PHYSICAL REVIEW E **67**, 045202(R) (2003)

Tracking fixed-point dynamics in an electrochemical system using delayed-feedback control

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(Received 14 October 2002; published 15 April 2003)

We report numerical and experimental results indicating successful tracking of stabilized fixed points solutions in an electrochemical system. By applying a continuous delayed-feedback technique, periodic oscillations are suppressed via stabilization of a steady-state fixed point. Subsequently, using a simple continuation method involving an update term, this stabilized fixed point is tracked through the bifurcation diagram as a system parameter is slowly varied. Under the influence of this tracking protocol, inception of oscillatory dynamics is precluded over large parameter domains and through bifurcations.

DOI: 10.1103/PhysRevE.67.045202 PACS number(s): 05.45.−a, 87.10.+e

Endeavors involving tracking of unstable solutions have emphasized on tracking periodic solutions rather than fixed point solutions. Tracking of stabilized periodic solutions was first proposed by Carroll *et al.* [1]. Using a composite of the Ott-Grebogi-Yorke (OGY) [2] chaos control algorithm and a continuation method, they were able to track a stabilized periodic orbit in a Duffing-like electric circuit. This work was followed by experimental reports involving tracking of stabilized periodic solutions in a chemical Belousov-Zhabotinsky (BZ) reaction [3] and a laser system [4]. In parallel, there had been reports [5–8] of targeting the system dynamics to nonoscillatory solutions using feedback techniques. Controlling the dynamics on a previously unstable fixed point could be of practical relevance in experimental situations where chaotic and/or periodic oscillations are potentially harmful and cause degradation in performance. In such cases, it would be desirable to suppress the emergence of oscillatory dynamics for a wide parameter range of operating conditions. This being the motivation for the present work, in this paper, we report the tracking of stabilized fixed point responses in a numerical model for electrochemical corrosion [9,10] and in an experimental electrochemical cell [11] using a delayed-feedback control strategy [12,13]. In both simulations and experiments, periodic dynamics were converted to a nonoscillatory (fixed point) state, and this stabilized state was subsequently tracked through the bifurcation diagram using an additional update term. Under the superimposed control, inception of the oscillatory behavior is precluded for a range of operating conditions, where normally periodic or chaotic behavior would be observed.

We consider an autonomous dynamical system that is described by a general set of differential equations

$$\dot{\mathbf{x}} = \mathbf{f}(\mathbf{x}(t), \mathbf{p}), \tag{1}$$

where $\mathbf{x}=(x_1, x_2, x_3, \ldots, x_n)$ and $\mathbf{p}=(p_1, p_2, p_3, \ldots, p_m)$ are the system variables and the control (bifurcation) parameters, respectively. Depending on the values of control parameters, a given system may exhibit a number of dynamical responses such as fixed point steady state(s), periodic or chaotic oscillations, etc. Considering that in our electrochemical experiments, there is an obvious measurable system variable x_1 (e.g., the anodic current I), the accessible control parameter p_1 (e.g., the anodic potential V) can be continuously perturbed such that

$$p_1(t) = p_1(0) + \gamma[x_1(t) - x_1(t-\tau)], \tag{2}$$

where $p_1(0)$ is the initial value of the parameter and the term $\gamma[x_1(t)-x_1(t-\tau)]$ gives the superimposed delayed feedback. It is important to note that this feedback changes the stability of the target dynamics without altering the location of fixed points and/or creating new periodic orbits in the phase space [5,13].

Implementation of the continuous delayed-feedback strategy can be categorized into the following two limiting cases.

(1) In the limit $\tau \to 0$, the delayed-feedback technique effectively reduces to the derivative control [5] which is also capable of stabilizing steady-state solutions (fixed points) [6].

(2) In the limit $\tau \to \tau_{\mathrm{UPO}}$, where τ_{UPO} is the period of an unstable periodic orbit (UPO) embedded in the chaotic attractor, the strategy is equivalent to the Pyragas method of controlling chaos [12].

