

Stabilization of unstable steady states and periodic orbits in an electrochemical system using delayed-feedback control

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We report numerical and experimental results indicating successful stabilization of unstable steady states and periodic orbits in an electrochemical system. Applying a continuous delayed-feedback technique not only periodic and chaotic oscillations are suppressed via stabilization of steady-state solutions but also the chaotic dynamics can be converted to periodic behavior. In all cases the feedback perturbation vanishes as a target state is attained. [S1063-651X(99)02205-9]

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I. INTRODUCTION

It is well documented [1–9] that dynamical control of chaotic behavior can be achieved by judiciously applying small perturbations to an accessible control parameter. These experiments employed flexible control strategies [10–12] to convert chaotic behavior to periodic responses. There have also been reports [14–18] on targeting the system dynamics to nonoscillatory solutions (for example, to unstable steady states) using feedback techniques [16,17]. Controlling the dynamics such that a previously unstable fixed point is attained could be of great practical importance in experimental situations where chaotic and/or periodic oscillations are potentially harmful and may cause degradation in performance.

In this article, we report the stabilization of unstable steady states and periodic responses in a numerical model for electrochemical corrosion [19] and in an electrochemical cell [20] using a delayed-feedback control strategy [13,21]. In both the numerical model and the experimental system we were able to convert chaotic and periodic oscillations to a steady state, and chaotic oscillations to a periodic response as well. The article is organized as follows. In the following section a brief introduction to the continuous delayed-feedback strategy is provided. Numerical and experimental results are presented in Secs. III and IV, respectively. We conclude in Sec. V along with a remark about future experiments.

II. DELAYED-FEEDBACK CONTROL

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

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

where $\mathbf{x} = (x_1, x_2, x_3, \dots, x_n)$ and $\mathbf{p} = (p_1, p_2, p_3, \dots, p_m)$ are the system variables and the control (bifurcation) parameters, respectively. Depending on the values of control pa-

rameters, a given system may exhibit a wealth of dynamical responses such as steady state(s), periodic or chaotic oscillations, etc. A discontinuous transition between these states is called a bifurcation, and the map showing the location of different dynamical states in the parameter space is called a bifurcation diagram. With conventional methods, however, one can map out the stable dynamical responses only. Stabilization of the system dynamics on previously unstable responses requires a special targeting procedure for which the delayed-feedback control appears to be a viable tool.

Considering that in our electrochemical experiments there is an easily measurable system variable x_1 (e.g., the anodic current), the accessible control parameter p_1 (e.g., the anodic potential) can be continuously perturbed such that

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

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 [16]. Recently Just *et al.* [21] elucidated the mechanism of delayed-feedback control using a linear stability analysis. Although their discussion concentrates on the problem of stabilizing unstable periodic solutions, the results are equally applicable for the stabilization of previously unstable steady states as well.

Earlier implementations of the continuous delayed-feedback strategy can be categorized into two limiting cases.

(1) In the limit $\tau \rightarrow 0$ the delayed-feedback technique effectively reduces to the derivative control [16] which is capable of stabilizing steady-state solutions (fixed points) [14,15].

(2) In the limit $\tau \rightarrow \tau_{\text{UPO}}$, where τ_{UPO} is the period of an unstable periodic orbit (UPO) embedded in the chaotic attractor, the strategy is the so-called Pyragas method of controlling chaos [13].

