Real-Time Optimization of a Tubular Reactor with Distributed Feed

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An adaptive extremum seeking control scheme for a class of nonlinear distributed parameter systems is presented. It addresses the real-time optimization of a parallel chemical reaction system in an isothermal tubular reactor with uniform distributed feed described by a set of hyperbolic partial differential equations. Only limited knowledge of the kinetics is assumed. An adaptive learning technique is introduced to design an extremum seeking algorithm that drives the system states to a set point that maximizes the value of an objective function. Lyapunov's stability theorem is used in the design of the extremum seeking controller structure and the development of the parameter learning laws. *AIChE J,* 52: 2120–2128, 2006
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Introduction

In the field of chemical reaction engineering, considerable research and development have been dedicated to the improvement of selectivity in parallel reaction systems via the development of new catalysts. This objective can, however, be achieved by alternative approaches. The concentration of reactants plays a central role in the product distribution in a parallel reaction system.[1] Moreover, reaction orders in the kinetic relations are of prime importance.[2,3] A high reactant concentration favors the highest apparent order reaction, while a low concentration favors the lowest apparent order reaction. In the classical study of tubular reactors, the reactants are fed at the reactor input and the product is collected at the output. It is obviously difficult, if not impossible, to obtain some desired concentration profile along the reactor by only acting on the inlet concentration. If, on the other hand, a distributed feeding strategy is possible, then the reactants can be introduced either at the reactor input or at different points along the reactor.

Different research works have investigated the use of feed distribution to increase the selectivity of parallel reactions.[4-8] The results of these works are based on the perfect knowledge of the structure and the parameters of the reaction kinetics. This is especially true in the application of optimal controller design, where a perfect knowledge of the process model is required to perform a reliable off-line optimization.[9] Since the reaction kinetics may be badly known in practice, optimal control based on approximate kinetic models can often result in poor performance of the system.

Since the optimal set point depends on the process parameters, we consider here control approaches that optimize process

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performance in the presence of model uncertainties, as in adaptive extremum seeking control.[10-12]

In this study we extend the previous results of adaptive extremum seeking to a class of problems described by a set of hyperbolic partial differential equations. We investigate an extremum seeking algorithm for isothermal tubular reactors with distributed feed. The problem consists of finding the optimal exit fraction of the product of interest resulting from a gas phase parallel reactions scheme. The tubular reactors with distributed feed include different feed policies, such as discrete feed equally spaced and/or equally distributed or continuous feed.[2,3] Performances of the tubular reactor are directly linked to the feed configuration. Increasing the number of injection points increases the performance of the reactor for favorable kinetics.[2] The discrete feed becomes continuous when the number of injection points tends to infinity. In this article, we focus on uniform (continuous) distributed feed, which can be realized in practice through, for example, a membrane reactor. The uniform distributed feed can be described with a single variable independent of the spatial coordinate. Since the distribution profile of the feed is uniform along the tubular reactor, the distributed feed is completely defined by its magnitude. This configuration allows study of the behavior of the adaptive extremum seeking controller for systems described by partial differential equations.

The reactor configuration considered in this study is a one-dimensional tubular reactor with two reactants, one being fed along the reactor length and the other being fed at the reactor input. Furthermore, the uniform distributed feed is kept within a realistic range by introducing input constraints into the definition of the problem. Indeed, a modified cost function is used to account for the input constraints.

Moreover, we assume a limited knowledge of the reaction kinetics. Therefore, a Lyapunov-based adaptive learning control technique is used to steer the system to its unknown extremum. We also show that a persistence of excitation (PE) condition is necessary to guarantee the convergence of the extremum seeking mechanism.

The article is organized as follows. First we present the model of the tubular reactor with distributed feed. We then describe the problem and provide some preliminary analysis. Then the parameter estimation algorithm and the controller are designed based on Lyapunov functions. Finally, we provide simulation results that illustrate the performance of the controller.

Reactor Modeling

We first derive the model of the isothermal tubular reactor with distributed feed using component balances. Note that this model is not directly used in the solution of the real-time optimization problem. The model is designed to check the efficiency of the extremum seeking controller on a simulated tubular reactor with distributed feed.

The reactor is supposed to operate in plug-flow without diffusion. The reaction considered in this study is assumed to be a gas phase reaction with mole number variation. Under these assumptions, a differential section of the tubular reactor can be represented as in Figure 1.

The variable N_i is the number of mole of component i in the differential volume dV, while F_i and $(-r_i)$ are the molar flow rate and the rate of disappearance of component i by the reaction per unit of volume, respectively. The quantity qdz is the volume flow introduced in the differential section by the distributed feed, and C_i^{in} is the concentration of component i in the distributed feed. The mass balance equation is:

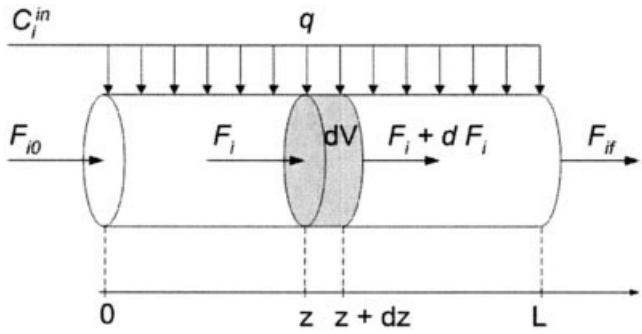


Figure 1. Tubular reactor with uniform distributed feed.

