An Isoperimetric Optimal Control Problem for a Non-Isothermal Chemical Reactor with Periodic Inputs

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Abstract

In this paper, we study the optimal control problem for a continuous stirred tank reactor (CSTR) that represents a reaction of the type "$A \rightarrow$ product". The reactor dynamics is described by a nonlinear system of ordinary differential equations controlled by two inputs: the inlet concentration and the inlet temperature. We formulate the problem of maximizing the average product of this reactor for a fixed consumption of the input component over a period of time. This kind of isoperimetric optimal control problem is analyzed by using the Pontryagin maximum principle with Lagrange multipliers. We show that the optimal controls are bang-bang and propose an upper bound for the number of switchings for the linearized problem with periodic boundary conditions. Numerical simulations confirm that our control strategy can be used to improve the reactor performance over a specified period of time in comparison to the steady-state operation.

Highlights

- Mathematical models of nonlinear reactions of the type "$A \rightarrow$ product" are considered.

- Periodic modulations of the inlet concentration and the temperature are used to control the reaction.

- Maximization of the reactor performance is treated as an isoperimetric optimal control problem.

- A theoretical estimate of the optimal number of switchings is obtained, and an algorithm for computing the control parameters is proposed.

- Simulation results illustrate the improvement of the performance by using bang-bang controls.

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1. Introduction

The optimization of periodic processes has been already the subject of several studies in chemical engineering and control theory (a survey of related works is presented in Section 2). The majority of publications in this area (cf. [1, 17, 18]) deals with smooth (or even constant) controls, so that no optimization problems with bang-bang strategies have been rigorously analysed up to now from the mathematical viewpoint. In this paper, we will consider a chemical reaction of the type "$A \to$ product" controlled by the inlet concentration $C_{Ai}(t)$ of A and the inlet temperature $T_i(t)$ at time t[1]. An attractive goal is to maximize the conversion of A to the product over a specified period of time $t \in [0, t_f]$, which can be formulated as the minimization of the outlet concentration $C_A(t)$ provided that the mean consumption of A is fixed as $\bar{C}_{Ai}$:

$$\frac{1}{t_f} \int_0^{t_f} C_A(t)\, dt \to \min,$$

$$\frac{1}{t_f} \int_0^{t_f} C_{Ai}(t)\, dt = \bar{C}_{Ai}.$$

We assume here that the process is operated periodically, so that the inputs and the state of the reactor are periodic functions of the period t_f. To the best of our knowledge, the above isoperimetric optimal control problem has not been solved up to now for mathematical models of chemical reactions. In order to evaluate the theoretical concept studied in this work, we selected one of the most simple reactors applied in chemical reaction engineering, namely the perfectly mixed continuously operated stirred tank reactor (CSTR). Analysing this concentrated system allowed us to extract generally valid trends in the most simple way exploiting analytical solutions. The selection of the model parameters was based on exploiting available knowledge regarding the kinetics of a standard liquid phase reaction, namely the hydrolysis of acetic acid anhydride. The features of this reaction are quite typical and thus allow some generalization with respect to main trends of periodic operation.

The main contributions of this paper can be summarized as follows.

- In Section 3, we formulate the problem of maximizing the performance of a single homogeneous n-th order reaction by using a time-varying inlet concentration (Problem 1). It is shown that each control satisfying the optimality conditions is bang-bang, and the procedure for computing the control parameters is described explicitly for the case $n = 2$. A numerical

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[1]We will introduce dimensionless variables to simplify the notations in the sequel.

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example illustrates that our bang-bang control strategy ensures better performance in comparison to the sinusoidal input modulation considered in the paper [14] in terms of the nonlinear frequency response function. In contrast to the approach of [11] dealing with a truncated Fourier series for a square wave input, we analyze the optimality conditions rigorously by taking into account the periodic boundary condition $C_A(0) = C_A(t_f)$.

- The isoperimetric optimal control problem (Problem 2) is considered in Section 4 for a nonlinear system of ordinary differential equations controlled by two inputs (the inlet concentration of A and the inlet temperature). The optimality conditions are analysed by the Pontryagin maximum principle with Lagrange multipliers (cf. [16]). In contrast to the results by N. Watanabe et al. [18], we consider here a cost function which is convex, but not strictly. We show that each optimal control is bang-bang and propose an upper bound for the number of switchings N for the linearized problem with periodic boundary conditions (Proposition 1). The switching times are defined by an auxiliary system of transcendental equations. For the case of small switching times, these transcendental equations can be approximated by polynomials. Our control design scheme is based on expansions of the composition of flows corresponding to extremal inputs.

- The simulation results in Section 5 confirm that bang-bang controls with two switchings can be used to improve the reactor performance over a specified period of time t_f in comparison to the steady-state operation.

2. Related Work

The paper by J. M. Douglas [1] was among the first theoretical studies, where the effects of time-varying inputs were estimated for nonlinear reaction models. A second-order isothermal reaction was considered there under the assumption that the feed composition is modulated by the sine function, and the method of small parameter was used to approximate the output behavior with correction terms of different orders of magnitude. It was shown that the frequency response of the system under consideration contains higher order harmonic terms in additional to the fundamental component. It follows from the analysis of the frequency response function that the average output value is close to the corresponding steady-state value for very low and very high frequency inputs. The case of simultaneous modulation of the feed composition and the flow rate with sinusoidal functions was considered in the paper [1] as well. It was pointed out that the maximum improvement in the conversion is obtained when both amplitudes are large, and the phase shift is close to 180°. The optimum design problem is addressed in the book [2] in the sense of steady-state operations, when the goal is to find the values of the design variables maximizing the profitability of the process (e.g., the capital charge factor).

In the paper [10], a CSTR model whose dynamical behavior is governed by a system of two nonlinear ordinary differential equations with one control

variable is considered. For the optimal control problem with periodic boundary conditions, the first variation of the cost function is evaluated with the use of the adjoint system of equations. If this first variation is nonzero for a given periodic process, then the performance can be improved by using "the hill climbing method in function space", described in the paper. Some simulation results, based on this method, are presented to illustrate the possibility of improving an initial steady-state control.