The delayed-feedback control is first tested on a model for electrochemical corrosion [9] described by three dimensionless differential equations:

$$\dot{Y} = p(1 - \theta_{\mathrm{OH}} - \theta_{\mathrm{O}}) - qY, \tag{3}$$

$$\dot{\theta}_{\mathrm{OH}} = Y(1 - \theta_{\mathrm{OH}} - \theta_{\mathrm{O}}) - [\exp(-\beta\theta_{\mathrm{OH}}) + r]\theta_{\mathrm{OH}} + 2s\theta_{\mathrm{O}}(1 - \theta_{\mathrm{OH}} - \theta_{\mathrm{O}}), \tag{4}$$

$$\dot{\theta}_{\mathrm{O}} = r\theta_{\mathrm{OH}} - s\theta_{\mathrm{O}}(1 - \theta_{\mathrm{OH}} - \theta_{\mathrm{O}}). \tag{5}$$

Variables θ_{O} and θ_{OH} represent the fractions of the electrode surface covered by two different chemical species, while Y represents the concentration of metal ions in the electrolyte. Parameters p, q, r, s, and β are determined by chemical reaction rates in the model [9]. We numerically integrate these equations using a fourth-order Runge-Kutta algorithm with a fixed step size ($h=1.0$). The system parameter p is chosen to be the control parameter whereas s is the bifurcation parameter. Figure 1 shows the numerically generated bifurcation diagram for the model system. It is computed by plotting the

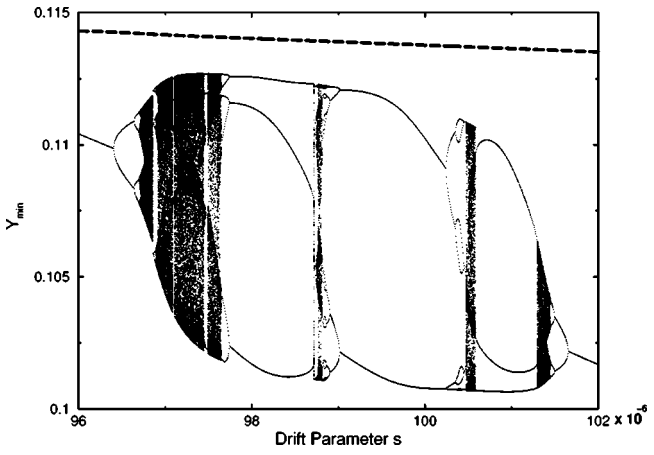

FIG. 1. The numerically computed bifurcation diagram of the three-dimensional electrochemical model [Eqs. (3)–(5)]. While the bifurcation parameter s is varied, the other system parameters p, q, r, and β are fixed at 2.0×10^{-4}, 1.0×10^{-3}, 2.0×10^{-5},and 5.0, repsectively. The superimposed dashed line denotes the tracked fixed point solution. The control parameters of Eq. (6) are $\gamma=-0.1$ and $\tau=24$ integration steps, and the continuation parameter of Eq. (9) is chosen as $\gamma_1=-0.06$.

minima of the Y variable of the model system as a function of s. It shows the typical period doubling route to chaos as the parameter s is increased. After passing through the large periodic windows, an inverse period doubling is observed until the period-1 dynamics is stable again.

Both periodic and chaotic responses can be converted to a steady-state behavior by continuously varying parameter p according to the following control formula:

$$p(t)=p(0)+\gamma[\theta_{\mathrm{OH}}(t)-\theta_{\mathrm{OH}}(t-\tau)]. \tag{6}$$