$$\frac{\partial N_i}{\partial t} = -dF_i + r_i dV + qC_i^{in} dz. \tag{1}$$

The molar flow rate F_i is the product between the volume flow v and the concentration C_i in the reactor, while the number of moles N_i is the product between the differential volume dV and the concentration C_i. Then, by dividing by dV, Eq. 1 becomes:

$$\frac{\partial C_i}{\partial t} = -\frac{v}{S}\frac{\partial C_i}{\partial z} - \frac{C_i}{S}\frac{\partial v}{\partial z} + r_i + \frac{q}{S}C_i^{in} \tag{2}$$

where S is the section of the tubular reactor.

Let us now derive the expression for $\partial v/\partial z$. The mass balance for the total concentration C_{tot} is given by summing Eq. 2 for all components (reactants, products, and inert gas):

$$\frac{\partial C_{tot}}{\partial t} = -\frac{v}{S}\frac{\partial C_{tot}}{\partial z} - \frac{C_{tot}}{S}\frac{\partial v}{\partial z} + \sum_{i=1}^{n} r_i + \frac{q}{S}C_{tot}^{in} \tag{3}$$

where n is the number of components and C_{tot}^{in} is the total concentration in the distributed feed. If the tubular reactor operates at constant pressure, the ideal gas law gives that the total concentration is a constant, that is:

$$\frac{\partial C_{tot}}{\partial t} = \frac{\partial C_{tot}}{\partial z} = 0. \tag{4}$$

Moreover, if the temperature and the pressure in the distributed feed are the same as in the feed at the reactor input, the total concentration in the distributed feed C_{tot}^{in} is the same as the total concentration C_{tot}. By rearranging the terms, Eq. 3 becomes:

$$\frac{1}{S}\frac{\partial v}{\partial z} = \frac{1}{C_{tot}}\sum_{i=1}^{n} r_i + \frac{q}{S}. \tag{5}$$

A general model for the tubular reactor with distributed feed is derived from Eqs. 2 and 5 by using the following dimensionless variables:

$$\tau = \frac{LS}{v_0}, \quad \theta = \frac{t}{\tau}, \quad \xi = \frac{z}{L}, \quad r_i = \frac{\tau r_i}{C_{tot}}$$
$$x_i = \frac{C_i}{C_{tot}}, \quad x_i^{in} = \frac{C_i^{in}}{C_{tot}}, \quad u = \frac{qL}{v_0}, \quad v' = \frac{v}{v_0}$$

where v_0 is the volume flow rate at the reactor input.

The dimensionless model of the tubular reactor with distributed feed is given by:

$$\frac{\partial x_i}{\partial \theta} = -v' \frac{\partial x_i}{\partial \xi} - x_i \frac{\partial v'}{\partial \xi} + r_i + u x_i^{in} \tag{6}$$

$$\frac{\partial v'}{\partial \xi} = \sum_{i=1}^{n} r_i + u$$

where $\theta \in \mathbf{R}^+$ and $\xi \in [0, 1]$ are the time and space variables; $x_i \in S_X \subset \mathbf{R}^+$ is the fraction of component i; $v' \in S_v \subset \mathbf{R}^+$ is the dimensionless flow rate; and $u \in S_u \subset \mathbf{R}^+$ is the distributed feed (control action). The initial condition is given by:

$$x_i(0, \xi) = x_i^{\theta_0} \tag{7}$$

while the boundary conditions are given by:

$$x_i(\theta, 0) = x_{i0}; \quad v'(\theta, 0) = v_0. \tag{8}$$

Problem Formulation

The problem considered in this article is to optimize a chemical reaction scheme with favorable kinetics[2] in a tubular reactor. As a case study, the following gas phase parallel chemical reactions are considered:

$$\tau_1 A + B \xrightarrow{k_1} \tau_C C + \tau_{E1} E$$

$$\tau_2 A + B \xrightarrow{k_2} \tau_D D + \tau_{E2} E \tag{9}$$

where the reaction kinetics depend on reactant partial pressures:

$$r_C = f_1(P_A, P_B) \tag{10}$$

$$r_D = f_2(P_A, P_B) \tag{11}$$

This general case can be illustrated by oxidation or oxidative dehydrogenation of hydrocarbon.[13-16] A more specific case is the oxidation of ethylene, which is used for the simulation of the proposed controller.

The objective is to maximize the fraction of the product of interest C at the reactor output. The selectivity of C can be defined by the ratio of the production rate of C with the production rate of the undesired by product D:

$$S = \frac{r_C}{r_D} = \frac{f_1(P_A, P_B)}{f_2(P_A, P_B)}. \tag{12}$$

The tubular reactor with uniform distributed feed of A can be useful if the selectivity of C is improved by maintaining a low concentration of A.[2,3]

Here we assume that the measurements of the fraction of C at the reactor output are available on-line and that the reaction rates are unknown. The objective is to find the value of the uniform distributed feed u that maximizes the reactor exit fraction of C at steady state.

Note that at steady state, the exit fraction of C (x_C^{ss}) is a function only of the distributed feed, denoted $\pi(u)$, all other parameters being constant.

