Synchronization of Chemical Systems Using External Forcing

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We report the synchronization of dynamics in a numerical model simulating electrochemical corrosion using external chaotic forcing, periodic forcing, and finally forcing including a random component (random forcing). For all three external forcings synchronization is achieved when the two response systems are at identical parameter conditions exhibiting similar behavior. However when the two response systems are at unequal parameter values exhibiting different dynamical behavior, synchronization is achieved only for forcing including a random variable (random forcing). This ability of random perturbations to achieve generalized synchronization makes it a candidate worthy of consideration in problems involving synchronization of nonidentical systems.

I. Introduction

Interest in synchronization of dynamics has increased because of its possible relevance to secure communications. It started out with efforts to synchronize identical systems,^{1–5} but lately much emphasis has been placed on synchronization of nonidentical systems^{6,7} (generalized synchronization).

Recently it was realized that chaotic dynamics could be tamed using external perturbations.8 In this article we extend this already existing idea⁸ and propose using external (identical) signals superimposed onto the dynamical equations of the two response systems to achieve synchronization.⁹ The problem considered here is different from the usual scenario where the goal is to synchronize the dynamics of the drive and the response system. In the case discussed here the drive system is merely a generator of the chaotic, periodic, or random signals which are superimposed on the dynamics of the two response systems. Since there is an absence of a target (synchronized) state for the two response systems, the final synchronized dynamics tend to be different from both the initial unsynchronized dynamics (of response systems) and the superimposed external dynamics of the drive system. The article is organized as follows: In the following section a brief introduction to the model system chosen for superposition of external signals is provided. In section III results from application of the periodic forcing are discussed. Numerical results from the application of external chaotic signals are presented in section IV. Finally, in section V results from the application of random driving are presented along with a brief comparison.

II. Model for Electrochemical Corrosion

The chemical system under consideration involves passivation of the reactive surface of a metal electrode in an electrochemical cell. The chemical kinetics of the passivation model includes the formation of two surface films, MOH (θ) and MO (θ_0), where M represents the metal atom. It combines elements from surface reaction models by Talbot and Oriani¹¹ for MOH and by Sato¹² for MO formation. The chemical kinetics leads to the dimensionless equations¹³

$$\dot{Y} = p(1 - \theta - \theta_{\rm O}) - qY \tag{1}$$

$$\dot{\theta} = Y(1 - \theta - \theta_{\rm O}) - [\exp(-\beta\theta) + r]\theta + 2s\theta_{\rm O}(1 - \theta - \theta_{\rm O})$$
(2)

$$\dot{\theta}_{\rm O} = r\theta - s\theta_{\rm O}(1 - \theta - \theta_{\rm O}) \tag{3}$$

where Y is the concentration of metal ions in the electrolyte. θ and θ_0 are the respective fraction of the metal surface covered by each film, p, q, r, and s are parameters related to chemical rate constants, and β represents the non-Langmuir nature of MOH film formation in the Talbot-Oriani model. The system has been studied in some detail 13 and is chaotic for parameter values $(p, q, r, s, \beta) = (2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 2.0 \times 10^{-5},$ 9.7×10^{-5} , 5.0). The discussion below deals with the behavior of the system in the neighborhood of this point in parameter space. In our numerical experiments two copies of this system are created, and the appropriate driving (periodic, chaotic, or random) is superimposed onto the evolution equation of one of the three independent variables (Y, θ, θ_0) . All the numerical calculations for this model system were performed using the fourth-order Runge-Kutta algorithm, with a step size of h =0.1.

III. Numerical Results for External Periodic Forcing

The external periodic signal is superimposed onto the dynamics of the two identical chemical systems exhibiting chaotic dynamics (eqs 1–3) at the parameter values (p, q, r, s, β) (2.0 × 10⁻⁴, 1.0 × 10⁻³, 2.0 × 10⁻⁵, 9.7 × 10⁻⁵, 5.0). The superimposed external periodic signal is chosen to be the periodic time series of a third chemical system but operating at a different region in parameter space ((p, q, r, s, β) = (2.0 × 10⁻⁴, 1.0 × 10⁻³, 0.0, 0.0, 5.0)). At this point in parameter space the system is two-dimensional and the dynamical response is a limit cycle.¹³ Under the influence of the external periodic signal (superimposed on the evolution equation) the altered



Figure 1. Synchronization of dynamics for the two response systems operating at identical parameter conditions using an external periodic forcing (time series of Y(3) from the 2-D model). The value of γ used to achieve synchronization is $\gamma = 0.004$.

equations of motions for the two response systems are

$$\dot{Y}(1) = p(1 - \theta(1) - \theta_0(1)) - qY(1) + \gamma(Y(1) - Y(3))$$
(4)