The appropriate values of γ and τ can determined by trial and error. Using the control of Eq. (6), one can stabilize the previously unstable fixed point. To calculate the update (continuation) term for tracking, fixed points (F_{s_1},F_{s_2}) are stabilized for two extreme values of the bifurcation parameter s, i.e., $s_1=96\times10^{-6}$ and $s_2=102\times10^{-6}$. Subsequently, we define a new scalar g [10],

$$g=(F_{s_2}-F_{s_1})/(s_2-s_1). \tag{7}$$

This enables us to estimate the fixed point location $[F_{fn}(\mathrm{approx})]$ at the new value of s,

$$F_{fn}(\mathrm{approx})=F_{fo}+g\,ds, \tag{8}$$

where F_{fo} is the old fixed point and ds is the change in the drift parameter. The continuation (update) term is given by

$$\gamma_1[\theta_{\mathrm{OH}}-F_{fn}(\mathrm{approx})], \tag{9}$$

when used in conjunction with the control term yields the following tracking protocol:

$$p(t)=p(0)+\gamma[\theta_{\mathrm{OH}}(t)-\theta_{\mathrm{OH}}(t-\tau)] + \gamma_1[\theta_{\mathrm{OH}}-F_{fn}(\mathrm{approx})] \tag{10}$$

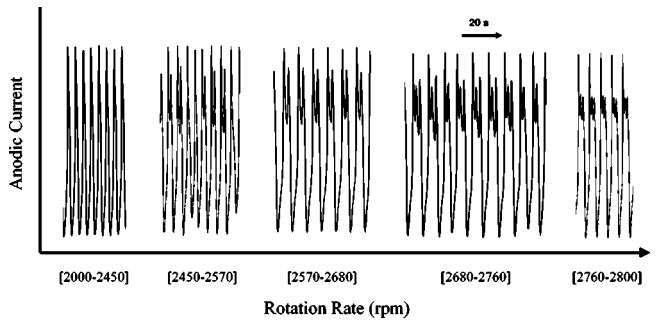

FIG. 2. A schematic of the experimental bifurcation diagram observed as the bifurcation parameter (rotation rate) is varied. It shows alternate windows of periodic and chaotic dynamics. The units of the anodic current I are in milliamperes.

that enables tracking via a continuous update of the fixed point $F_{fn}(\mathrm{approx})$. Compared to the tracking of the periodic solutions [1,3,4], where an update of the control term is required, tracking of fixed point solution is achieved by introducing the continuation term of Eq. (9). The control parameters of the delayed-feedback control, γ and τ are not updated and hence remain unchanged throughout the tracking procedure. A stabilized fixed point that is tracked through the bifurcation diagram is shown superimposed by the dashed line in Fig. 1.

The experimental system is an EG&G Princeton applied research model K60066 three-electrode electrochemical cell setup to study the potentiostatic electrodissolution of copper in an acetate buffer [11]. The anode is a rotating copper disk (5 mm diameter) shrouded by teflon. The electrolyte is an acetate buffer, a mixture of 70 cm^3 glacial acetic acid and 30 cm^3 of 2 $\mathrm{mol\,dm}^{-3}$ sodium acetate. The anodic potential is measured relative to a saturated calomel reference electrode, while the cathode is a platinum foil disk (2.5 cm^2 area). Under potentiostatic conditions, the circuit potential is continuously adjusted by a potentiostat (EG&G princeton applied research model 362) to maintain a desired set value of the anodic potential V, and the anodic current I is measured between the anode and cathode. Time series current data are collected and stored in a computer by sampling the anodic current using a data acquisition card with the sampling frequency fixed at 25 Hz.

Figure 2 shows the schematic of a typical experimental bifurcation diagram for the anodic current I. It shows adjacent periodic windows separated by episodes of chaotic behavior as the rotation rate of the anode is varied. This behavior, common in chemical systems, is called mixed-mode oscillations. In our experiments, it is nearly impossible to construct a continuous bifurcation diagram such as shown in Fig. 1 in real time because of the problems involving transient dynamics and hysteresis. Moreover, since the operating conditions change continuously, the details of the experimental bifurcation diagram are not reproducible. However the, qualitative features remain preserved.

