Dynamics of the electrochemical reaction behavior under the influence of random perturbations

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Abstract. The paper deals with the effect of random perturbations on critical phenomena in a dynamic model of an electrochemical reaction. Classical theories predict that solutions of differential equations will leave any neighborhood of a stable limit cycle if white noise is added to the system. The effect of external disturbances on the limit cycle is investigated, the sensitivity of the cycle to the noise is found. An analysis of the noise-induced transitions is performed.

1. Introduction
The unstable operation of any industrial unit is accompanied by some losses. The instability of the operating mode of an electrochemical reactor [1–5] leads, in some cases, to its shutdown, reduction of productivity, or, in the worst cases, to the explosion. Therefore, the clarification of the stability conditions can be interpreted as a part of the problem of reliability, operability and even economy of the technological process [6].

The main goal of this work is to study the electrochemical reactor behaviour under the influence of naturally present random perturbations. In a chemical system, the role of random perturbations can be played by various impurities, thermal vibrations and many other external factors. The presence of random perturbations can fundamentally change the behaviour of the system.

2. Dynamic model of electrochemical reactor
In this paper we investigate the Koper-Sluyters electrocatalytic reaction [1] mechanism underlying an electrochemical reactor. The dimensionless dynamic model of the reaction in the case of potentiostatic control is described by the system

\[
\frac{du}{dt} = -k_a e^{\gamma \theta/2} u (1 - \theta) + k_d e^{-\gamma \theta/2} \theta + 1 - u, \tag{1}
\]

\[
\beta \frac{d\theta}{dt} = k_a e^{\gamma \theta/2} u (1 - \theta) - k_d e^{-\gamma \theta/2} \theta - k_e e^{\alpha_0 \zeta} E \theta, \tag{2}
\]

where \(u\) is the dimensionless interfacial concentration of \(X\); \(\theta\) is the dimensionless amount of \(X\) that is adsorbed on the electrode surface; \(E\) is the interfacial potential; \(\beta\) is the coverage ratio of the adsorbate; \(\alpha_0\) is the symmetry factor for the electron transfer; and \(\zeta = F/(RT)\), where \(R\)
is the universal gas constant, $F$ is Faraday’s constant, and $T$ is the temperature. The physical meaning of the parameter $\gamma$ has always been a subject of dispute. In most of the literature [1], it is interpreted as an interaction parameter. Positive $\gamma$ signifies attractive and negative $\gamma$ signifies repulsive adsorbate interactions.

The system (1), (2) is singularly perturbed due to smallness of the parameter $\beta$ [7, 9] that allows us to use the geometric theory of integral manifolds of the singularly perturbed systems of ODE [8,10–13] for the study of the dynamics of the system.

A detailed analysis of the deterministic model was carried out using methods of the theory of singular perturbations and numerical methods. The main system processes were identified in [14–17]. A new type of reaction regime was singled out, under which the modelling trajectory at first move along the stable slow integral manifold and then continue for a while along the unstable slow integral manifold. Such trajectories of singularly perturbed systems are called canards, see for example [18–26] and references therein. These regimes are critical since they play the role of a border between two main types of reaction modes: nonperiodic regime and relaxation oscillations (limit cycle) [27,28].

2.1. Critical regime of the model

In [14] it has been shown that the critical point is a stable focus when it lies on the stable part of the slow curve and it is an unstable focus when it situated on the unstable part. In the second case, the relaxation oscillations are observed in the system.

The transition between these two situations corresponds to the case when the critical point coincides with the jump point, the stable equilibrium of the system becomes unstable, and at the same instant the stable limit cycle is originated, i.e., the Andronov–Hopf bifurcation occurs. With further minor modifications of the control parameter, say $k_e$, the critical point shifts on the unstable part of the slow curve, staying in small (of order $O(\beta)$ as $\beta \to 0$) the neighbourhood of the jump point. As parameter $k_e$ changes further this limit cycle grows, and at a value $k_e = k_e^*$ (so-called canard point) it becomes the canard cycle [29–35] with the following it canard explosion [36–39]. Recall that the trajectories which at first move along the stable slow integral manifold and then continue for a while along the unstable slow integral manifold are called canards [11,40].

