.2. Stability Criteria and Feedback Circuits. We describe here briefly the relationship between feedback circuits and classical dynamical analysis. (See also refs 2, 9, and 11). We write the characteristic equation as

$$P_\lambda(A) = |\lambda I - A| = \lambda^n + c_1\lambda^{n-1} + ... + c_k\lambda^{n-k} + ... + c_{n-1}\lambda + c_n = 0 \tag{2-4}$$

where the coefficients c_k, $k = 1, ..., n$ are the sum of the diagonal minors of order k of the matrix A. (See, for example, ref 15.) On the other hand, upon expanding the Jacobian determinant A, any k-dimensional principal subdeterminants with indices in I_k may be displayed in the form

$$D(I_k) = \sum_{J_k} (-1)^{\epsilon} P(I_k, J_k) \tag{2-5}$$

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over all permutations J_k, where ϵ is the number of even-dimensional components of P_k. Letting

$$S_k = \sum_{I_k} D(I_k) \tag{2-6}$$

over all k-principal subdeterminants in A, it readily appears that

$$(-1)^k S_k = c_k, \quad k = 1, 2, ..., n \tag{2-7}$$

Therefore sign and magnitude of circuits, and hence the type of feedback circuits, are closely related to the eigenvalues of A. Note also that every k-element circuit L_k is represented in S_k with a signature $\tau = (-1)^{k+1}$, i.e., negative for even dimensional and positive for odd dimensional circuit. Routh-Hurwitz stability criteria, that is, negative real parts of the eigenvalues as necessary and sufficient conditions of stability, imply

$$(-1)^k S_k > 0, \quad S_3 > S_1 \times S_2 \tag{2-8}$$

Moreover, if $S_1 = 0$, then $S_{2r+1} = 0$, $r = 1, ..., (n - 1)$.

Some important consequences are easily derived as follows. (See also ref 11.) Denote L_R the circuit structure found in the matrix A, corresponding to the region R where it has constant entries.

(1) Using the signature argument, positive circuits, having a "negative" contribution to S_k, always tend to destabilize a steady state. Hence their presence promotes instability in the system.

(2) Asymptotic stability, with $S_1 < 0$, requires at least one negative one-dimensional circuit, whereas L_R must contain at least one negative circuit of two-dimensions or higher to satisfy $S_3 < 0$.

2.3. Dynamical Role of Feedback Circuits. *2.3.1. Multistationarity*. It has been established that a positive feedback circuit is a necessary condition for multistationarity. In fact, a positive feedback circuit ensures instability in the system by causing the real part of some eigenvalues to become positive. See refs 6, 9, and 11 for more details.

2.3.2. Periodic Behavior. Assume the vector field F in R^n is C^1 and *positive quasi-monotonic*, that is, $\partial F_i/\partial x_j \geq 0$ whenever $i \neq j$. This implies all feedback circuits are positive except possibly the one-element ones. Such a vector field has a monotonic solution flow. In particular there are no attracting periodic orbits.[16,17] An immediate important consequence is that, in the absence of negative circuits other than one-element circuits, the system cannot have any stable periodic behavior such as stable limit cycles. See also ref 10.

In summary negative feedback circuits typically promote oscillations, whereas multistationarity is promoted by positive circuits. Whenever they actually fulfill this dynamical role, they are said to be *functional*.[7,8]

3. Model For Electrochemical Corrosion

We study the following electrochemical corrosion model initially developed by Talbot and Oriani.[18] A metal M is dissolving in an electrolyte solution. Any given point on the surface of the metal at any given time is either bare or covered with adsorbed MOH. The adsorbed metal hydroxide (MOH) passivates the underlying metal. Under appropriate parameter conditions this model system reproduces the salient features of the dynamical behavior observed during potentiostatic dissolution of copper in an acetate buffer.[19] It is summarized by the following set of chemical reactions.