 $\dot{\theta}(1) = Y(1)(1 - \theta(1) - \theta_{\rm O}(1)) - [\exp(-\beta\theta(1)) + r]\theta(1) + 2s\theta_{\rm O}(1)(1 - \theta(1) - \theta_{\rm O}(1))$ (5)

$$\dot{\theta}_{0}(1) = r\theta(1) - s\theta_{0}(1)(1 - \theta(1) - \theta_{0}(1))$$
 (6)

and

$$\dot{Y}(2) = p(1 - \theta(2) - \theta_0(2)) - qY(2) + \gamma(Y(2) - Y(3))$$
(7)

$$\dot{\theta}(2) = Y(2)(1 - \theta(2) - \theta_0(2)) - [\exp(-\beta\theta(2)) + r]\theta(2) + 2s\theta_0(2)(1 - \theta(2) - \theta_0(2))$$
(8)

$$\dot{\theta}_{0}(2) = r\theta(2) - s\theta_{0}(2)(1 - \theta(2) - \theta_{0}(2))$$
(9)

The external driving (*Y*(3)) superimposed onto the evolution equations (eqs 4 and 7) of the two response system is $\gamma(Y(1) - Y(3))$ (eq 4) and $\gamma(Y(2) - Y(3))$ (eq 7), respectively. Figure 1 shows the difference of the θ variable of the two response systems (($\theta(1) - \theta(2)$) converging to zero upon initiation of the forcing at the 20 000th step of integration. The minimum value of γ able to achieve synchronization was $\gamma = 0.004$.

As the superimposed periodic driving to the two systems is nonvanishing, the synchronized final dynamics are different from the initially unsynchronized chaotic dynamics of the two response systems. In fact the final synchronized state of the two response systems is periodic because of the entrainment of the dynamics under the influence of the periodic signal (via superposition). Hence the two response (initially unsynchronized) systems attain synchronization subsequent to the initiation of the external periodic driving, and the final synchronized state is periodic. Figure 2 shows the final periodic attractor (identical for the two response systems) corresponding to synchronized dynamics.

Synchronization is also achieved if the periodic time series of the *Y* variable are replaced with the periodic time series of the other independent variable (θ) .

Efforts to synchronize the two response systems operating at different parameter conditions exhibiting different dynamical



Figure 2. Two-dimensional projection of the final periodic attractor for the two response systems after the initiation of the synchronizing periodic forcing ($\gamma = 0.004$).



Figure 3. Synchronization of dynamics for the two response systems operating at identical parameter conditions using an external chaotic forcing (time series of Y(3)). The value of γ used to achieve synchronization is $\gamma = 0.003$.

behavior (generalized synchronization) were unsuccessful using the external periodic driving.

IV. Numerical Results for External Chaotic Forcing

The two response systems (eqs 4–9) are at parameter values (similar to previous section) such that they exhibit chaotic dynamics. The superimposed external forcing is chosen to be the chaotic time series from a third copy of the same chemical system but operating at a different region in parameter space ($(p, q, r, s, \beta) = (2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 2.0 \times 10^{-5}, 9.685 \times 10^{-5}, 5.0)$) (different s). The external driving (Y(3)) superimposed onto the evolution equations (eq 4 and eq 7) of the two response system is $\gamma(Y(1) - Y(3))$ (eq 4) and $\gamma(Y(2) - Y(3))$ (eq 7), respectively. Figure 3 shows the difference of the θ variable for the two response systems ($\theta(1) - \theta(2)$) converging to zero upon initiation of the driving (at the 20 000th step of integration). The minimum value of γ enable to attain synchronization was $\gamma = 0.003$.

As the superimposed chaotic driving (Y(3)) to the two response systems is nonvanishing, the synchronized final dynamics (complex nonchaotic) are different from the initially unsynchronized chaotic dynamics (for the response systems) and from the chaotic dynamics of the drive system. The nonchaotic nature of the final attractor (chaos suppression) is





Figure 4. Two-dimensional projection of the attractor for the two response systems before and after the initiation of the synchronizing chaotic forcing ($\gamma = 0.003$). (a) The two-dimensional projection of the chaotic attractor corresponding to unsynchronized dynamics. (b) The two-dimensional projection of the final nonchaotic attractor corresponding to synchronized dynamics.

verified by calculating the Lyapunov exponents. However, for small values of γ , the final synchronized attractor (identical for both the response systems) retains some features of the initial chaotic attractor corresponding to unsynchronized dynamics (similar for both the response systems). Figure 4a shows the two-dimensional projection of the chaotic attractor corresponding to unsynchronized dynamics. Figure 4b displays the twodimensional nonchaotic attractor for the two response systems (synchronized state) for $\gamma = 0.003$. It preserves some initial features of the unsynchronized chaotic attractor of Figure 4a. However, as the value of γ is increased, the final synchronized dynamics depart from the initial unsynchronized state manifested by the destruction of any correlation between the two (before and after) attractors.

Similar to periodic forcing, synchronization was also achieved when instead of using a chaotic time series of *Y* variable one uses the chaotic time series of the other two independent variables (θ , θ_0). Figure 5 shows the attainment of synchronization using driving of the form $\gamma(Y(1) - \theta_0(3))$ (added to eq 4) and $\gamma(Y(2) - \theta_0(3))$ (added to eq 7) superimposed to the dynamics of the two response systems.