From first sight, the threshold in the qualitative behaviour of the solutions of the system corresponds to the Andronov–Hopf bifurcation point. However, when the value of the control parameter is close to the Andronov–Hopf bifurcation point, the size of the limit cycle is so small that the behaviour of the system’s solution is practically indistinguishable from the slow mode. If, in the case of the slow regime, the trajectories approach the stable equilibrium, practically coinciding with the jump point, in the latter case they tend to a small limit cycle, nearly coinciding with the same jump point. And only when the control parameter attains the canard point, provided the equilibrium is on the unstable part of the slow curve, but in the sufficiently small vicinity of the jump point, the qualitative change in the system’s behaviour can be observed. Namely, the growth of the limit cycle occurs in such a way that it becomes possible to speak of the existence of the canard trajectory. In other words, the appreciable change in size and/or in the form of the limit cycle is observed for small variations of the control parameter, i.e. the canard explosion takes place. Thus, the canard point is the critical value of the control parameter.

The canards and the parameter value $k_e^*$ allow asymptotic expansions in powers of the small parameter $\beta$ [11,35,40]:

\begin{equation}
\Phi(\theta, \beta) = \Phi(\theta) + \beta \Phi_1(\theta) + \beta^2 \Phi_2(\theta) + \ldots,
\end{equation}

\begin{equation}
k_e^* = \chi(\beta) = \chi_0 + \beta \chi_1 + \beta^2 \chi_2 + \ldots.
\end{equation}
In order to find these asymptotic expansions for the canard and the canard point, we substitute the formal expansions (3) and (4) into the invariance equation [13]

\[ \frac{du}{d\theta} g(u, \theta) = \beta f(u, \theta). \]  

which follows from the system (1), (2). As a result we have:

\[ u_0(\theta) = \frac{(k_d e^{-\gamma \theta/2} + \chi_0 e^{\alpha \zeta E}) \theta}{k_a e^{\gamma \theta/2} (1 - \theta)}, \]  

\[ u_1(\theta) = -k_a u_0(\theta)(1 - \theta)e^{\gamma \theta/2} + k_d e^{-\gamma \theta/2} + 1 - u_0(\theta) + \chi_1 e^{\alpha \zeta E} \theta u'_0(\theta), \]  

\[ \chi_0 = \frac{k_a (1 - \bar{\theta}) e^{\gamma \bar{\theta}/2} - k_d e^{-\gamma \bar{\theta}/2} \bar{\theta}}{(k_a (1 - \bar{\theta}) e^{\gamma \bar{\theta}/2} - 1) e^{\alpha \zeta E} \bar{\theta}}, \]  

\[ \chi_1 = -\frac{k_a u_1(\bar{\theta})(1 - \bar{\theta}) e^{\gamma \bar{\theta}/2} + u_1(\bar{\theta}) + k_a u_1(\bar{\theta}) u'_1(\bar{\theta})(1 - \bar{\theta}) e^{\gamma \bar{\theta}/2}}{e^{\alpha \zeta E} \bar{\theta} u'_1(\bar{\theta})}, \]  

where the value \( \theta = \bar{\theta} \) corresponding to the jump point.

The equations (6)–(9) define the first-order approximations for the canard passing through the jump point \( (u(\bar{\theta}), \bar{\theta}) \) and the canard point of the system (1), (2).

During the study, we have set aside the problem of the random perturbations influence on the critical regime of the model [41–49]. Since such a regime is modelled by a canard, it is necessary to investigate how the shape, size and possibility of its existence change under influence of the perturbation [50–53].

\[ \frac{du}{dt} = -k_a e^{\gamma \theta/2} u(1 - \theta) + k_d e^{-\gamma \theta/2} + 1 - u + \epsilon w = f(u, \theta), \]  

\[ u(\theta) = \frac{(k_d e^{-\gamma \theta/2} + \chi_0 e^{\alpha \zeta E}) \theta}{k_a e^{\gamma \theta/2} (1 - \theta)}, \]  

\[ u_1(\theta) = -k_a u_0(\theta)(1 - \theta)e^{\gamma \theta/2} + k_d e^{-\gamma \theta/2} + 1 - u_0(\theta) + \chi_1 e^{\alpha \zeta E} \theta u'_0(\theta), \]  

\[ \chi_0 = \frac{k_a (1 - \bar{\theta}) e^{\gamma \bar{\theta}/2} - k_d e^{-\gamma \bar{\theta}/2} \bar{\theta}}{(k_a (1 - \bar{\theta}) e^{\gamma \bar{\theta}/2} - 1) e^{\alpha \zeta E} \bar{\theta}}, \]  