Assumption 1. The function $\pi(u)$ is continuously differentiable and admits a maximum on S_U.

Since an analytical solution of Eq. 6 is difficult, if not impossible, to obtain even if the reaction rates are known, an analytical expression for $\pi(u)$ cannot be derived. Let us, therefore, choose to parameterize this unknown function by using a universal approximation,[10,12] for example:

$$\pi(u) = W^T S(u) + \mu_l(\theta) \tag{13}$$

where $\mu_l(\theta)$ is the NN approximation error and where S_U, the basis function vector, is given by:

$$S(u) = [s_1(u), s_2(u), \ldots, s_l(u)]^T \tag{14}$$

$$\text{with } s_i(u) = \exp\left(\frac{-(u - \varphi_i)^T (u - \varphi_i)}{\sigma_i^2}\right) - \frac{(u - \varphi_i)^2}{\sigma^2} \tag{15}$$

for $i = 1,2,\ldots,l$. The basis weights parameters W are assumed to take values in a compact set of R^l. The functional approximation of the equilibrium manifold is dependent on the center of the receptive field φ_i and on the width of the Gaussian function σ_i. The convexity of the functions $s_i(u)$ is ensured by choosing an appropriate value for σ. Universal approximation results stated in [17] and [18] indicate that if l is chosen sufficiently large, then $W^T S(u)$ can approximate any continuous function to any desired accuracy on a compact set. Then, even if the network of chemical reactions (Eq. 9) is more complex than a two reaction system, the neural network approximation technique can approximate the more complex function $\pi(u)$ without increasing the computational complexity.

From a physical point of view all variables in the system (Eq. 6) are continuous functions of space and time. As a result, the steady-state exit fraction of C (x_C^{ss}) noted $\pi(u(\theta))$ is a continuous function of time. The continuity of the approximation error is then guaranteed by the continuity of basis functions. Moreover, the continuity and the bound of the approximation error justify the following assumption:

Assumption 2. The NN approximation error satisfies $|\partial \mu_l(\theta)/\partial \theta| < d\bar{\mu}_l$ with constant $d\bar{\mu}_l > 0$ over a compact set in R^+.

Controller Design

In this section of the article the control law is designed to steer the system to its unknown maximum. Constraints are imposed in the optimization problem to maintain the control action in a realistic range.

This section is organized as follows. The cost function to optimize is first defined. The control law to steer the system to the estimate optimum is then derived. Finally, an adaptive technique is designed that ensures the convergence of the reactor system to the true unknown optimum.

Optimum condition

The objective of the control strategy is to track the optimum steady-state fraction of C at the reactor output. The corresponding optimization problem is stated as follows:

$$\max_u x_C^{ss}$$

$$\text{s.t.} \quad u_{\min} < u < u_{\max} \tag{16}$$

To solve this problem, we define a cost function with log-barrier functions[19] to account for the input constraints:

$$J(u) = x_C^{ss} + \mu_1 \log(u - u_{\min} - \varepsilon_1) + \mu_2 \log(u_{\max} - u - \varepsilon_2) \tag{17}$$

where μ_1, μ_2, ε_1, ε_2 are strictly positive constants. An appropriate choice of these constants allows preserving the shape of the function x_C^{ss} on the available range of the control action while the cost function will sharply decrease close to the boundaries.

Under the assumption of convexity of $J(u)$, the cost function is maximized when

$$\frac{\partial J}{\partial u}(u^*) = 0. \tag{18}$$

By using the parameterization (Eq. 13), the cost function becomes:

$$J(u) = W^T S(u) + \mu_1 \log(u - u_{\min} - \varepsilon_1) + \mu_2 \log(u_{\max} - u - \varepsilon_2). \tag{19}$$

Since the true parameters W are assumed to be unknown, the following cost function is considered:

$$\hat{J}(u) = \hat{W}^T S(u) + \mu_1 \log(u - u_{\min} - \varepsilon_1) + \mu_2 \log(u_{\max} - u - \varepsilon_2) \tag{20}$$

where $\hat{W}$ is the estimate of W. Consequently, the optimum $\hat{u}^*$ is the solution of:

$$\frac{\partial \hat{J}}{\partial u}(\hat{u}^*) = 0. \tag{21}$$

The gradient of $\hat{J}(u)$ with respect to u is equal to:

$$\Gamma_1 = \left(\hat{W}^T \frac{\partial S(u)}{\partial u} \right) + \frac{\mu_1}{u - u_{\min} - \varepsilon_1} - \frac{\mu_2}{u_{\max} - u - \varepsilon_2} \tag{22}$$

while the Hessian of $\hat{J}(u)$ with respect to u is written as follows:

$$\Gamma_2 = \left(\hat{W}^T \frac{\partial^2 S(u)}{\partial u^2} \right) - \frac{\mu_1}{(u - u_{\min} - \varepsilon_1)^2} - \frac{\mu_2}{(u_{\max} - u - \varepsilon_2)^2}. \tag{23}$$

In the following subsection, an appropriate control law is designed to steer the system to its maximum.

Optimum tracking

The objective of the control law is to drive the control action to the value that cancels the gradient Γ_1.

Let us define:

$$z_s = \Gamma_1 - d(\theta) \tag{24}$$

where $d(\theta)$ is a dither signal. The dither signal is introduced to guarantee the convergence of the estimate parameters. This assertion is demonstrated in the last subsection.