$$\mathrm{M} \xrightarrow{k_1} \mathrm{M}^+ + \mathrm{e}^-$$

$$\mathrm{M}^+ + \mathrm{H_2O} \underset{k_{-2}}{\overset{k_2}{\rightleftharpoons}} (\mathrm{MOH})_{aq} + \mathrm{H}^+$$

$$\mathrm{M}^+ + \mathrm{A}^- \xrightarrow{k_3} \mathrm{MA}$$

$$\mathrm{MOH_{aq}} \underset{k_{-4}}{\overset{k_4}{\rightleftharpoons}} \mathrm{MOH_{ad}} \tag{3-1}$$

It is assumed that A^-, H_2O, and H^+ are present in excess so that their concentrations can be assumed to remain constant. These concentrations are then absorbed into the rate constants. Since hydration is generally fast, the second reaction is assumed to be always in equilibrium; so $[\mathrm{MOH_{aq}}]$ is proportional to $[\mathrm{M}^+]$. We absorb the equilibrium constant into k_4. The two independent variables in the corrosion process are the concentration of metal ion, C, and the fractional coverage of MOH, θ_{OH}. Following Talbot and Oriani, we allow for the possibility that k_4 and k_{-4} may vary with θ_{OH}, and we express these rate constants as

$$k_4 = k'_4 \tilde{f}(\theta_{OH}), \quad k_{-4} = k'_{-4}\tilde{g}(\theta_{OH}) \tag{3-2}$$

where the functions $\tilde{f}$ and $\tilde{g}$ include any such dependence. Taking $C = [\mathrm{M}^+]$, we obtain

$$\frac{dC}{d\tau} = k_1\theta_M - k_3 C$$

$$\frac{d\theta_{OH}}{d\tau} = k'_4 C\theta_M \tilde{f}(\theta_{OH}) - [k'_{-4}\tilde{g}(\theta_{OH})]\theta_{OH} \tag{3-3}$$

where $\theta_M = 1 - \theta_{OH}$. It is convenient to rewrite these rate equations in terms of dimensionless quantities. The following dimensionless variables are defined:[18-20]

$$\tau = k'_{-4}t, \quad K = \frac{k'_4}{k'_{-4}}, \quad x = KC, \quad p = \frac{k_1 K}{k'_{-4}}, \quad q = \frac{k_3}{k'_{-4}} \tag{3-4}$$

In terms of these dimensionless variables, setting $y = \theta_{OH}$, and following Talbot and Oriani, we choose $\tilde{f}(y) = 1$ and $\tilde{g}(y) = e^{-\beta y}$ and obtain the two-dimensional model with three control parameters (p, q, β).

$$\dot{x} = p(1 - y) - qx = f(x, y)$$

$$\dot{y} = x(1 - y) - ye^{-\beta y} = g(x, y) \tag{3-6}$$

with the state variables x and y in the interval [0,1], for physical reasons. As discussed elsewhere (see refs 18−20), the model system exhibits interesting global dynamics as a function of the ratio p/q for $q = 0.001$ and β. Indeed, using stability analysis and numerical integration, it has been shown to exhibit bistability for appropriate parameter values adjacent to the oscillatory regime.[18,20] These dynamics are recovered here by a feedback circuit based method complemented by a nullcline analysis.

3.1. Feedback Circuit Analysis. Solving $f(x, y) = g(x, y) = 0$ for fixed points (steady states) of system 3-6 yields

$$x = l_p(y) = \frac{p}{q}(1 - y)$$

$$p(1 - y)^2 - qye^{-\beta y} = 0 \tag{3-7}$$

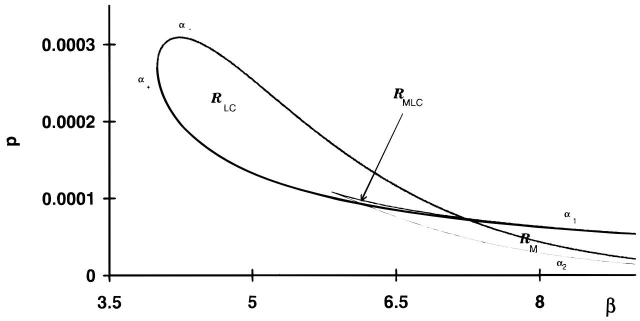

Figure 1. Mapping for $q = 0.001$. The region, called $\mathbf{R}_{LC}$, encompassed by the α_- and α_+ represents the area in parameter space where self-sustained oscillations occur. The region, called $\mathbf{R}_M$, encompassed by α_1 and α_2 represents the area in parameter space where multistationarity occurs. The overlap area, $\mathbf{R}_{MLC}$, is the region of coexistence of multistationarity and sustained oscillations.