At the anodic potential and rotation rate given in Fig. 3, the electrochemical system exhibits period-1 current oscillations. The unstable fixed point is stabilized (control is turned on at $t=135$ s) by continuously perturbing the anodic poten-

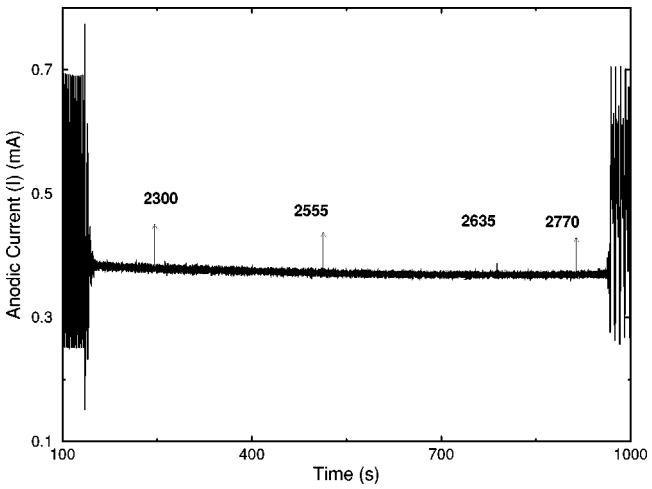

FIG. 3. The tracked fixed point in the experimental electrochemical system. The initial control of period-1 dynamics (rotation rate=2200 rpm) and anodic voltage $V(0)$ is 0.670 V. The control is achieved by using the delayed-feedback control strategy [Eq. (6)] with the following control parameters, $\gamma=-0.23$ and $\tau=65$ sampling steps. Subsequent tracking of the fixed point solution is attained using an update term in conjunction with the initial control term. The continuation parameter of Eq. (9) used is $\gamma_1=-0.07$. Four instantaneous values of the rotation rate; 2300 rpm, 2555 rpm, 2635 rpm, and 2770 rpm are shown as the tracking is performed and rotation rate is varied. Finally, the tracking is switched off at rotation rate=2770 rpm and the system starts exhibiting period-3 oscillations.

tial $V(t)$ according to the following control formula:

$$V(t)=V(0)+\gamma[I(t)-I(t-\tau)]. \tag{11}$$

Similar to the simulations, before implementing tracking, control is achieved for two extreme values of the bifurcation parameter (rotation rate) and the scalar g computed. Subsequently, updated values of the fixed points $[F_{fn}(\text{approx})]$ for different values of the bifurcation parameter (rotation rate) are obtained and the continuation term analogous to that of Eq. (9) is calculated. Finally a tracking procedure analogous to that of Eq. (10) is implemented in experiments. Figure 3 shows the successful tracking of the stabilized fixed point as the drift parameter (rotation rate) is varied. Four snapshot values of the rotation rate during the tracking procedure are also shown in Fig. 3. Eventually, the tracking algorithm is finally turned off and the system starts exhibiting period-3 oscillations which is the autonomous (uncontrolled) dynamics of the system for this value of the bifurcation (drift) parameter (rotation rate). Under the influence of this tracking procedure, it is possible to preclude inception of oscillatory dynamics for a large interval of the bifurcation parameter.

The presented numerical and experimental results indicate that delayed-feedback control in conjunction with a simple update method can be successfully applied to stabilize and subsequently track unstable steady-state fixed points in dissipative electrochemical systems. Moreover, these results indicate that it is possible to use tracking in order to preclude the emergence of oscillatory dynamics in systems. This could be of importance in situations where oscillations lead to degradation of performance. The update term [Eq. (9)] uses a simple linear interpolation scheme that approximates the new value of fixed point as the drift parameter (s in simulations and rotation rate in experiments) is varied. Although this is a very simple technique it seems to work well for our model and experiments. It could be harder to achieve successful tracking for certain extreme situations such as (a) systems where the location of the fixed point has a strong nonlinear dependence on the bifurcation parameter and (b) systems with extremely rapid and abrupt variations in operating conditions. However, since in most practical situations, drift is usually associated with a slow continuous change in experimental conditions, our simple tracking method is applicable and hence worthy of consideration.

Note added. Recently, it was brought to our attention that there exists unpublished work [14] similar to ours involving tracking of fixed points. These experiments were carried out in an another chemical system and use a different algorithm to achieve tracking.

ACKNOWLEDGMENTS

This work was supported by CONACyT, Mexico. Technical assistance from Marco Rivera is appreciated.

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