The convergence of z_s to zero is analyzed by using Lyapunov theory.

Let us consider the Lyapunov function candidate:

$$V_1 = \frac{1}{2} z_s^2. \tag{25}$$

By taking the time derivative of V_1, we obtain:

$$\dot{V}_1 = z_s \left(\Gamma_2 \dot{u} + \dot{\hat{W}}^T \frac{\partial S}{\partial u} - \dot{d}(\theta) \right) \tag{26}$$

To achieve the negative definiteness of $\dot{V}_1$, the following dynamical control law is used:

$$\dot{u} = \frac{1}{\Gamma_2} \left(-k_z z_s - \dot{\hat{W}}^T \frac{\partial S}{\partial u} + \dot{d}(\theta) \right) \tag{27}$$

where $k_z > 0$ is a gain to be assigned. The substitution of Eq. 27 into Eq. 26 yields:

$$\dot{V}_1 = -k_z z_s^2. \tag{28}$$

Hence, according to Lyapunov theory, the control law described by Eq. 27 guarantees the convergence to the maximum of the cost function (Eq. 20). Since the kinetic parameters are

assumed to be unknown, an update law for these parameters is needed to reconstruct the unknown function $\pi(u)$.

Adaptation law

The fraction x_c is the sum between the steady-state fraction (x_C^{ss}) and the transient fraction (Δx_C):

$$x_C(\theta, 1) = x_C^{ss}(u(\theta), 1) + \Delta x_C(\theta). \tag{29}$$

By using the neural network approximation, the variation of C at the reactor output can be expressed by:

$$\dot{x}_C(\theta, 1) = W^T \frac{\partial S}{\partial u}\dot{u} + \frac{\partial \mu_l}{\partial \theta} + \frac{\partial \Delta x_C(\theta)}{\partial \theta}. \tag{30}$$

Even if x_c is measured at the reactor output, its time evolution is unknown because W is unknown. The estimate $\hat{x}_C$ is generated by the following estimation law:

$$\dot{\hat{x}}_C(\theta, 1) = \hat{W}^T \frac{\partial S}{\partial u}\dot{u} + c(\theta)^T\dot{\hat{W}} + k_x(x_C - \hat{x}_C) \tag{31}$$

where $c(\theta)$ are time-varying vector valued functions and k_x is a strictly positive constant.

The dynamics of the estimation error ($\tilde{x}_C = x_C - \hat{x}_C$) is given by:

$$\dot{\tilde{x}}_C = \tilde{W}^T \frac{\partial S}{\partial u}\dot{u} + \frac{\partial \mu_l}{\partial \theta} + \frac{\partial \Delta x_C(\theta)}{\partial \theta} - c(\theta)^T\dot{\hat{W}} - k_x\tilde{x}_C \tag{32}$$

where $\tilde{W} = W - \hat{W}$ is the vector of parameter estimation errors. Note that in the rest of the article, $\tilde{x}_C(\theta, 1)$ will be denoted by $\tilde{x}_C$.

The next step is to prove that the estimation error $\tilde{x}_C$ and the parameter estimation errors $\tilde{W}$ converge to zero. In that case, the optimum of the approximate function reached in the optimization step will be in the neighborhood of the real optimum of the system.

Let us define:

$$\eta = \tilde{x}_C - c(\theta)^T\tilde{W} \tag{33}$$

and the following Lyapunov function:

$$V_2 = \frac{1}{2}\eta^2. \tag{34}$$

Its time derivative is given by the following expression:

$$\dot{V}_2 = \eta\left(\tilde{W}^T \frac{\partial S}{\partial u}\dot{u} + \frac{\partial \mu_l}{\partial \theta} + \frac{\partial \Delta x_C(\theta)}{\partial \theta} - k_x\tilde{x}_C - \dot{c}(\theta)^T\tilde{W}\right). \tag{35}$$

Substitution of Eq. 33 into Eq. 35 yields:

$$\dot{V}_2 = \eta\left(\tilde{W}^T \frac{\partial S}{\partial u}\dot{u} + \frac{\partial \mu_l}{\partial \theta} + \frac{\partial \Delta x_C(\theta)}{\partial \theta} - k_x\eta - k_x c(\theta)^T\tilde{W} - \dot{c}(\theta)^T\tilde{W}\right). \tag{36}$$

Let us consider that $\dot{c}(\theta)$ is given by:

$$\dot{c}(\theta) = -k_x c(\theta) + \dot{u}\frac{\partial S}{\partial u} \tag{37}$$

with $k_x > 0$.

Substitution of Eq. 37 into Eq. 36 gives:

$$\dot{V}_2 = -k_x\eta^2 + \frac{\partial \mu_l}{\partial \theta}\eta + \frac{\partial \Delta x_C(\theta)}{\partial \theta}\eta. \tag{38}$$

Assumption 3. The time derivative of $\Delta x_C(\theta)$ is bounded: $|\partial \Delta x_C(\theta)/\partial\theta| < d\bar{\Delta}x_C$ with $d\bar{\Delta}x_C > 0$.