with parameters $p > 0$, $q > 0$, $\beta > 0$, and $0 \le x,y < 1$. These equations define a region R_{ss} of steady states in the state space. We derive the mapping function

$$\alpha = \frac{p}{q} = F(\beta, y) = \frac{y e^{-\beta y}}{(1-y)^2} \qquad (3\text{-}8)$$

whose extrema are solutions of

$$\frac{d\alpha}{dy} = \alpha' = \frac{q(\beta y^2 + (1-\beta)y + 1)}{e^{\beta y}(1-y)^3} = 0 \qquad (3\text{-}9)$$

This admits the two distinct positive reals roots

$$y_{1,2} = \frac{-1 + \beta \mp \sqrt{\beta^2 - 6\beta + 1}}{2\beta} \qquad (3\text{-}10)$$

for the parameter values $\beta > 5.82$. Hence the function α is monotonic decreasing for $y_1 < y < y_2$, with a maximum $\alpha(\beta, y_1)$ at y_1 and a minimum $\alpha(\beta, y_2)$ at y_2. Therefore the mapping in the parameter space ($\alpha = p/q, \beta$) for multistationarity is bounded by

$$\alpha_1 = \alpha(\beta, y_1) = \frac{y_1 e^{-\beta y_1}}{(1-y_1)^2} \quad \text{and}$$

$$\alpha_2 = \alpha(\beta, y_2) = \frac{y_2 e^{-\beta y_2}}{(1-y_2)^2} \qquad (3\text{-}11)$$

This region is denoted $\mathbf{R}_M$ (see Figure 1) for $q = 0.001$ (region encompassed by α_1 and α_2.) In the region R_{ss} system 3-6 has the Jacobian matrix

$$A = \begin{pmatrix} -q & -p \\ 1-y & a_{22}(p, q, \beta) \end{pmatrix} \qquad (J)$$

with

$$a_{22}(p, q, \beta) = -x + (\beta y - 1)e^{-\beta y} = \frac{p}{q}\left(\frac{y-1}{y}\right)(\beta y^2 - \beta y + 1) \qquad (3\text{-}12)$$

The corresponding qualitative Jacobian matrix is

$$A_q = \begin{pmatrix} - & - \\ + & \text{sign}(a_{22}(p, q, \beta)) \end{pmatrix} \qquad (3\text{-}13)$$

Hence the circuit structure contains a negative one-element circuit L_x^- and a two-element negative L_{xy}^-. At this stage, the necessary condition for a (damped) periodic behavior is realized. Getting $a_{22}(p, q, \beta) > 0$ is the only way to ensure a positive circuit and, hence, multistationarity or sustained oscillations. Note from eq 3-12 that

$$\text{sign}(a_{22}(p, q, \beta)) = -\text{sign}(\beta y^2 - \beta y + 1) \qquad (3\text{-}14)$$

and the quadratic function

$$Q_\beta(y) = \beta y^2 - \beta y + 1 \qquad (3\text{-}15)$$

admits the two distinct positive real roots

$$Y_{1,2} = \frac{1}{2}\left(1 \mp \sqrt{1 - \frac{4}{\beta}}\right), \quad \text{for } \beta > 4 \qquad (3\text{-}16)$$