A single non-isothermal reaction is considered in the paper [8] by assuming that the coolant flow-rate is controlled within given bounds. The problem of minimizing a quadratic cost functional is formulated without periodic boundary conditions, and the candidates for optimal controls are chosen as polynomials or bang-bang controls with unknown swiching times. Then an iterative scheme for computing the control parameters is presented based on numerical integration of the state equations and Rosenbrock's method for unconstrained optimization.

The paper [17] exploits the second variation of the cost function in the frequency domain for examples of chemical reactions involving at most two control signals. To simplify the formula for the second variation, the authors assumed that the control variations are sinusoidal functions with one principal frequency. The process response has been also simulated for bang-bang controls with the use of a numerical hill-climbing method, however, no results concerning the computation of switching times have been presented.

The question whether the performance of an isothermal CSTR may be improved by using periodic perturbations of a given steady-state control is studied in the paper [18]. If a steady-state input is in the interior of the set of control values, and if certain conditions on the transfer function of the linearized system hold, then the above steady-state control is not optimal. In this case, sinusoidal perturbations of the input with large frequencies can be used to improve the reactor performance in comparison to its steady-state operation. It is shown that, for the reaction of order $n > 1$, the reactor performance can be improved by fluctuating the feed concentration only.

In the papers [6, 9, 15], an isothermal reaction scheme of the type "$\nu_1 A_1 + \nu_2 A_2 \to product$" with the power law rate $r = k C_1^{n_1} C_2^{n_2}$ is considered under the assumption that the sum of the inlet concentrations of A_1 and A_2 is constant. By applying Hölder's inequality to the integral of the product concentration, a priori estimates of the degree of conversion are obtained for different values of n_1 and n_2 in [6]. In particular, the conversion cannot be improved with respect to the steady-state operation if $0 < n_1 < 1$, $0 < n_2 < 1$, and $n_1 + n_2 \leq 1$. This property is also established in the paper [7] for an isothermal plug-flow tube reactor model by exploiting the convexity of the function that generates the solutions in the method of characteristics. The Legendre–Clebsch condition is used in [15] to check the optimality of steady state operations of a CSTR. In particular, it is shown that the steady-state operation is not optimal if $\nu_1 > 1$ for homogeneous systems. If the inlet concentrations are bang-bang controls with a given period t_f, and if there is only one switching point $t_s \in (0, t_f)$, then t_s is uniquely defined from the corresponding steady-state concentration. Under these assumptions, the domains of conversion improvements and conversion

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deteriorations are constructed in the (n_1, n_2)-plane by using numerical simulations in the paper [9]. It is pointed out that a reaction with a decreasing total number of moles increases the region for conversion improvements.

The problem of maximizing the average production rate of a chemical reaction can also be treated within the economic model predictive control (EMPC) framework. A survey of recent results in this area is presented in [3]. In particular, the EMPC approach is applied for an irreversible second-order exothermic reaction. The mathematical model of this reaction is represented by material and energy balance equations, and the economic measure is defined as an integral functional whose integrand is proportional to the square of the concentration variable with the coefficient depending on the temperature. Such objective function is used to maximize the average production rate over a given finite time horizon assuming that the input signal satisfies an integral constraint. Note that the system trajectory is not required to be periodic for the above problem formulation. Chapter 4 of the monograph [4] deals with applications of Lyapunov's direct method for the EMPC design. A second-order exothermic reaction is considered as an example, and a quadratic Lyapunov function with respect to the deviation of the state from its stationary value is used in the control design. The authors note that a contractive Lyapunov-based constraint may be applied to the EMPC to ensure that the temperature converges to a neighborhood of the optimal stead-state value. The case of a non-isothermal CSTR with three parallel reactions is considered in the book [4] as well, and the economic measure is chosen to penalize energy usage and reactant consumption, credit the production rate of the desired product, and penalize the deviation of the temperature from its median value.

In contrast to the above publications, in this paper we concentrate on an isoperimetric formulation of the optimal control problem with periodic boundary conditions, and we do not aim to penalize the deviation of the state variables from their steady-state values, assuming that large deviations may have the potential to improve the efficiency of the conversion. To the best of our knowledge, neither analytic expressions for the optimal control candidates nor algebraic equations for computing the switching times have been obtained in the literature so far.

3. Modulation of the Inlet Concentration

Let us first consider a homogeneous isothermal reaction of the type "$A \rightarrow$ product" (cf. [12, 14]):

$$\frac{dC_A}{dt} = \frac{F}{V}(C_{Ai} - C_A) - C_A^n k_0 e^{-E_A/(RT)}, \qquad (1)$$

where C_A is the reactant A concentration, C_{Ai} is the inlet concentration of A, t is the time, n is the order of the reaction, V is the reactor volume, F is the volumetric flow-rate of the reaction stream, T is the temperature, k_0 is the preexponential factor in the Arrhenius equation, E_A is the activation energy,

and R is the universal gas constant. The differential equation (1) admits a steady-state solution $C_A(t) = \bar{C}_A > 0$ for $C_{Ai}(t) = \bar{C}_{Ai} = $ const and $F = $ const if

$$\frac{\bar{C}_{Ai}}{\bar{C}_A} = 1 + \frac{V\bar{C}_A^{n-1}}{F}k_0 e^{-E_A/(RT)}. \tag{2}$$