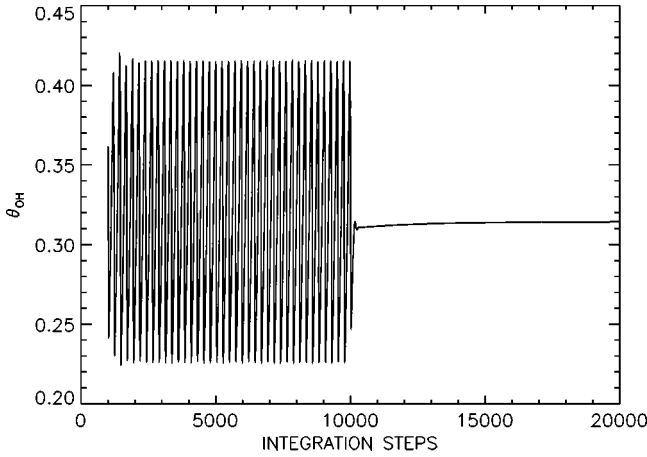


FIG. 1. Stabilization of an unstable steady state in the model Eqs. (3)–(5) exhibiting periodic oscillations. The system parameters p, q, r, s, β are $2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 2.0 \times 10^{-5}, 9.5 \times 10^{-5}, 5.0$, while the control parameters in Eq. (6) are $\gamma = -0.1$ and $\tau = 24$ integration steps.

In an effort to stabilize both unstable steady-state and periodic solutions, our numerical and experimental investigations involve choosing the value of $0 < \tau \leq \tau_{\text{UPO}}$.

III. NUMERICAL RESULTS

The delayed-feedback control is first tested on a model for electrochemical corrosion [19] described by three dimensionless differential equations:

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

$$\begin{aligned} \dot{\theta}_{\text{OH}} = & Y(1 - \theta_{\text{OH}} - \theta_{\text{O}}) - [\exp(-\beta\theta_{\text{OH}}) + r]\theta_{\text{OH}} \\ & + 2s\theta_{\text{O}}(1 - \theta_{\text{OH}} - \theta_{\text{O}}), \end{aligned} \quad (4)$$

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

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. Previous numerical studies have shown that depending on the parameter values this model may result in simple periodic or chaotic oscillations as well [19]. We numerically integrate these equations using a fourth order Runge-Kutta algorithm with a fixed stepsize ($h = 4.0$). For the purposes of control we change the value of p .

At the parameter values given in Fig. 1 the model system exhibits periodic oscillations. Figure 1 also shows that the periodic response can be easily converted to a steady-state behavior by continuously varying parameter p according to the following control formula:

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

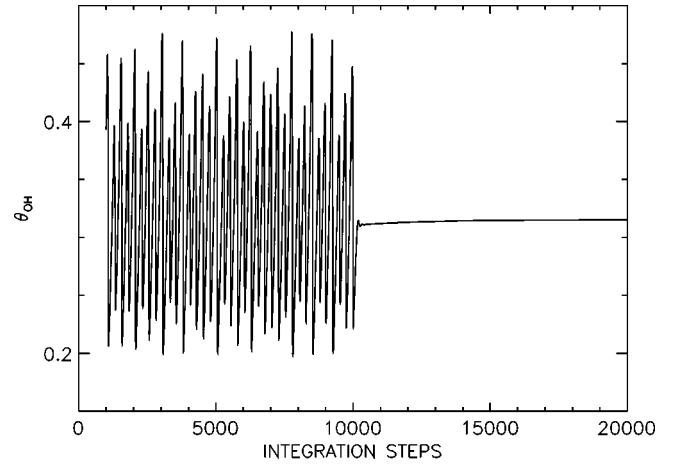


FIG. 2. Stabilization of an unstable steady state in the model Eqs. (3)–(5) exhibiting chaotic oscillations. The system parameters p, q, r, s, β are $2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 2.0 \times 10^{-5}, 9.7 \times 10^{-5}, 5.0$, while the control parameters are $\gamma = -0.11$ and $\tau = 23$ integration steps.