The rate of growth of the transient $\Delta x_C(\theta)$ is given by:

$$\frac{\partial x_C(\theta)}{\partial \theta} = \frac{\partial x_C^{ss}(u(\theta))}{\partial u}\frac{\partial u(\theta)}{\partial \theta} + \frac{\partial \Delta x_C(\theta)}{\partial \theta}$$

From a physical point of view, all variables in Eq. 6 are continuous bounded functions of time. The rate of growth of the exit fraction of C ($\partial x_C(\theta)/\partial\theta$) is then a continuous bounded function of time. For the same reasons, the steady state solution of Eq. 6 gives that $\partial x_C^{ss}(u(\theta))/\partial u$ is a continuous bounded function of time. Finally, the bound of the dynamic control (Eq. 71) fulfills Assumption 3.

By using the upper bounds $|\partial \mu_l/\partial\theta| < d\bar{\mu}_l$ and $|\partial \Delta x_C(\theta)/\partial\theta| < d\bar{\Delta}x_C$, we get:

$$\dot{V}_2 \le -k_x\eta^2 + d\bar{\mu}\eta + d\bar{\Delta}x_C\eta. \tag{39}$$

By completing the square, Eq. 39 becomes:

$$\dot{V}_2 \le -\left(k_x - \frac{k_1}{2} - \frac{k_2}{2}\right)\eta^2 + \frac{1}{2k_1}d\bar{\mu}_l^2 + \frac{1}{2k_2}d\bar{\Delta}x_C^2 \tag{40}$$

where k_1 and k_2 are positive constants. The exponential convergence of the state η is guaranteed by using an appropriate gain function k_x. Indeed, by setting the gain equal to:

$$k_x = k_\eta + \frac{k_1}{2} + \frac{k_2}{2} \tag{41}$$

where $k_\eta > 0$, we obtain the following inequality:

$$\dot{V}_2 \le -2k_\eta V_2 + \frac{1}{2k_1}d\bar{\mu}_l^2 + \frac{1}{2k_2}d\bar{\Delta}x_C^2. \tag{42}$$

By integrating Eq. 42 we get:

$$V_2(\theta) \le \alpha_1 e^{-2k_\eta(\theta-\theta_0)} + \alpha_2 \tag{43}$$

where $\alpha_1 = V_2(\theta_0)$ and $\alpha_2 = (1/2k_1k_\eta)d\bar{\mu}_l^2 + (1/2k_2k_\eta)d\bar{\Delta}x_C^2$.

The exponential convergence of the state η is given by:

$$\|\eta\| \leq \alpha_3 e^{-k_\eta(\theta-\theta_0)} + \alpha_4 \tag{44}$$

where $\alpha_3 = \sqrt{2V_2(\theta_0)}$ and $\alpha_4 = (1/\sqrt{k_1k_\eta})|d\bar{\mu}_l| + (1/\sqrt{k_2k_\eta})|d\bar{\Delta}x_C|$.

Consequently, Eq. 44 shows that the state η converges to a small neighborhood of the origin.

It remains to show that the estimation error $\tilde{x}_C$ and the parameter estimation errors $\tilde{W}$ converge to a small neighborhood of the origin. This can be achieved by using an appropriate choice of the parameter update law. Since $\tilde{x}_C$ is known, we can choose the parameter update law as follows:

$$\dot{\hat{W}} = \gamma_w c(\theta)\tilde{x}_C. \tag{45}$$

To guarantee that our parameters remain in a compact set Ω_w defined by:

$$\Omega_w = \{\hat{W} \mid \|\hat{W}\| \leq w_m\} \tag{46}$$

we use the following projection algorithm:

$$\dot{\hat{W}} = \begin{cases} \gamma_w c(\theta)\tilde{x}_C & \text{if } \|\hat{W}\| < w_m \text{ or} \\ & \text{if } \|\hat{W}\| = w_m \\ & \text{and } \hat{W}^T c(\theta)\tilde{x}_C \leq 0 \\ \gamma_w\left(I - \dfrac{\hat{W}\hat{W}^T}{\hat{W}^T\hat{W}}\right)c(\theta)\tilde{x}_C & \text{otherwise} \end{cases} \tag{47}$$

In the following, we show that under suitable assumptions, the parameter update law (Eq. 47) guarantees the convergence of the parameter estimation errors to zero.

Lemma 1. Consider the differential equation:

$$\dot{z}(\theta) = -\phi(\theta)\phi(\theta)^T z(\theta) \tag{48}$$

and assume that there exist two positive constants T and k such that:

$$\int_\theta^{\theta+T} \phi(\tau)\phi(\tau)^T d\tau \geq kI \tag{49}$$

then the origin is a globally exponentially stable equilibrium of the system.

The proof of this lemma can be found in [20].

Assumption 4. The dither signal $d(\theta)$ is such that the time-varying vector $c(\theta)$ satisfies:

$$\int_\theta^{\theta+T} c(\tau)c(\tau)^T d\tau \geq k_N I_N$$

for positive constants T and k_N.

Under Assumption 4, Lemma 1 shows that the origin of the differential equation:

$$\dot{\tilde{W}} = -\gamma_w c(\theta)c(\theta)^T\tilde{W} \tag{50}$$

is an exponentially stable equilibrium. Moreover, the proof of Lemma 1 shows that:

$$-\tilde{W}^T c(\theta)c(\theta)^T\tilde{W} \leq -k_w\|\tilde{W}\|^2 \tag{51}$$

with k_w a positive constant.