By introducing the dimensionless time $\tau = Ft/V$ and dimensionless variables

$$x = \frac{C_A - \bar{C}_A}{\bar{C}_A}, \quad u = \frac{C_{Ai} - \bar{C}_{Ai}}{\bar{C}_{Ai}}\left(1 + \frac{V\bar{C}_A^{n-1}}{F}k_0 e^{-E_A/(RT)}\right), \tag{3}$$

we rewrite the material balance equation (1) in the following form:

$$\frac{dx}{d\tau} = -x - \alpha\big((x+1)^n - 1\big) + u, \quad \alpha = \frac{V\bar{C}_A^{n-1}}{F}k_0 e^{-E_A/(RT)}, \tag{4}$$

where the parameter α evaluates the ratio of the time given to the reaction (the mean residence time V/F) over the time needed be the reaction (expressed by the rate expression parameters and concentration). It corresponds to the frequently used Damköhler number Da (see, e.g., [5]). If the control parameter u is fixed to some constant value $\bar{u}$, then differential equation (4) admits a steady-state solution $x(\tau) = $ const. In particular,

$$x(\tau) = \frac{\bar{u}}{1 + \alpha} \quad \text{for } n = 1, \tag{5}$$

and

$$x(\tau) = -\frac{2\alpha + 1}{2\alpha} \pm \sqrt{\left(\frac{2\alpha + 1}{2\alpha}\right)^2 + \frac{\bar{u}}{\alpha}} \quad \text{for } n = 2. \tag{6}$$

We are interested in maximizing the conversion of A to the product in comparison to these particular solutions by using time-varying inputs $u(\tau)$. For this purpose we introduce the class of admissible controls, which consists of all measurable functions $u(\tau) \in [u_{min}, u_{max}]$ depending on $\tau \in [0, \tau_f]$, where $\tau_f > 0$ and $u_{min} < u_{max}$ are given numbers. We also assume that the mean value of the control $\bar{u}$ is fixed and formulate the following optimal control problem.

Problem 1. *Given $\tau_f > 0$ and $\bar{u} \in [u_{min}, u_{max}]$, find an admissible control $\hat{u} : [0, \tau_f] \to [u_{min}, u_{max}]$ that minimizes the cost*

$$J = \frac{1}{\tau_f}\int_0^{\tau_f} x(\tau)\, d\tau \tag{7}$$

among all solutions $x(\tau)$ of the differential equation (4) corresponding to the class of admissible controls $u(\tau)$ such that

$$x(0) = x(\tau_f), \quad \frac{1}{\tau_f}\int_0^{\tau_f} u(\tau)\, d\tau = \bar{u}. \tag{8}$$

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To solve this isperimetric optimal control problem, we apply the Pontryagin maximum principle with the Hamiltonian

$$H(p_0, p, x, u, \eta) = p_0 x - p\Big(x + \alpha\big((x+1)^n - 1\big) - u\Big) + \eta(u - \bar{u}\tau_f), \ p_0 \le 0. \quad (9)$$

If $\hat{u}(\tau)$ is an optimal control for the above problem on $\tau \in [0, \tau_f]$ and $\hat{x}(\tau)$ is the corresponding solution of equation (4), then there exist constants $p_0 \le 0$, η, and a continuous function $p : [0, \tau_f] \to \mathbb{R}$, $(p_0, p(\tau), \alpha) \ne 0$, such that $\hat{u}(\tau)$ maximizes the Hamiltonian $H(p_0, p(\tau), \hat{x}(\tau), u, \eta)$ for each $\tau \in [0, \tau_f]$ along the trajectory $(\hat{x}(\tau), p(\tau))$ (see, e.g., [16]). Here $p(\tau)$ satisfies the adjoint equation:

$$\frac{dp}{d\tau} = -\left.\frac{\partial H}{\partial x}\right|_{x=\hat{x}(\tau)} = -p_0 + p\Big(1 + n\alpha(\hat{x}(\tau) + 1)^{n-1}\Big). \quad (10)$$

The general solution of the differential equation (10) is

$$p(\tau) = \left(p(0) - p_0 \int_0^\tau \exp\left\{-\int_0^s \big(1 + n\alpha(1 + \hat{x}(v))^{n-1}\big)\, dv\right\} ds\right) \\ \times \exp\left\{\int_0^\tau \Big(1 + n\alpha\,(1 + \hat{x}(s))^{n-1}\Big) ds\right\}. \quad (11)$$

We see that the maximum of H given by formula (9) is achieved for the following function

$$\hat{u}(\tau) = \begin{cases} u_{max}, & p(\tau) + \eta > 0, \\ u_{min}, & p(\tau) + \eta < 0. \end{cases} \quad (12)$$

Thus, each optimal control $u = \hat{u}(\tau)$ for Problem 1 is bang-bang (piecewise constant) on $\tau \in [0, \tau_f]$. If $p(\tau)$ is monotone, we conclude that each optimal control $\hat{u}(\tau)$ has at maximum one switching on $\tau \in [0, \tau_f]$, i.e. $\hat{u}(\tau)$ may be represented either as

$$\hat{u}(\tau) = \begin{cases} u_{max}, & \tau \le \tau^* = \frac{\tau_f(\bar{u} - u_{min})}{u_{max} - u_{min}}, \\ u_{min}, & \tau > \tau^*, \end{cases} \quad (13)$$

or

$$\hat{u}(\tau) = \begin{cases} u_{min}, & \tau \le \tau^* = \frac{\tau_f(u_{max} - \bar{u})}{u_{max} - u_{min}}, \\ u_{max}, & \tau > \tau^*, \end{cases} \quad (14)$$

Here the switching time $\tau^* \in [0, \tau_f]$ is defined from the isoperimetric condition $\int_0^{\tau_f} \hat{u}(\tau)\, d\tau = \bar{u}\tau_f$.