The appropriate values of γ and τ (see figure legend) were determined by trial and error. The same control formula but with slightly different γ and τ values can be successfully applied to convert chaotic oscillations to a steady-state response (Fig. 2). Note that in both cases upon reaching the target state the perturbation vanishes as $\theta_{\text{OH}}(t) = \theta_{\text{OH}}(t - \tau)$. This important feature of the delayed-feedback control warrants that no new steady-state solution is being created but only the stability of the originally unstable fixed point is reversed. If the system under control were moved away from the fixed point, for example, by temporarily changing the value of another control parameter or one of the system variables, the effect of such perturbations would decay due to the delayed-feedback control on parameter p . Once the control is turned off, however, the steady-state solution becomes unstable again. Thus (in the absence of control) the smallest perturbation would move the system away from the fixed point to finally settling on the original periodic orbit (not shown in the figures).

Figure 3 shows the conversion of chaotic oscillations to period-one dynamics using the Pyragas method. The value of $\tau = \tau_{\text{UPO}}$ in the control formula [Eq. (6)] was calculated by utilizing a return map in the following manner. Instead of plotting consecutive extrema of one of the system variables, a return map is created by plotting the number of iteration steps N encountered between successive minima of the same system variable. Hence the constructed map is, in fact, a temporal return map N_{n+1} vs N_n . Using the intersection of this map with the line of identity enables one to approximate the value of τ_{UPO} for the period-one orbit. As the delay time in Eq. (6) now agrees with τ_{UPO} , the feedback signal naturally vanishes upon approaching the stabilized periodic orbit. Note that the delay times applied in Figs. 1 and 2 are an order of magnitude lower than the natural period of the UPO embedded in the chaotic attractor. The presented numerical results indicate that by appropriately choosing the value of time delay between 0 and τ_{UPO} , the delayed-feedback algorithm may work in the experimental setting as well, resulting in the stabilization of different unstable dynamical states (steady states or periodic responses) by choice.

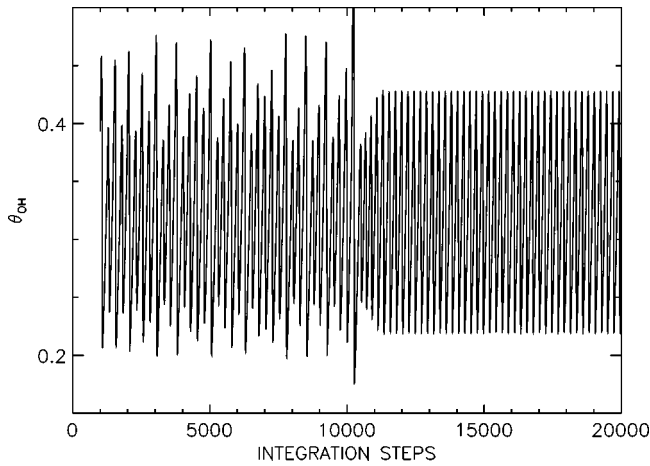


FIG. 3. Controlling chaos in the model Eqs. (3)–(5) using the Pyragas method. The system parameters are the same as in Fig. 2, while the control parameters are $\gamma = -0.11$ and $\tau = \tau_{\text{UPO}} = 210$ integration steps.

IV. EXPERIMENTAL RESULTS

The experimental system was an EG&G Princeton Applied Research Model K60066 three-electrode electrochemical cell set up to study the potentiostatic electrodisso- lution of copper in an acetate buffer [20]. The anode is a rotating copper disk (5 mm diameter) shrouded by Teflon. The electrolyte is an acetate buffer, a mixture of 70 cm³ glacial acetic acid and 30 cm³ of 2 mol dm⁻³ sodium acetate. The anodic potential is measured relative to a saturated calomel reference electrode (SCE), while the cathode is a platinum foil disk (2.5 cm² 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.