Let us define the Lyapunov function:

$$V_w = \frac{1}{2\gamma_w}\tilde{W}^T\tilde{W}. \tag{52}$$

The rate of change of V_w along the trajectories of the system is given by:

$$\dot{V}_w = -\tilde{W}^T c(\theta)c(\theta)^T\tilde{W} - \tilde{W}^T c(\theta)\eta + \begin{cases} 0 & \text{if } \|\hat{W}\| < W_m \text{ or} \\ & \text{if } \|\hat{W}\| = w_m \\ & \text{and } \hat{W}^T c(\theta)\tilde{x}_C \leq 0 \\ \hat{W}^T\dfrac{\hat{W}\hat{W}^T}{\hat{W}^T\hat{W}}(c(\theta)c(\theta)^T\tilde{W} + c(\theta)\eta) & \text{otherwise} \end{cases} \tag{53}$$

The property of the projection algorithm shows that:

$$\dot{V}_w \leq -\tilde{W}^T c(\theta)c(\theta)^T\tilde{W} - \tilde{W}^T c(\theta)\eta. \tag{54}$$

By completing the squares we get:

$$\dot{V}_w \leq -\frac{1}{2}\tilde{W}^T c(\theta)c(\theta)^T\tilde{W} + \frac{1}{2}\eta^2. \tag{55}$$

Introducing Eq. 51 into Eq. 55 leads to:

$$\dot{V}_w \leq -\frac{k_w}{2}\|\tilde{W}\|^2 + \frac{1}{2}\eta^2 = -k_w\gamma_w V_w + \frac{1}{2}\eta^2. \tag{56}$$

Integration of Eq. 56 yields:

$$V_w(\theta) \leq V_w(\theta_0)e^{-k_w\gamma_w(\theta-\theta_0)} + \int_{\theta_0}^{\theta} e^{-k_w\gamma_w(\theta-\tau)}\frac{\eta(\tau)^2}{2}d\tau. \tag{57}$$

The analytical expression of $V_w(t)$ is derived by introducing Eq. 43 into Eq. 57:

$$V_w(\theta) \leq V_w(\theta_0)e^{-k_w\gamma_w(\theta-\theta_0)} + \int_{\theta_0}^{\theta} e^{-k_w\gamma_w(\theta-\tau)}(\alpha_1 e^{-2k_q(\theta-\theta_0)} + \alpha_2)d\tau \tag{58}$$

$$\leq \alpha_5 e^{-\lambda_1(\theta-\theta_0)} + \alpha_6 \tag{59}$$

where

$$\lambda_1 = \min\{k_w\gamma_w, 2k_\eta\}, \tag{60}$$

$$\alpha_5 = \max\left\{V_w(\theta_0), \left|\frac{\alpha_1}{k_w\gamma_w - 2k_\eta}\right|, \frac{\alpha_2}{k_w\gamma_w}\right\}, \tag{61}$$

$$\alpha_6 = \frac{\alpha_2}{k_w\gamma_w}. \tag{62}$$

The exponential convergence of the parameter estimation error is given by:

$$\|\tilde{W}\| \leq \alpha_7 e^{-\lambda_2(\theta-\theta_0)} + \alpha_8 \tag{63}$$

where

$$\lambda_2 = \min\left\{\frac{k_w\gamma_w}{2}, k_\eta\right\}, \tag{64}$$

$$\alpha_7 = \max\left\{\sqrt{2\gamma_w V_w(\theta_0)}, \sqrt{\left|\frac{2\gamma_w\alpha_1}{k_w\gamma_w - 2k_\eta}\right|}, \sqrt{\frac{2\alpha_2}{k_w}}\right\}, \tag{65}$$

$$\alpha_8 = \sqrt{\frac{2\alpha_2}{k_w}}. \tag{66}$$

The above proof shows that the parameter estimation errors converge to an adjustable neighborhood of the origin by increasing the controller gains.

By using Eq. 33, the estimation error is bounded as follows:

$$\|\tilde{x}_C\| \leq \|\eta\| + \|c(\theta)\|\,\|\tilde{W}\|. \tag{67}$$

Equations 44 and 63 show that $\|\eta\|$ and $\|\tilde{W}\|$ are bounded. The exponential convergence of the estimation error $\tilde{x}_C$ is then demonstrated if the signal $c(\theta)$ is also bounded. The signal $c(\theta)$ is the solution of:

$$\dot{c}(\theta) = -k_x c(\theta) + \dot{u}\frac{\partial S}{\partial u}. \tag{68}$$

Under the assumption that $\|\dot{u}(\partial S/\partial u)\|$ is bounded by β, the following expression is satisfied:

$$\|c(\theta)\| \leq \|c(\theta_0)\|e^{-k_x(\theta-\theta_0)} + \frac{\beta}{k_x}. \tag{69}$$

Let us show that the bound β exists and is finite.

From Eq. 14 we get:

$$\left\|\frac{\partial S}{\partial u}\right\| \leq 4\,\frac{(u_{\max} - u_{\min})}{\sigma_m^2}\,\sqrt{l} \tag{70}$$

under the assumption that $u_{\min} < \varphi_i < u_{\max}$ and where $\sigma_m = \min\{\sigma, \sigma_i\}$ for $1 \leq i \leq l$.