One can check that the differential equation (4) with $u = \text{const}$ is integrable in elementary functions if $n = -1; 1/2; 1; 2$. It was already reported in the papers [6] and [14] that *no efficiency improvement of the first-order reaction ($n = 1$) can be achieved by using time-varying controls in comparison to the steady-state operation.*

Let us consider now the optimal control problem for the differential equation (4) with $n = 2$:

$$\frac{dx}{d\tau} = -\alpha x^2 - (1 + 2\alpha)x + u. \quad (15)$$

For this case, based on [6], an improvement by using periodic modulations of the input concentration can be expected. Recall that the steady-state solutions of this differential equations with $u(\tau) = \bar{u}$ are given by formula (6). The solutions $x = \hat{x}(\tau)$ corresponding to the control $u = \hat{u}(\tau)$ of the form (13) can be written as follows:

$$\hat{x}(\tau) = \begin{cases} \frac{v_{max} - 2\alpha - 1}{2\alpha} - \frac{v_{max}}{\alpha(1 + C_1 e^{\tau v_{max}})}, v_{max} = \sqrt{(1+2\alpha)^2 + 4\alpha u_{max}}, \tau \leq \tau^*, \\ \frac{v_{min} - 2\alpha - 1}{2\alpha} - \frac{v_{min}}{\alpha(1 + C_2 e^{(\tau - \tau_f)v_{min}})}, v_{min} = \sqrt{(1+2\alpha)^2 + 4\alpha u_{min}}, \tau > \tau^*. \end{cases}$$
(16)

The constants C_1 and C_2 are defined by the conditions $\hat{x}(0) = \hat{x}(\tau_f)$ and $\hat{x}(\tau^* - 0) = \hat{x}(\tau^* + 0)$:

$$\frac{v_{max}}{1 + C_1} - \frac{v_{min}}{1 + C_2} = \frac{v_{max} - v_{min}}{2},$$

$$\frac{v_{max}}{1 + C_1 e^{\tau^* v_{max}}} - \frac{v_{min}}{1 + C_2 e^{(\tau^* - \tau_f)v_{min}}} = \frac{v_{max} - v_{min}}{2}.$$
(17)

The controls (14) can be considered analogous to the case (13) by replacing u_{max} with u_{min}.

In order to illustrate the efficiency of our control design scheme, let us consider the second-order reaction ($n = 2$) represented by equation (1) with the following parameters:

$$\bar{C}_{Ai} = 1 \frac{mol}{m^3} \text{ (mean inlet concentration of reactant } A\text{)};$$

$$k = k_0 e^{-E_A/(RT)} = 10^{-3} \frac{m^3}{s \cdot mol} \text{ (rate constant)};$$

$$\tau_c = \frac{V}{F} = 10^2 \, s \text{ (contact time)}.$$

These parameter values have been used in the paper [14] for the frequency response analysis of nonlinear reactors with a sinusoidal input. The steady-state concentration $\bar{C}_A > 0$ corresponding to the above $\bar{C}_{Ai}$ is obtained from the algebraic equation (2): $\bar{C}_A \approx 0.916 \frac{mol}{m^3}$. If M is the maximum magnitude of the dimensionless input concentration $\frac{C_{Ai} - \bar{C}_{Ai}}{\bar{C}_{Ai}}$, then the control $u \in [u_{min}, u_{max}]$ in formula (3) is constrained by

$$u_{max} = -u_{min} = M(1 + \alpha),$$

where $\alpha = k\tau_c \bar{C}_A \approx 0.0916$. We consider the isoperimetric optimal control problem for the differential equation (15) with the cost (7) and constraints (8) with $\bar{u} = 0$, i.e., we assume that the same average amount of A is used for all admissible inputs as in the steady-state. We analyse candidates for the optimal control $u = \hat{u}(\tau)$ and the optimal trajectory $x = \hat{x}(\tau)$ on $\tau \in [0, \tau_f]$ given by formulas (13) and (16), respectively. We take the dimensionless time period $\tau_f = 2\pi$ and $M = 0.75$, so that $\tau^* = \pi$, $u_{max} \approx 0.8187$, and the parameters C_1, C_2 of the function $\hat{x}(\tau)$ are computed from the system of algebraic equations (17):

$$C_1 \approx 9.5708, \ C_2 \approx -252.87.$$

The time plot of the corresponding solution $\hat{x}(\tau)$ of the differential equation (15) is shown in Fig. 1 together with the horizontal line corresponding to the cost

$$\hat{J} = \frac{1}{\tau_f} \int_0^{\tau_f} \hat{x}(\tau)\, d\tau \approx -0.018.$$

We see that $\hat{J} < 0$, so the proposed control $u = \hat{u}(\tau)$ gives better conversion of A to the product in comparison with the steady-state operation.
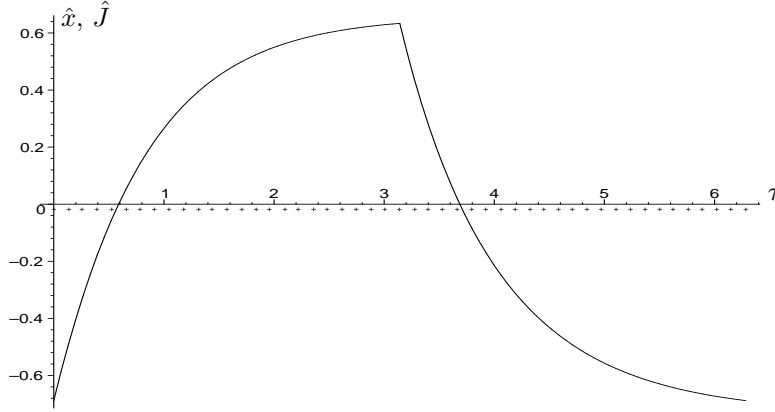

Figure 1: The graph of $\hat{x}(\tau)$ (solid line) and the value of $\hat{J}$ (dotted line).

On the one hand, our reactor performance improvement due to the bang-bang control of form (13) can be expressed in absolute units and percents as follows:

$$\Delta C_1 = \frac{1}{\tau_f} \int_0^{T*} C_A(\tau)\, d\tau - \bar{C}_A = \hat{J}\bar{C}_A \approx -0.0166\, \frac{mol}{m^3}, \ \Delta C_1/\bar{C}_A \approx -1.81\%.$$

As in the paper [14], the minus sign indicates that the mean outlet concentration of A for a time-varying input is less than the steady-state outlet concentration.