At the anodic potential and rotation rate given in Fig. 4(a) the electrochemical system exhibits periodic current oscillations. The unstable fixed point is being stabilized (control is

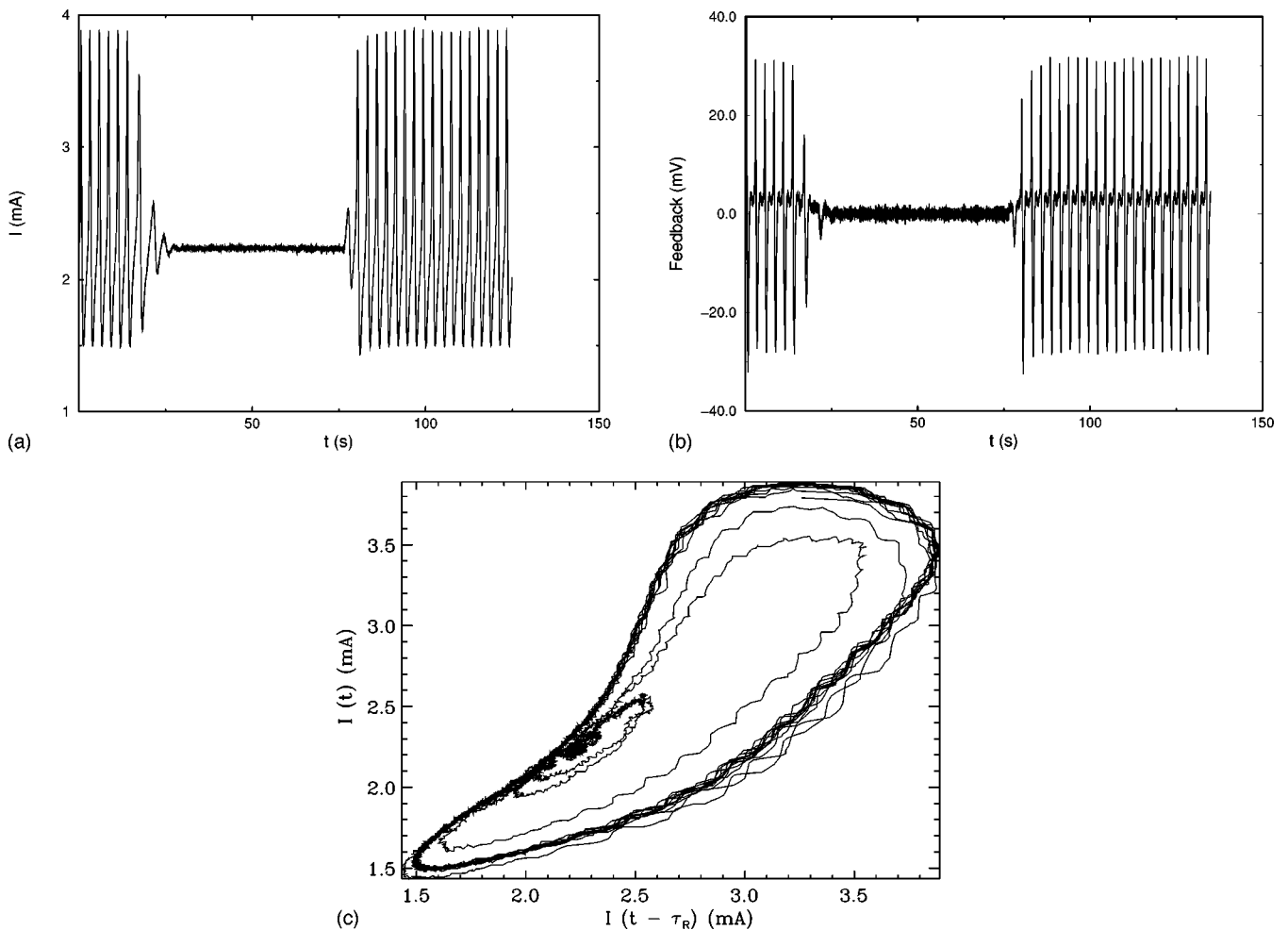


FIG. 4. Stabilization of an unstable steady state in the electrochemical system exhibiting periodic oscillations. The rotation rate is 2400 rpm, while the anodic potential $V(0)$ is 0.670 V. The control parameters used in Eq. (7) are $\gamma = -0.15$ mV/mA and $\tau = 20$ sampling steps. (a) Anodic current plotted over a period during which the delayed-feedback control is switched off, on, and off again. (b) Calculated feedback signal plotted over the same period but applied during the control session only. (c) Phase-space reconstruction for a segment of time series current data from Fig. 4(a). The value of τ_R used for reconstruction is the period of 30 sampling steps.