Finally similar to results for periodic forcing, efforts to synchronize the two response systems operating at different parameter conditions exhibiting different dynamical behavior (generalized synchronization) were unsuccessful using the external chaotic driving.



Figure 5. Synchronization of dynamics for the two response systems operating at identical parameter conditions using an external chaotic forcing (time series of θ_0). The value of γ used to achieve synchronization is $\gamma = 0.003$.



Figure 6. Synchronization of dynamics for the two response systems operating at identical parameter conditions using an external random forcing (Y - R). The value of γ used to achieve synchronization is $\gamma = 0.0001$.

V. Numerical Results for External Random Forcing

The two response systems (eqs 4-9) again are exhibiting chaotic dynamics (parameter similar to section III). The random component (R) of the superimposed external forcing comes from the random number generator of the machine $(-1 \le R \le 1)$. The external driving (superimposed every time step) onto the evolution equations (eq 4 and eq 7) of the two response systems is $\gamma(Y(1) - R)$ (eq 4) and $\gamma(Y(2) - R)$ (eq 7). Figure 6 shows the difference of the θ variable for the two response systems $(\theta(1) - \theta(2))$ converging to zero upon initiation of the driving at the 20 000th step of integration. The minimum value of γ required to attain synchronization was $\gamma = 0.0001$, an order of magnitude lower than that required for external periodic and chaotic drivings. Similar to the previous two cases the final attractor departs (in appearance) from the initial chaotic dynamics proportional to the values of γ used to achieve synchronization. It was also verified that the final dynamics were different from the superimposed random forcing so as to ensure that the synchronized state was not completely dominated by the random signal.

The added advantage of using the random driving was that it is possible to achieve synchronization of the two response systems even though they operate at different parameter



Figure 7. Synchronization of dynamics for the two response systems operating at different parameter conditions (system 1, (p, q, r, s, β)) $(2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 2.0 \times 10^{-5}, 9.7 \times 10^{-5}, 5.0)$); system 2, $((p, q, r, s, \beta) = (2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 0.0, 0.0, 5.0)$) exhibiting different qualitative behavior using an external random forcing (Y - R). The value of γ used to achieve synchronization is $\gamma = 0.001$. (a) The difference of the variable converging to zero manifesting synchronization. (b) The dramatic change in the dynamics of the response system under the influence of the synchronizing forcing.

conditions and exhibit different dynamics (generalized synchronization) Figure 7a shows the difference of the θ variable of the two dissimilar response systems ($\theta(1) - \theta(2)$) converging to zero upon initiation of the driving at the 20 000th step of integration. Figure 7b exhibits the time series of the response system ($\theta(2)$) chosen to be at a parameter value where the dynamics is a period one oscillation. It clearly manifests the dramatic change of dynamics under the influence of the synchronizing random forcing. Again it was verified that the time series corresponding to the synchronized state was not similar to the external random signal.

Moreover we were able to achieve synchronization if we chose the random forcing superimposed onto the evolution equations (eq 4 and eq 7) of the two response systems to be of the form γR (eq 4) and γR (eq 7), respectively (pure random driving). This is similar to adding a global random forcing to a dynamical system. Figure 8 shows the difference of the θ variable for the two response systems ($\theta(1) - \theta(2)$) converging to zero upon initiation of the driving at the 20 000th step of integration. The final synchronized state was again found to be different from the unsynchronized dynamics and from the external random signal.



Figure 8. Synchronization of dynamics for the two response systems operating at identical parameter conditions using an external random forcing (*R*). The value of γ used to achieve synchronization is $\gamma = 0.0003$.



Figure 9. Synchronization of dynamics for the two response systems operating at different parameter conditions (system 1, (p, q, r, s, β)) $(2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 2.0 \times 10^{-5}, 9.7 \times 10^{-5}, 5.0)$); system 2, $((p, q, r, s, \beta) = (2.0 \times 10^{-4}, 1.0 \times 10^{-3}, 0.0, 0.0, 5.0))$ exhibiting different qualitative behavior using an external random forcing (*R*). The value of γ used to achieve synchronization is $\gamma = 0.0005$.

Finally using the random driving of the form γR (eq 4) and γR (eq 7) we were also able to synchronize the two response systems operating at different parameter conditions, as shown in Figure 9.

In conclusion results of successful synchronization are presented using nonvanishing external periodic chaotic and random forcings. However in comparison (of the three forcings), the superiority of the random forcing is clearly evident, as not only is it able to achieve synchronization for similar response systems but it also is equally efficient in attaining generalized synchronization. The results for the random driving are independent of the manner of implementation of the forcing ($\gamma(Y - R)$ or γR) (in the sense that synchronization is achieved for either driving) and are applicable to actual experimental situations. Moreover the fact that forcing of the form γR is successful indicates a possible relevance of using random forcings to achieve synchronization of spatiotemporal systems. Our initial results using random driving to synchronize spatiotemporal systems are encouraging.

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