On the other hand, the performance improvement with a sinusoidal input of the dimensionless amplitude M and the dimensionless frequency $\omega = 2\pi/\tau_f = 1$ can be estimated by the nonlinear frequency response function (see, e.g., [14]):

$$\Delta C_2 = 2 \left(\frac{M}{2} \right)^2 G_2(\omega, -\omega)\bar{C}_A \approx -0.0099\, \frac{mol}{m^3}, \ \Delta C_2/\bar{C}_A \approx -1.08\%,$$

where $G_2(\omega, -\omega)$ is the asymmetrical second-order frequency response function [14, Formula (14-26)]:

$$G_2(\omega, -\omega) = -\frac{k\tau_c \bar{C}_A (1 + k\tau_c \bar{C}_A)^2}{(1 + 2k\tau_c \bar{C}_A)((1 + 2k\tau_c \bar{C}_A)^2 + \omega^2)}.$$

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As ΔC_1 and ΔC_2 are negative, we conclude that both bang-bang and sinu-soidal periodic controls can be used to improve the performance of the reactor in comparison with its steady-state operation. However, the inequality $\Delta C_1 < \Delta C_2$ indicates that the bang-bang strategy results in better performance for the case considered.

4. Non-Isothermal Reaction

In this section, we assume that a non-isothermal reaction of the type "$A \rightarrow$ product" is characterized by the steady-state inlet concentration of the reactant $\bar{C}_{Ai}$ and the steady-state outlet concentration $\bar{C}_A$ for some constant temperature in the reactor $\bar{T}$ and constant volumetric flow-rate $\bar{F}$. In the dimensionless variables

$$x_1(\tau) = \frac{C_A - \bar{C}_A}{\bar{C}_A}, \quad x_2(\tau) = \frac{T - \bar{T}}{\bar{T}}, \quad \tau = Ft/V, \tag{18}$$

such a reaction of order n is governed by the following differential equations [12, 13]:

$$dx_1/d\tau = k_1 e^{-\varkappa} - \phi_1 x_1 - k_1 (x_1 + 1)^n e^{-\varkappa/(x_2+1)} + u_1,$$
$$dx_2/d\tau = k_2 e^{-\varkappa} - \phi_2 x_2 - k_2 (x_1 + 1)^n e^{-\varkappa/(x_2+1)} + u_2, \tag{19}$$

where $x_1(\tau)$ is the dimensionless outlet concentration of A, $x_2(\tau)$ is the di-mensionless temperature in the reactor, the input $u_1(\tau)$ is applied to control the inlet concentration of A (and/or the flow-rate), and $u_2(\tau)$ corresponds to the temperature of the inlet stream. The dimensionless control variables and parameters of system (19) can be expressed as follows:

$$u_1(\tau) = k_1 \Phi e^{-\varkappa} + (1 + k_1 e^{-\varkappa})(1 + \Phi) \frac{C_{Ai}(\tau) - \bar{C}_{Ai}}{\bar{C}_{Ai}},$$
$$u_2(\tau) = \beta \Phi + (1 + \beta)(1 + \Phi) \frac{T_i(\tau) - \bar{T}_i}{\bar{T}_i}, \tag{20}$$

$$k_1 = k_0 \bar{C}_A^{n-1} \frac{V}{\bar{F}}, \; k_2 = \frac{\Delta H_R k_0 \bar{C}_A^n V}{\rho c_p \bar{T} \bar{F}}, \; \varkappa = \frac{E_A}{R\bar{T}}, \; \Phi = \frac{F - \bar{F}}{\bar{F}},$$
$$\phi_1 = \Phi + 1, \; \phi_2 = \phi_1 + \frac{U_0 A_w}{\rho c_p \bar{F}}, \; \beta = k_2 e^{-\varkappa} + \frac{U_0 A_w (\bar{T} - T_j)}{\rho c_p \bar{F} \bar{T}}. \tag{21}$$

Here $T_i(\tau)$ is the temperature of the inlet stream (and $\bar{T}_i$ is its steady-state value), T_j is the temperature of the heating/cooling fluid, $C_{Ai}(\tau)$ is the inlet concentration of A, F is the volumetric flow-rate of the reaction stream, ΔH_R is the reaction heat, U_0 is the overall heat transfer coefficient, A_w is the surface area for heat exchange, ρ is the density, and c_p is the heat capacity.

System (19) has an equilibrium $x_1 = x_2 = 0$ with $u_1 = u_2 = 0$ that corre-sponds to the steady state of the reactor.

4.1. Pontryagin Maximum Principle

For a given time horizon $\tau_f > 0$, we assume that the class of admissible controls $\mathcal{U}_{\tau_f}$ for system (19) consists of all measurable functions $u : [0, \tau_f] \to \mathbb{R}^2$ such that $u_i^{min} \leq u_i(\tau) \leq u_i^{max}$ for $\tau \in [0, \tau_f]$, $i = 1, 2$. Our goal is to maximize the productivity of the reactor, which means the minimization of the mean value of $x_1(\tau)$ over the period $\tau \in [0, \tau_f]$ by using the admissible controls with a fixed mean value of $u_1(\tau)$. To be more precise, we consider the following isoperimetric optimal control problem in this section.

Problem 2. *Given $\tau_f > 0$, $\bar{u}_1$, x_i^0, u_i^{min}, u_i^{max}, $i = 1, 2$, the goal is to find a control $\hat{u}(\cdot) \in \mathcal{U}_{\tau_f}$ that minimizes the cost*

$$J = \frac{1}{\tau_f} \int_0^{\tau_f} x_1(\tau) \, d\tau$$

along the solutions $(x_1(\tau), x_2(\tau))$ of system (19) corresponding to the admissible controls $u(\cdot) \in \mathcal{U}_{\tau_f}$ such that

$$\frac{1}{\tau_f} \int_0^{\tau_f} u_1(\tau) \, d\tau = \bar{u}_1, \ x_i(0) = x_i(\tau_f) = x_i^0, \quad i = 1, 2. \tag{22}$$