turned on at $t=20$ sec) by continuously perturbing the anodic potential $V(t)$ according to the following control formula:

$$V(t) = V(0) + \gamma[I(t) - I(t - \tau)]. \quad (7)$$

The applied values of γ and τ are given in the figure legend. As a result of the delayed-feedback on the anodic potential, the current oscillations decay to eventually yielding a dc response from the system. Though the current signal is a bit noisy, we may say that the previously unstable steady state has been successfully stabilized. When the control is turned off at $t=75$ sec, the system moves away from the fixed point and reverts to executing periodic oscillations again. Values of the feedback signal are plotted in Fig. 4(b). Note that the values are shown over the whole time scale of the experiment but the feedback signals have been imposed during the control session (between 20 and 75 sec) only. The applied corrections to the potential are max $\pm 5\%$ of the rest potential $V(0)$, and they basically go to zero as the system stabilizes itself on the fixed point. Figure 4(c) depicts the dynamics in a reconstructed phase space for a segment of the time series data in Fig. 4(a). When the control is turned on the system leaves the limit cycle, spirals in, and eventually approaches the previously unstable fixed point inside the periodic orbit.

The same control formula [Eq. (7)] but with slightly different γ and τ values can be applied to convert chaotic current oscillations to a steady-state response as well. At the anodic potential and rotation rate given in Fig. 5(a) the electrochemical system exhibits chaotic current oscillations. During control (between 40 and 140 sec), the chaotic oscillations decay yielding a noisy dc response. When control is turned off the system moves away from the fixed point and reverts to executing chaotic oscillations again. Values of the applied feedback signals are plotted in Fig. 5(b). That the control is less successful than in Fig. 4(a) is clearly reflected by the oscillatory character of the feedback signal upon attainment of the target steady state as shown in Fig. 5(b). On the other hand, both Figs. 5(a) and 5(b) prove that by the application of the feedback perturbation the stability of the steady state is, indeed, being reversed.

Implementation of the Pyragas method to stabilize the UPO embedded in the chaotic attractor involved a precontrol procedure to determine τ_{UPO} similar to that described in Sec. III. A temporal return map has been constructed by plotting the time elapsed (sampling time \times number of samples taken) between successive minima of the anodic current, and the intersection of this map with the line of identity gave us an approximate value for τ_{UPO} . Figure 6(a) shows time series current data while control is off, turned on (at $t=70$ sec), and then shut off again. During the control session, the chaotic current oscillations are tamed (within the experimental error) to a periodic response. When the control is turned off (at $t=135$ sec) the system moves away from the periodic orbit and reverts to executing chaotic oscillations again. The occasional spiking in the applied feedback signal shown in Fig. 6(b) may originate from the imperfect targeting of the periodic orbit due to several reasons: Experimental errors in measuring the current, a limit of variation in the anodic po-