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3. Stochastic model of electrochemical reactor

In this section, a model of an electrochemical reaction of the Koper-Sluyters type with allowance for random perturbations is considered. It is assumed that the system has a white noise of low intensity. In this case, the model can be represented by the following form:

\[ \frac{du}{dt} = -k_a e^{\gamma \theta/2} u(1 - \theta) + k_d e^{-\gamma \theta/2} + 1 - u + \epsilon w = f(u, \theta), \]  

\[ u(\theta) = \frac{(k_d e^{-\gamma \theta/2} + \chi_0 e^{\alpha \zeta E}) \theta}{k_a e^{\gamma \theta/2} (1 - \theta)}, \]  

\[ u_1(\theta) = -k_a u_0(\theta)(1 - \theta)e^{\gamma \theta/2} + k_d e^{-\gamma \theta/2} + 1 - u_0(\theta) + \chi_1 e^{\alpha \zeta E} \theta u'_0(\theta), \]  

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\[ \chi_1 = -\frac{k_a u_1(\bar{\theta})(1 - \bar{\theta}) e^{\gamma \bar{\theta}/2} + u_1(\bar{\theta}) + k_a u_1(\bar{\theta}) u'_1(\bar{\theta})(1 - \bar{\theta}) e^{\gamma \bar{\theta}/2}}{e^{\alpha \zeta E} \bar{\theta} u'_1(\bar{\theta})}, \]  

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where, \( w_1 \) and \( w_2 \) are independent Wiener processes. The case of small noise amplitude is considered.

Without loss of generality the parameters of the system are chosen to be \( \epsilon = 0.2, \gamma = 8.99, k_a = 10, k_d = 100, \alpha_0 = 0.05, f = 38.7, E = 0.207564 \) unless other values are specified in figure captions.

Figures 1–3 display the process without the effect of random perturbations. Figures 4–6 demonstrate the same process with the effect of random perturbations.

- Figure 3: The slow curve (red line) and the trajectory (black line) of the system (1), (2) without the effect of random perturbations.
- Figure 4: Plot of \( u = u(t) \) with the effect of random perturbations. 
- Figure 5: Plot of \( \theta = \theta(t) \) with the effect of random perturbations.

We start with the analysis of the sensitivity of the stochastic equilibrium of a dynamical system depending on the control parameter \( k_e \) to noise.

4. **Theoretical sensitivity to random perturbations**

The stochastic sensitivity function method [54] is applied to analyze the sensitivity of the stochastic equilibrium of a dynamical system to the random perturbations. This method based on the calculation of the stochastic sensitivity matrix \( W \). \( W \) a positively definite symmetric matrix. It characterizes the spread of random trajectories of the system around the equilibrium position. The eigenvalues of the matrix \( W \) are so-called the theoretical characteristics of noise sensitivity.

The matrix \( W \) is found from the solution of the matrix equation [55]:

\[ \frac{\partial W}{\partial k_e} = \frac{\partial g(u, \theta)}{\partial k_e} \]
FW + WF^T + S = 0, \hspace{1cm} (12)

where \( F = \left( \begin{array}{cc} \frac{\partial f}{\partial u} & \frac{\partial f}{\partial \theta} \\ \frac{\partial g}{\partial u} & \frac{\partial g}{\partial \theta} \end{array} \right) \)_{\bar{u}, \bar{\theta}}, \quad S = \left( \begin{array}{cc} 1 & 0 \\ 0 & 1 \end{array} \right)

For the two-dimensional system the matrix \( W \) has the following form:

\[
W = \begin{pmatrix} w_{11} & w_{12} \\ w_{12} & w_{22} \end{pmatrix}
\]