According to the Pontryagin maximum principle for problems with isoperimetric constraints [16, Theorem 4.1], if $\hat{u}(\tau)$ is an optimal control on $\tau \in [0, \tau_f]$ for Problem 2 and $\hat{x}(\tau)$ is the corresponding solution of system (19), then there exist constants $p_0 \leq 0$, η_1, and continuous functions $p_i : [0, \tau_f] \to \mathbb{R}$, $i = 1, 2$, such that $(p_0, p_1(\tau), p_2(\tau), \eta_1) \neq 0$ and $\hat{u}(\tau)$ maximizes the Hamiltonian

$$H(x, u, p_0, p, \eta_1) = p_0 x_1 + p_1 \left(k_1 e^{-\varkappa} - \phi_1 x_1 - k_1 (x_1 + 1)^n e^{-\varkappa/(x_2+1)} + u_1 \right)$$

$$+ p_2 \left(k_2 e^{-\varkappa} - \phi_2 x_2 - k_2 (x_1 + 1)^n e^{-\varkappa/(x_2+1)} + u_2 \right) + \eta_1 (u_1 - \bar{u}_1 \tau_f) \tag{23}$$

for each $\tau \in [0, \tau_f]$ along the trajectory $(\hat{x}(\tau), p(\tau))$. Here, the $p_i(\tau)$ satisfy the adjoint equations:

$$dp_1/d\tau = -\frac{\partial H}{\partial x_1} = -p_0 + \phi_1 p_1 + n(k_1 p_1 + k_2 p_2)(x_1 + 1)^{n-1} e^{-\varkappa/(x_2+1)},$$

$$dp_2/d\tau = -\frac{\partial H}{\partial x_2} = \phi_2 p_2 + \frac{\varkappa(k_1 p_1 + k_2 p_2)(x_1 + 1)^n}{(x_2 + 1)^2} e^{-\varkappa/(x_2+1)}, \ p_0 \leq 0.$$

By computing the pointwise maximum of H with respect to $u_i \in [u_i^{min}, u_i^{max}]$ in formula (23), we conclude that the optimal controls are bang-bang:

$$\hat{u}_1(\tau) = \frac{u_1^{max} + u_1^{min}}{2} + \frac{u_1^{max} - u_1^{min}}{2} \text{sign}\,(p_1(\tau) + \eta_1),$$

$$\hat{u}_2(\tau) = \frac{u_2^{max} + u_2^{min}}{2} + \frac{u_2^{max} - u_2^{min}}{2} \text{sign}\, p_2(\tau), \quad \tau \in [0, \tau_f]. \tag{24}$$

4.2. Control of the Linearized System

Let us linearize the differential equations (19) in a neighborhood of the equilibrium point $x_1 = x_2 = 0$:

$$\frac{dx}{d\tau} = \tilde{A}x + u, \quad x = \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} \in \mathbb{R}^2, \ u = \begin{pmatrix} u_1 \\ u_2 \end{pmatrix} \in [u_1^{min}, u_1^{max}] \times [u_2^{min}, u_2^{max}], \quad (25)$$

where

$$\tilde{A} = \begin{pmatrix} -\phi_1 - n\tilde{k}_1 & -\varkappa\tilde{k}_1 \\ -n\tilde{k}_2 & -\phi_2 - \varkappa\tilde{k}_2 \end{pmatrix}, \quad \tilde{k}_1 = k_1 e^{-\varkappa}, \ \tilde{k}_2 = k_2 e^{-\varkappa}.$$

We will assume that the matrix $\tilde{A}$ is nonsingular for the sequel.

Then the adjoint equations corresponding to Problem 2 for system (25) take the following form:

$$\frac{dp(\tau)}{d\tau} = -\tilde{A}^* p(\tau) - \begin{pmatrix} p_0 \\ 0 \end{pmatrix}, \quad p(\tau) = \begin{pmatrix} p_1(\tau) \\ p_2(\tau) \end{pmatrix} \in \mathbb{R}^2. \quad (26)$$

Here the asterisk denotes the transpose. As it has been shown in the previous subsection, each optimal control $\hat{u}(\tau)$ can be represented in the form (24), where $p_1(\tau)$ and $p_2(\tau)$ are the components of a solution to system (26):

$$\begin{pmatrix} p_1(\tau) \\ p_2(\tau) \end{pmatrix} = e^{-\tau\tilde{A}^*}\bar{p} - \left(\tilde{A}^*\right)^{-1}\begin{pmatrix} p_0 \\ 0 \end{pmatrix}, \quad (27)$$

with some vector $\bar{p} \in \mathbb{R}^2$.

Proposition 1. *Let*

$$D = (\phi_1 + \phi_2 + n\tilde{k}_1 + \varkappa\tilde{k}_2)^2 - 4(\phi_1\phi_2 + \phi_1\varkappa\tilde{k}_2 + n\phi_2\tilde{k}_1) > 0. \quad (28)$$

Then each control $\hat{u}_i(\tau)$ of the form (24) with $p_i(\tau)$ given by (27), $i = 1, 2$, has at maximum 2 switchings in the interval $[0, \tau_f]$.

Proof. If condition (28) is satisfied, then the matrix $-\tilde{A}^*$ has two distinct real eigenvalues:

$$\lambda_{1,2} = \frac{\phi_1 + \phi_2 + n\tilde{k}_1 + \varkappa\tilde{k}_2}{2} \pm \frac{\sqrt{D}}{2}.$$

Thus, there exists an invertible 2×2-matrix P such that

$$-\tilde{A}^* = P \begin{pmatrix} \lambda_1 & 0 \\ 0 & \lambda_2 \end{pmatrix} P^{-1},$$

and formula (27) can be rewritten as

$$\begin{aligned}
p_1(\tau) &= c_1 P_{11} e^{\lambda_1 \tau} + c_2 P_{12} e^{\lambda_2 \tau} - \left(\tilde{A}^*\right)^{-1}_{11} p_0, \\
p_2(\tau) &= c_1 P_{21} e^{\lambda_1 \tau} + c_2 P_{22} e^{\lambda_2 \tau} - \left(\tilde{A}^*\right)^{-1}_{21} p_0, \quad c_1, c_2 = \text{const.}
\end{aligned} \quad (29)$$

Here P_{ij} and $\left(\tilde{A}^*\right)^{-1}_{ij}$ denote the components of P and $\left(\tilde{A}^*\right)^{-1}$, respectively.