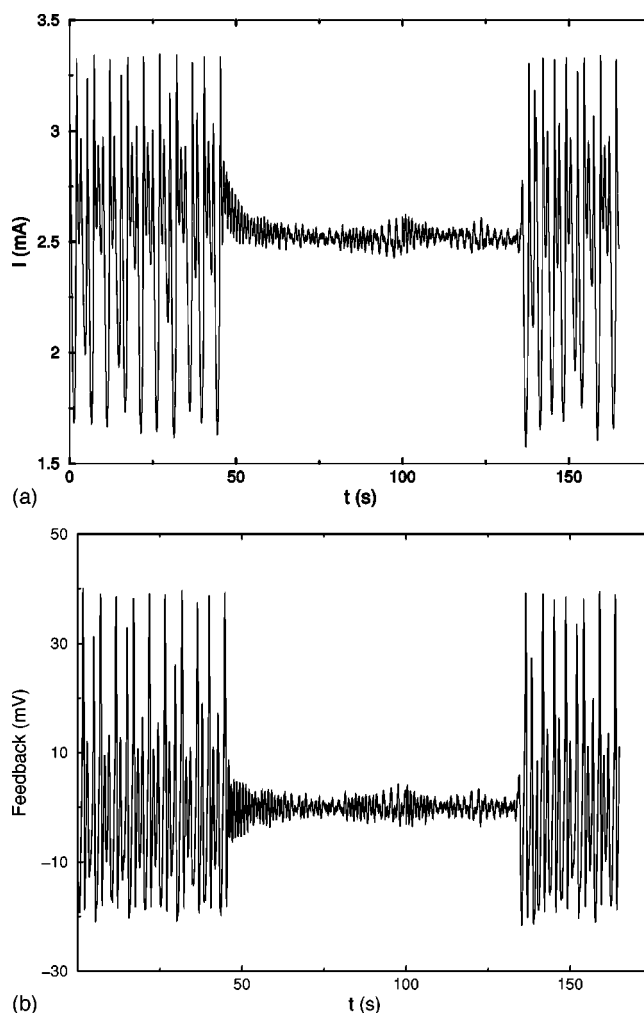


FIG. 5. Stabilization of an unstable steady state in the electrochemical system exhibiting chaotic oscillations. The rotation rate is 2700 rpm, the anodic potential $V(0)$ is 0.720 V. The control parameters used in Eq. (7) are $\gamma = -0.15$ mV/mA and $\tau = 15$ sampling steps. (a) Anodic current plotted over a period during which the control is switched off, on, and off again. (b) Calculated feedback signal plotted over the same period but applied during the control session only.

tential by the potentiostat (± 0.001 mV) and the error in determining τ_{UPO} . Figure 6(c) depicts the dynamics in the reconstructed phase space for a segment of the time series in Fig. 6(a). As the control is turned on, the system trajectories depart from the chaotic attractor and settle down on the previously unstable limit cycle. This figure clearly shows that the UPO is, indeed, embedded in the chaotic attractor and only its stability is being changed by the introduction of the feedback perturbation.

V. CONCLUSIONS

The presented numerical and experimental results indicate that delayed feedback control can be successfully applied to stabilize unstable steady states and periodic orbits in dissipative electrochemical systems. The great advantage of the applied self-controlling feedback is that it renders unification to two distinct control strategies, namely, the derivative control strategy and the Pyragas method for controlling chaos. By