Substitute this matrix in the equation (12). After several transformations, we find the elements of the matrix \( W \):

\[
w_{11} = \frac{-1 - 2f_{\theta}w_{12}}{2f_u}, \hspace{1cm} (13)
\]

\[
w_{22} = \frac{-1 - 2g_{u}w_{12}}{2g_{\theta}}, \hspace{1cm} (14)
\]

\[
w_{12} = \frac{f_u f_{\theta} + g_u g_{\theta}}{2(f_u^2 g_{\theta} + g_u^2 f_u - f_u f_{\theta} g_u - f_u g_u g_{\theta})}, \hspace{1cm} (15)
\]

and the eigenvalues of the matrix:

\[
\lambda_{1,2} = \frac{w_{11} + w_{22} \pm \sqrt{(w_{11} + w_{22})^2 - 4(w_{11}w_{22} - w_{12}^2)}}{2}, \hspace{1cm} (16)
\]

where

\[
f_u = \frac{\partial f}{\partial u} (\bar{u}, \bar{\theta}), \quad f_{\theta} = \frac{\partial f}{\partial \theta} (\bar{u}, \bar{\theta}), \quad g_u = \frac{\partial g}{\partial u} (\bar{u}, \bar{\theta}), \quad g_{\theta} = \frac{\partial g}{\partial \theta} (\bar{u}, \bar{\theta}).
\]

Figure 6. The slow curve (red line) and the trajectory (black line) of the system (1), (2) with the effect of random perturbations.

Figure 7 demonstrates Theoretical sensitivity to the random perturbations in the stable point concerning to parameter \( k_e \). Note that one of the eigenvalues is very small, so the degree of stochastic sensitivity is determined by the highest eigenvalue. This figure shows that the stochastic equilibrium becomes more sensitive to random perturbations when the value of the control parameter \( k_e \) is higher.
5. Noise-induced transitions

Qualitative changes are possible in the stochastic model under the noise influence: when a certain critical value of the noise intensity $\epsilon_{cr}$ is reached, a transition from one deterministic attractor (stable point) to another (limit cycle) occurs. Random trajectories leave the pool of attraction of the deterministic attractor and wind up the limit cycle. Such qualitative changes in the system are called noise-induced transitions. Consider the change in the stochastic phase portrait depending on the intensity of the noise.

Figures 8 and 9 represent a small noise in the model. The scatter of random states is always in the neighbourhood of the equilibrium.

Rare transitions occur through the unstable cycle to the limit cycle and back with increasing noise intensity. In that case, the oscillations of mixed type are observed, see Figures 10 and 11.

However, with the growth of the control parameter and its approach to the zone where the stable point loses its stability, the oscillations take the form of a larger-amplitude, see Figures 12 and 13. Transitions become more frequent with a further increase of noise intensity.

Thus, using the stochastic sensitivity function, we can predict the value of the noise intensity $\epsilon_{cr}$ corresponding to the beginning of the transitions.

We demonstrate transitions induced by noise for the control parameter $k_e = 0.85$ and find out that the critical value of the noise intensity approximately equals to $\epsilon_{cr} \approx 0.009495$.

After searching for the critical values of the noise intensity for the value of the parameter $k_e$ from the stable zone, we obtain the dependence of the $\epsilon_{cr}$ from the control parameter.
Figure 10. Noise-induced transitions for the parameter $k_e = 0.85$, $\epsilon = 0.0098$.

Figure 11. Noise-induced transitions for the parameter $k_e = 0.85$, $\epsilon = 0.0098$. Scatter of random states of the $\theta = \bar{\theta}(t)$.

Figure 12. Noise-induced transitions for the parameter $k_e = 0.85$, $\epsilon = 0.02$.

Figure 13. Noise-induced transitions for the parameter $k_e = 0.85$, $\epsilon = 0.02$. Scatter of random states of the $\theta = \bar{\theta}(t)$.

As it can be seen from Figure 14, the increasing control parameter value leads to decreasing of the noise intensity value, at which the transitions between attractors begin to appear.

Figure 14. Dependence of the critical noise intensity value from the control parameter $k_e$. 
6. Conclusion
The dynamic model of the electrocatalytic reaction under the influence of naturally present random perturbations was explored.

The results obtained in the work indicate that the situation can become unstable not only because of improperly actions but also due to small changes in some parameters. With such changes, the system can undergo the transition from small oscillations to the critical regime simulated by the canard, which corresponds to oscillations in the system and unstable reaction behaviour.

7. References
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Acknowledgment

This work was supported by the Ministry of Education and Science of the Russian Federation as a part of the Program “Research and development on priority directions of scientific-technological complex of Russia for 2014–2020” (Project RFMEFI58716X0033).