If $p_i(\tau) = const$, then the control $\hat{u}_i(\tau)$ in (24) has no switchings in $\tau[0, \tau_f]$, otherwise the switching times τ are defined by the equation

$$p_i(\tau) = -\eta_i, \tag{30}$$

where we put $\eta_2 = 0$ for the convenience of notation, $i = 1, 2$. We rewrite equation (30) with respect to the new variable $y = e^{\lambda_1 \tau} > 0$ by using (29) as follows:

$$F(y) \equiv y + cy^\mu = q, \tag{31}$$

where c, q, and $\mu = \lambda_2/\lambda_1 \neq 1$ are real parameters. Without loss of generality, we have assumed here that $c_1 P_{i1} \neq 0$, since otherwise we may replace λ_1 with λ_2. The function $F(y)$ has at most one critical point $y^* > 0$ defined by the equation $F'(y^*) = 1 + c\mu y^{*\mu-1}$. Thus, the function $F(y)$ is either strictly monotone on the segment $\mathcal{I}$ with the endpoints $y_1 = 1$ and $y_2 = e^{\lambda_1 \tau_f}$, or there is a unique extremum point y^* in $\mathcal{I}$. These arguments imply that equation (31) has at most 2 solutions on $\mathcal{I}$, which completes the proof of Proposition 1.

We see that each of the two controls $\hat{u}_1(\tau)$ and $\hat{u}_2(\tau)$ has at maximum 2 switchings if the conditions of Proposition 1 are satisfied, hence, the corresponding vector function $\hat{u}(\tau) \in \mathbb{R}^2$ has no more than 4 switchings.

To find the candidates for solutions to Problem 2, we introduce a family of bang-bang controls $\tilde{u}(\tau)$ with N switchings in $[0, \tau_f]$, $0 \leq N \leq 4$. Let

$$0 = \tau_0 < \tau_1 < ... < \tau_{N+1} = \tau_f,$$

and let

$$\tilde{u}(\tau) = u^j \quad \text{for } \tau \in [\tau_j, \tau_{j+1}), \tag{32}$$

where

$$u^j \in \left\{ \begin{pmatrix} u_1^{min} \\ u_2^{min} \end{pmatrix}, \begin{pmatrix} u_1^{min} \\ u_2^{max} \end{pmatrix}, \begin{pmatrix} u_1^{max} \\ u_2^{min} \end{pmatrix}, \begin{pmatrix} u_1^{max} \\ u_2^{max} \end{pmatrix} \right\} = U,$$

for each $j = 0, 1, ..., N$. The solution $x(\tau)$ of system (25) corresponding to the inital condition $x(0) = x^0$ and control $\tilde{u}(\tau)$ can be represented as follows:

$$x(\tau) = e^{(\tau-\tau_j)\tilde{A}} x(\tau_j) + e^{(\tau-\tau_j)\tilde{A}} \int_0^{\tau-\tau_j} e^{-s\tilde{A}} ds\, u^j$$

$$= e^{(\tau-\tau_j)\tilde{A}} x(\tau_j) + \tilde{A}^{-1} \left(e^{(\tau-\tau_j)\tilde{A}} - I \right) u^j, \quad \tau \in [\tau_j, \tau_{j+1}], \; j = 0, 1, ..., N.$$

This formula implies that

$$x(\tau_f) = e^{\tau_f \tilde{A}} x^0 + \tilde{A}^{-1} \sum_{j=0}^N e^{(\tau_f - \tau_{j+1})\tilde{A}} \left(e^{(\tau_{j+1} - \tau_j)\tilde{A}} - I \right) u^j.$$

The periodic boundary condition $x(0) = x(\tau_f)$ is thus reduced to the following:

$$\sum_{j=0}^N \left(e^{\Delta_j \tilde{A}} - I \right) \left(\prod_{i=j+1}^N e^{\Delta_i \tilde{A}} \right) u^j = \tilde{A} \left(I - e^{\tau_f \tilde{A}} \right) x^0, \tag{33}$$

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and the isoperimetric constraint in (22) takes the form

$$\sum_{j=0}^{N} \Delta_j u_1^j = \tau_f \bar{u}_1, \quad \sum_{j=0}^{N} \Delta_j = \tau_f, \tag{34}$$

where $\Delta_j = \tau_{j+1} - \tau_j > 0$, and $u_1^j \in \{u_1^{min}, u_1^{max}\}$ denotes the first coordinate of u^j, $j = 0, 1, ..., N$. In the scalar form, the relations (33) and (34) represent a system of 4 equations with respect to $N+1$ unknowns $(\Delta_0, \Delta_1, ..., \Delta_N)$, provided that $\tau_f > 0$, $x^0 \in \mathbb{R}^2$, $\bar{u}_1 \in \mathbb{R}^1$, and $(u^0, u^1, ..., u^N) \in U^{N+1}$ are given.

We will use the formula

$$e^{\Delta_j \tilde{A}} = I + \Delta_j \tilde{A} + \frac{\Delta_j^2}{2} \tilde{A}^2 + O(\Delta_j^3)$$

to introduce a polynomial approximation of condition (33) for small Δ_j:

$$\sum_{j=0}^{N} \Delta_j \left(I + \frac{\Delta_j}{2} \tilde{A} \right) \prod_{i=j+1}^{N} \left(I + \Delta_i \tilde{A} + \frac{\Delta_i^2}{2} \tilde{A}^2 \right) u^j = -\tau_f \tilde{A} \left(I + \frac{\tau_f}{2} \tilde{A} \right) x^0. \tag{35}$$

If, for given $\tau_f > 0$, $x^0 \in \mathbb{R}^2$, $\bar{u}_1 \in \mathbb{R}^1$, $(u^0, u^1, ..., u^N) \in U^{N+1}$, the system of algebraic equations (34) and (35) has a solution $(\Delta_0, \Delta_1, ..., \Delta_N)$ with positive components, then *the piecewise constant function $u = \tilde{u}(\tau)$ given by (32) with $\tau_0 = 0$ and $\tau_j = \sum_{i=0}^{j-1} \Delta_i$, $j = 1, 2, ..., N+1$, is considered as an approximation of a possible optimal control for Problem 2 (with N switchings in the linearized case).* The switching times $\{\tau_j\}$ may be defined exactly by solving equation (33) instead of (35). As it follows from the above considerations, each optimal control (in the sense of Problem 2) can be obtained for system (25) by using this procedure with $N \leq 4$, if the conditions of Proposition 1 are satisfied.