It entails $a_{22}(p, q, \beta) > 0$ for $\beta > 4$, and $Y_1 < y < Y_2$. From eq 3-10, this also holds true for multistationarity. Indeed we have $Y_1 < y_1 < y_2 < Y_2$. Hence the region $\mathbf{R}_M$ of multistationarity is embedded in the region $\mathbf{R}_{L+}$ of the parameter space with a positive circuit. Moreover, the mapping function $\alpha(\beta, y)$ is monotonic increasing for $Y_1 < y < y_1$ and $y_2 < y < Y_2$, but monotonic decreasing for $y_1 < y < y_2$. Hence the region $\mathbf{R}_{L+}$ of positive circuit is the union of $\mathbf{R}_M$ and the region bounded by

$$\alpha_- = \alpha(\beta, Y_1) = \frac{Y_1 e^{-\beta Y_1}}{(1-Y_1)^2} \quad \text{and}$$

$$\alpha_+ = \alpha(\beta, Y_2) = \frac{Y_2 e^{-\beta Y_2}}{(1-Y_2)^2} \qquad (3\text{-}17)$$

(see Figure 1) for $q = 0.001$. From stability analysis of bounded planar systems, perturbation of a unstable focus or node out of the region of multistationarity leads to sustained oscillations. Moreover, an unstable focus or node requires $P > 0$ and $S > 0$, where S and P are respectively sum and product of the eigenvalues.

$$P = p\,\frac{(1-y)}{y}(\beta y^2 + (1-\beta)y + 1)$$

$$S = a_{22}(p, q, \beta) - q \qquad (3\text{-}18)$$

Therefore, the multistationarity region $\mathbf{R}_M$ is given by $P < 0$, i.e., $\beta y^2 + (1-\beta)y + 1 < 0$. Moreover, $S > 0$ implies $a_{22}(p, q, \beta) > q > 0$. This entails that the parameter domain for $S > 0$ is included in $\mathbf{R}_{L+}$, where $a_{22}(p, q, \beta) > 0$. This domain tends to the region $\mathbf{R}_{L+}$ as q tends to 0, in particular for small values such as $q = 0.001$.

At this stage sustained oscillations may occur only in the region $\mathbf{R}_{L+}$. We then use the nullcline technique to actually map out the region $\mathbf{R}_{LC}$ of sustained oscillations, given in Figure 1. This region overlaps $\mathbf{R}_M$ yielding a region $\mathbf{R}_{MLC}$ of coexistence of multistationarity and sustained oscillations. For instance, taking $p = 0.0001$ and $\beta = 6.0$ for $q = 0.001$, we obtain two unstable nodes with a saddle point in between, surrounded by a limit cycle. See Figure 2.

3.2. Nullcline Analysis. Information about the global flow in state space can be obtained using the nullcline technique.[21-23] This technique is implemented in the two-dimensional corrosion model and complements the feedback circuit approach.

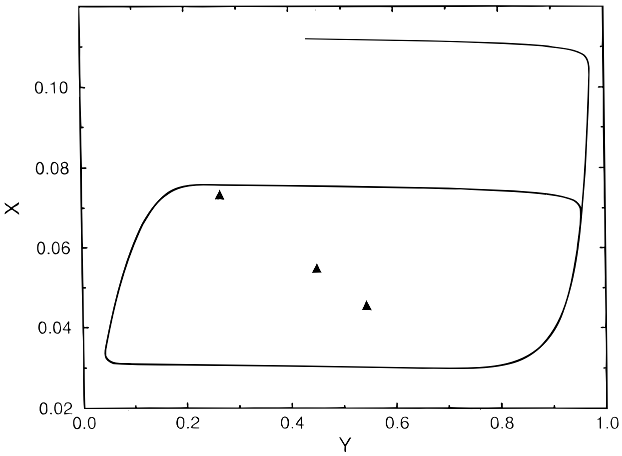

Figure 2. State space portrait for the parameters $p = 0.0001$, $q = 0.001$, and $\beta = 6.0$. It clearly indicates the coexistence of multistationarity (three fixed points indicated by $\triangle$) along with stable oscillatory dynamics (limit cycle) and is consistent with the predicted overlap region of Figure 1. Stability analysis reveals that all three fixed points are unstable, two unstable nodes and a saddle point.