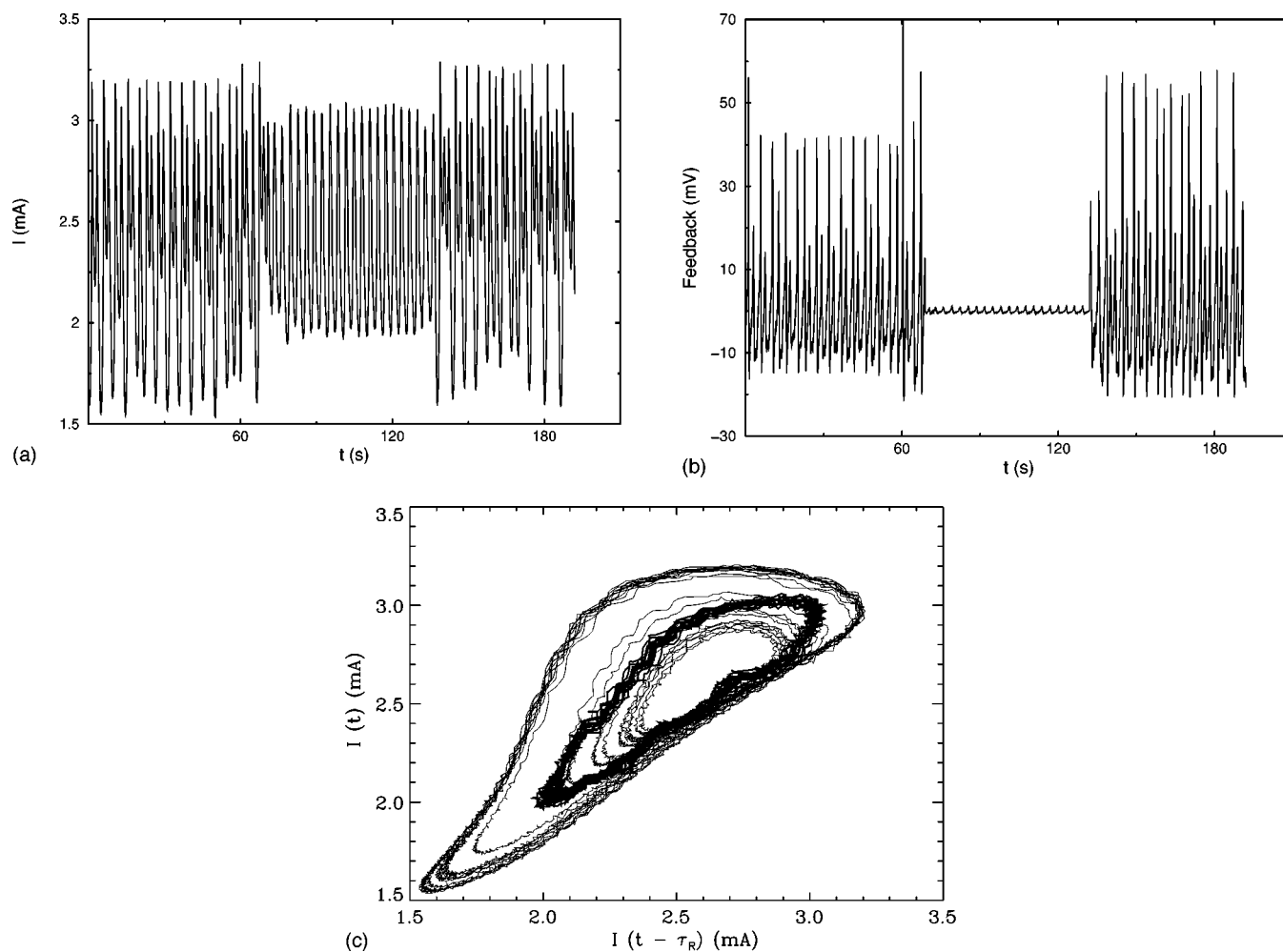


FIG. 6. Controlling chaos in the electrochemical system using the Pyragas method. The rotation rate is 2900 rpm, while the anodic potential $V(0)$ is 0.715 V. The control parameters used in Eq. (7) are $\gamma = -0.19$ mV/mA and $\tau = 65$ sampling steps. (a) Anodic current plotted over a period time during which the control is switched off, on, and off again. (b) Calculated feedback signal plotted over the same period but applied during the control session only. (c) Phase-space reconstruction for a segment of time series data from Fig. 6(a). The value of τ_R is the same as in Fig. 4(c).

judicious variations of the two available control parameters γ and τ , stabilization of unstable steady states and periodic orbits can be attained by choice. Our results also prove that the feedback perturbations do not change the projections of the unstable steady states and UPOs in the original phase space, and the additional degree of freedom due to the perturbation changes only the stability of these objects. Moreover, since the control signal vanishes subsequent to attainment of the target state, it is ensured that the system has not drifted to a regime where the target dynamics were naturally stable. Initial results for the implementation of this control strategy to tracking of fixed points and periodic orbits in

slowly drifting systems [22] and for mapping out otherwise unattainable unstable dynamical states in the bifurcation diagram of other electrochemical systems as well are encouraging.

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