The bound of the dynamic control law is derived from Eq. 27:

$$\|\dot{u}\| \leq \frac{1}{|\Gamma_2|}\left(k_z\|z_s\| + \|\dot{\hat{W}}\|\left\|\frac{\partial S}{\partial u}\right\| + \|\dot{d}(\theta)\|\right). \tag{71}$$

The strictly positivity of Γ_2 is guaranteed by the convexity condition. Moreover, the norms $\|z_s\|$, $\|\dot{\hat{W}}\|$, and $\|\partial S/\partial u\|$ are bounded from Eqs. 28, 63, and 70, respectively. The dynamic control law is then bounded by choosing a bounded dither signal. This shows that $\|\dot{u}(\partial S/\partial u)\|$ is bounded by a positive real value β.

The convergence of the estimation error $\tilde{x}_C$ is then given by:

$$\|\tilde{x}_C\| \leq \alpha_3 e^{-k_\eta(\theta-\theta_0)} + \alpha_4 + \left(\|c(\theta_0)\|e^{-k_x(\theta-\theta_0)} + \frac{\beta}{k_x}\right) \times (\alpha_7 e^{-\lambda_2(\theta-\theta_0)} + \alpha_8) \leq \alpha_9 e^{-\lambda_3(\theta-\theta_0)} + \alpha_{10} \tag{72}$$

where

$$\lambda_3 = \min\{k_\eta, k_x, \lambda_2, k_x + \lambda_2\}, \tag{73}$$

$$\alpha_9 = \max\left\{\alpha_3, \alpha_7\|c(\theta_0)\|, \alpha_7\frac{\beta}{k_x}, \alpha_8\|c(\theta_0)\|\right\}, \tag{74}$$

$$\alpha_{10} = \max\left\{\alpha_4, \alpha_8\frac{\beta}{k_x}\right\}. \tag{75}$$

The convergence of the estimated gradient z_s and the estimation error $\tilde{x}_C$ provides the convergence of the closed-loop system to an adjustable neighborhood of the unknown steady-state optimum.

Theorem 1. Consider the parallel reactions (Eq. 9) taking place in a tubular reactor with uniform distributed feed. The control law (Eq. 27) with the parameter update law (Eq. 47) and the persistent excitation condition such that:

$$\int_\theta^{\theta+T} c(\tau)c(\tau)^T \geq k_N I_N$$

for positive constants T and k_N, drive the system (Eq. 6) to the maximum exit concentration of the product of interest.

Proof. The proof is straightforward from the above arguments and is omitted.

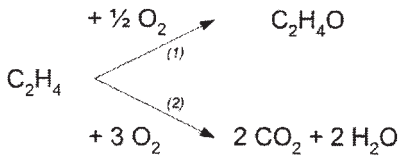
Simulation Results

This section presents simulation results of the proposed adaptive extremum seeking controller applied to ethylene epoxidation. The epoxidation of ethylene produces ethylene oxide, which is an important industrial chemical in the production of ethylene glycol and other chemicals.[21]

The reaction scheme is generally considered to be given by the following parallel network[13,15]:

Table 1. Kinetic Parameters Values

Parameter	r_1	r_2
k_0, mol/l s bar$^{1+n_i}$	1.45 10^5	1.96 10^6
E_{act}, KJ/mol	60.7	73.2
K_E, bar^{-1}	6.50	4.33
n_i	0.58	0.30



The rate of oxidation of the ethylene oxide is generally much smaller than the other two reactions. Lafarga et al.[13] proposed the following reaction rates for both reactions:

$$r_i = \frac{k_i P_E P_O^{n_i}}{(1 + K_E^i P_E)^2} \tag{76}$$

where P_E and P_O are the partial pressure of ethylene and oxygen, respectively, and k_i, K_E, and n_i are kinetic parameters presented in Table 1.

The selectivity of ethylene oxide increases if the partial pressure of ethylene decreases. The ethylene is then distributed along the reactor while the oxygen is fed at the reactor input.

The tubular reactor with distributed feed is simulated using the model described by Eq. 6 The fractions of ethylene, oxygen, and ethylene oxide are noted by x_A, x_B, and x_C, respectively. The initial conditions are set equal to:

$$x_A(0, \xi) = 0, \quad x_B(0, \xi) = 0.1, \quad x_C(0, \xi) = 0$$

while we consider the boundary conditions are set equal to:

$$x_{A0} = 0, \quad x_{B0} = 0.1, \quad x_{C0} = 0, \quad v_0' = 1.$$

The composition of the distributed feed is:

$$x_A^{in} = 0.4, \quad x_B^{in} = 0, \quad x_C^{in} = 0.$$

The centers of neural network approximation functions are evenly spaced points on the interval [0, 1] and the width σ_i is set to 2.50 for $1 \leq i \leq 6$ while σ is set to 1.25.