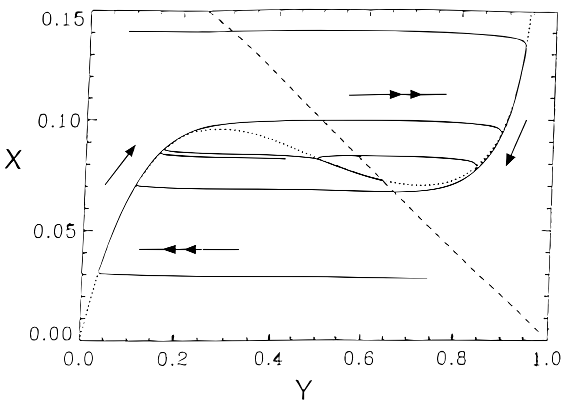

Figure 3. Global picture of the flow obtained from qualitative analysis of the two-dimensional system using nullclines. The dashed line is the nullcline for $f(x, y)$, and the dotted curve is the nullcline for $g(x, y)$. All the initial conditions converge to the large limit cycle since the fixed point solution is an unstable node. The system parameters are $p = 2 \times 10^{-4}$, $q = 1 \times 10^{-3}$, and $\beta = 5.0$.

Equating $f(x, y) = 0$ and $g(x, y) = 0$ in eq 3-6 yields the two nullclines for the model system. The points of intersection of these two nullclines are the fixed point solutions of the system. The flow in state space (global) can be obtained qualitatively from the shape of these nullclines.

Figure 3 shows the nullclines for our two-dimensional system for the following parameter values: $p = 2 \times 10^{-4}$, $q = 1 \times 10^{-3}$, and $\beta = 5.0$. At these parameter values and for typical values of $x = 0.1$ and $y = 0.4$ we have

$$\frac{dx}{dt} \ll \frac{dy}{dt} \tag{3-19}$$

thus making y the fast (dominant) variable (except for regions very near the nullcline $g(x, y)$). The double and the single arrows in Figure 3 represent the velocity vector fields for the fast (y) and slow (x) variables, respectively. The nullcline $g(x, y)$ has an unstable segment between its two extrema since the velocity vectors are directed away from it.

If the system is initialized at a point in state space far from either nullcline, it moves toward the nullcline $g(x,y)$ along a path parallel to the y axis under the influence of a velocity vector field for the fast variable y. Once the system reaches the nullcline $g(x, y)$, it moves along this nullcline toward the $f(x, y)$ nullcline until it reaches either an unstable segment of the $g(x, y)$ (as in Figure 3), or a stable fixed point at the intersection of $f(x, y)$ and $g(x, y)$. If the trajectory reaches an unstable segment of $g(x, y)$ before a fixed point is reached, then the system will move away from $g(x, y)$ in a direction parallel to the y axis toward a stable segment where the above process repeats.

An example of such a trajectory leading to a large stable limit cycle in the presence of an unstable fixed point is shown in Figure 3. When such large self-sustained oscillations are possible, the system is said to possess self-sustained excitability. In Figure 3 this large limit cycle is the only stable attractor; hence almost all the initial conditions are attracted to it.

It is also possible, for different parameter values, that the intersection of the nullclines is on the unstable segment and in close proximity to the extrema of the $g(x, y)$. The emerging limit cycle in this case is small and confined to the immediate neighborhood of this extrema. Such a system is said to exhibit self-sustained oscillations. All self-sustained excitable systems are also self-sustained oscillatory, but the converse does not hold.

3.2.1. Mapping of the Parameter Space. We apply the nullcline analysis discussed in the previous section to find the range in parameter space ($\alpha = p/q$, β), for small values of p and q, where the model equations have an oscillatory solution. Using eq 3-7, the defining condition for $f(x, y)$ (nullcline for the slow variable) yields

$$x = x_f(y) = -\frac{p}{q}(y - 1) \tag{3-20}$$

This is an equation for a straight line with slope $= -(p/q) = x/(y-1)$ and passing through $y = 1$. The other nullcline (for the dominant variable) $g(x, y) = 0$ gives

$$x = x_g(y) = \frac{ye^{-\beta y}}{(1 - y)} \tag{3-21}$$

The extrema for the nullcline $g(x, y)$ is calculated by equating $dx/dy = 0$. It entails the real roots

$$y_\pm = \frac{\beta \pm \sqrt{(\beta^2 - 4\beta)}}{2\beta} \tag{3-22}$$

of the quadratic function $\beta y^2 - \beta y + 1$ for the parameter values $\beta > 4$. Existence of an unstable segment for the nullcline $g(x, y)$, i.e., for $\beta \geq 4$, is a neccessary but not a sufficient condition for the emergence of oscillatory behavior.

Spontaneous oscillatory solutions exist when all the points of intersection for the two nullclines occur on the unstable segment of the nullcline $g(x, y)$. These oscillatory solutions then are required by the Poincaré–Bendixson theorem.[23]

From eq 3-20, the slope of the straight lines passing through the two extrema are

$$-\left(\frac{p}{q}\right)_{e1} = -\frac{x_{e1}}{(1 - y_{e1})} \quad \text{and} \quad -\left(\frac{p}{q}\right)_{e2} = -\frac{x_{e2}}{(1 - y_{e2})} \tag{3-23}$$

where (x_{e1}, y_{e1}) and (x_{e2}, y_{e2}) are the coordinates of the extrema for the nullcline $g(x, y)$ in the state space. If the slope of the nullcline $f(x, y)$ for the slow variable (x) is such that

$$\left(\frac{p}{q}\right)_{e1} < \text{slope} < \left(\frac{p}{q}\right)_{e2} \tag{3-24}$$

then all the fixed point solutions of the system are on the unstable segment of the nullcline for $g(x, y)$ and hence unstable. This results in the system exhibiting spontaneous oscillatory behavior by virtue of the Poincaré–Bendixson theorem.

This yields a range in the parameter space ($\alpha = p/q$, β), $q = 0.001$, where the two-dimensional system exhibits self-sustained oscillations. This range is identical to the one given in Figure 1. This has been verified by numerical integration for $q = 0.001$.

4. Results and Discussion

Usually, using stability analysis, one can only attain the local dynamics of the system, and hence one needs numerical integration in conjunction with stability analysis to obtain the global dynamics. However, in this paper, we were able to predict the global dynamics of a two-dimensional model related to electrochemical corrosion without resorting to actual integration of the system using a feedback circuit analysis (section 3.1) and an intuitive graphical analysis of the shape of the nullclines (section 3.2). Specifically, we were able to map out the regions in parameter space where the model system exhibits stable periodic oscillations or/and multistationarity. The results obtained using the two different techniques are consistent and correct (as verified by numerical integration of the system).

An important advantage of the feedback circuit analysis is that it links specific regulatory structures (feedback circuits) with specific dynamical properties. Here, for this two-dimensional system, we found that a single term $a_{22}(p, q, \beta)$ of the Jacobian matrix is crucial for both types of nontrivial behavior, multistationarity and limit cycles. However, in most of the parameter space, it is not possible to switch from one to the other type by changing β. As can be seen in Figure 1 for $q = 0.001$, the region allowing such transition is rather small. This property is best understood by considering the nullclines for various values of β and $\alpha = p/q$. Since the nullcline x depends only on the ratio $\alpha = p/q$ and the *S*- shaped nullcline y only on parameter β, the number and the location of the steady states also depend only on these two parameters. However, the very nature of the steady states can be affected by the choice of specific values for parameters p and q.

The mappings in Figure 1 for $q = 0.001$ and the subsequent conclusions do not necessarily apply when the parameter q is allowed to vary, even if the ratio $\alpha = p/q$ is maintained fixed. Indeed the region of sustained oscillations cannot be defined only in terms of the ratio $\alpha = p/q$. However, using the methods outlined in this paper, it is possible to map the domain of sustained oscillations for any value of q, with similar results for small values.

The feedback circuits technique used in this paper is easy to implement and can quickly demarcate the parameter space into interesting (oscillations, multistationarity) and noninteresting (single stable fixed point) regions. Hence it could prove useful in surveying dynamical response of models simulating physical, chemical, and biological systems. Recently, in refs 13 and 14, three- and four-dimensional systems have been analyzed and shown to exhibit complex periodic dynamics or even chaotic behavior. In preparation is the implementation of this nullcline-feedback circuit based method to predict the chaotic dynamics exhibited by a three-dimensional electrochemical model.

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