Table 2. Controller Parameters

Parameters	Value
$u_{\min}$, $u_{\max}$	0, 1
μ_1, μ_2	0.001, 0.001
ε_1, ε_2	0.001, 0.001
k_d	1
k_z	1
k_x	1
γ_w	10^6
w_m	100
$d(0)$	0
$c(0)$	$[000000]^T$
$\hat{W}(0)$	$[111111]^T$

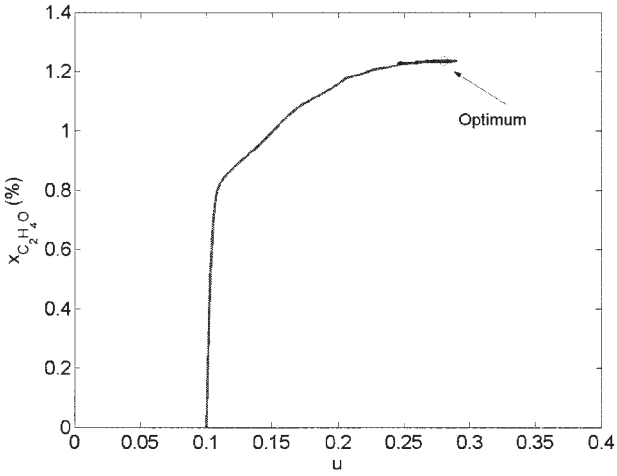


Figure 2. Phase diagram.

To ensure sufficient excitation, the dither signal is defined as follows[10-12]:

$$\dot{d}(\theta) = -k_d d(\theta) + a(\theta) \tag{77}$$

with k_d a strictly positive constant and:

$$a(\theta) = \sum_{i=1}^{10} (A_{1i}\sin(\omega_i\theta) + A_{2i}\cos(\omega_i\theta)) \tag{78}$$

where A_{1i} and A_{2i} are chosen as unit random numbers and ω_i are different frequencies.

The design parameters are presented in Table 2.

The simulation results are shown in Figures 2-4. Figure 2 shows the research path of the adaptive extremum seeking algorithm. We see that the control action u is kept within the constraints. Figures 3 and 4 present the evolution of the corresponding control input and exit fraction of ethylene oxide,

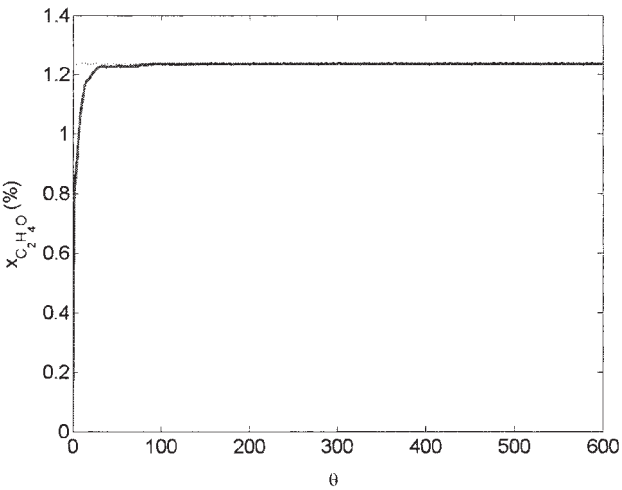


Figure 3. Concentration of ethylene oxide at the reactor output x_{C2H40} ("—") and its maximum x_{C2H40}^* ("- -").

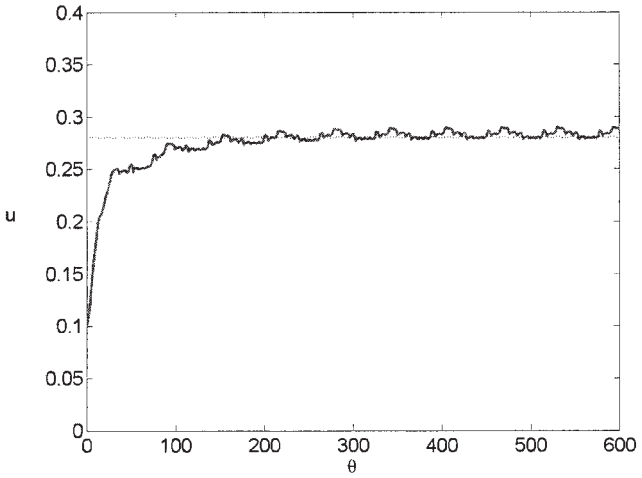


Figure 4. Control input u ("—") and the optimal control input u* ("- -").

respectively. The simulations demonstrate that the adaptive controller recovers the unknown optimum with only measurements of the exit fraction of ethylene oxide.

Note that the choice of the controller gains is not trivial. The convergence rate equations (Eqs. 26, 63, and 72) show that the rate of convergence to the true optimum increases if the gains k_z, k_x, and γ_w increase. Moreover, the radius of the neighborhood of convergence decreases if the gains k_x and γ_w increase. However, a large value for the gain k_x can cause a rapid convergence of the signal $c(\theta)$ to a neighborhood of zero. If the signal $c(\theta)$ becomes too small, the persistence of excitation condition is no longer satisfied in practice and the adaptation of the parameter estimates stops. Therefore, the gain k_x must not be too large in order to guarantee the convergence of the estimate parameters, yet not too small in order to guarantee a rate of convergence sufficiently fast.

Conclusions

We have solved a class of extremum seeking control problems for tubular reactors with uniform distributed feed and unknown reaction kinetic rate expressions subject to control action constraints. An adaptive learning technique was used to derive a relation between the exit fraction of the product of interest and the distributed feed. The adaptive extremum seeking algorithm converges to a small neighborhood of the unknown optimum when the dither signal satisfies a persistence of excitation condition.

Future works will concentrate on the case where the product of interest is not available for on-line measurement and the more general case where the distributed feed is not uniform.

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