5. Simulation Results

As an example, we consider the hydrolysis reaction of acetic acid anhydride to acetic acid:

$$(CH_3CO)_2O + H_2O \rightarrow 2\,CH_3COOH.$$
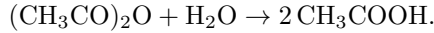
We assume the following realistic parameters for this example, cf. [12]:

$$k_0 = 139390\, s^{-1},\ E_A = 44.35\, \frac{kJ}{mol},$$

$$\Delta H_R = -55.5\, \frac{kJ}{mol},\ \rho c_p = 4.186\, \frac{kJ}{K \cdot l},\ R = 8.3144598\, \frac{J}{K \cdot mol},$$

$$V = 0.384\, l,\ \bar{F} = 0.00175\, \frac{l}{s},$$

$$\bar{C}_A = 0.8662\, \frac{mol}{l},\ \bar{C}_{Ai} = 3.640\, \frac{mol}{l},$$

$$\bar{T} = 331.93\, K,\ \bar{T}_i = 295.15\, K.$$

We take the order $n = 1$, for which no improvement is achievable by means of periodic input modulations in the isothermal case. The above values are used to compute the dimensionless parameters by (21) for the non-isothermal adiabatic case ($A_w = 0$) with a constant flow-rate ($F = \bar{F}$) as follows:

$$
\Phi = 0, \ \phi_1 = \phi_2 = 1, \ \varkappa \approx 16.07, \ k_1 \approx 3.059 \cdot 10^7, \ k_2 \approx -1.058 \cdot 10^6,
$$
$$
k_1 e^{-\varkappa} \approx 3.21, \ k_2 e^{-\varkappa} \approx -0.111. \tag{36}
$$

We assume that the reaction is controlled by both the inlet concentration $C_i(\tau) \in [\bar{C}_A - \Delta C_{Ai}, \bar{C}_A + \Delta C_{Ai}]$ and the inlet temperature $T_i(\tau) \in [\bar{T}_i - \Delta T_i, \bar{T}_i + \Delta T_i]$, where we take $\Delta C_{Ai} = \bar{C}_A$ and $\Delta T_i = 20 \, K$ for the simulation. This implies the following constraints on the controls $u_1(\tau)$ and $u_2(\tau)$ by using (20):

$$
u_1^{max} = -u_1^{min} = \frac{(1 + k_1 e^{-\varkappa}) \Delta C_{Ai}}{\bar{C}_A} \approx 4.21,
$$
$$
u_2^{max} = -u_2^{min} = \frac{(1 + k_2 e^{-\varkappa}) \Delta T_i}{\bar{T}_i} \approx 0.06. \tag{37}
$$

We see that the condition (28) of Proposition 1 holds for the linearized system (25):

$$
D \approx 2.03 > 0.
$$

In order to illustrate the possibility of improving the performance of the nonlinear reaction governed by equations (19), we apply a control $u = \tilde{u}(\tau)$ of the form (32) with $N = 2$ switchings for $x^0 = 0$, $\tau_f = 0.628$, $\bar{u}_1 = 0.107$, and

$$
u^0 = u^2 = \begin{pmatrix} u_1^{max} \\ u_2^{max} \end{pmatrix}, \ u^1 = \begin{pmatrix} u_1^{min} \\ u_2^{min} \end{pmatrix}. \tag{38}
$$

Note that our choice of the time horizon τ_f corresponds to the dimensionless frequency $\omega = 2\pi/\tau_f \approx 10$. To define the switching times τ_1 and τ_2, we first compute their initial approximations τ_1^* and τ_2^* for the linearized problem by solving equations (33) and (34):

$$
\tau_1^* = \Delta_0 \approx 0.204, \ \tau_2^* = \tau_1^* + \Delta_1 \approx 0.51. \tag{39}
$$

Then the constraints (22) are satisfied for the solution $x(\tau)$ of the nonlinear system (19) if the parameters in (32) are chosen as follows:

$$
\tau_1 \approx 0.2, \ \tau_2 \approx 0.506. \tag{40}
$$

We have computed these values by solving the equation $x(\tau_f) = x(0) = x^0$ with respect to (τ_1, τ_2) in a neighborhood of (τ_1^*, τ_2^*) numerically by using the `fsolve` function in `Maple`.
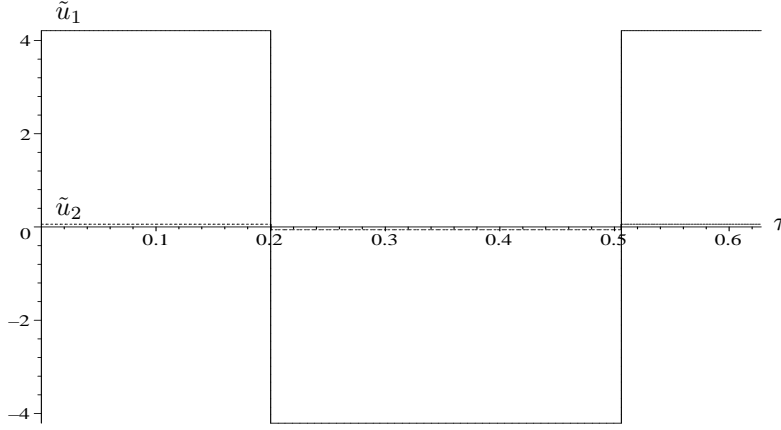
Figure 2: Bang-bang controls $\tilde{u}_1(\tau)$ (solid line) and $\tilde{u}_2(\tau)$ (dashed line) for system (19).
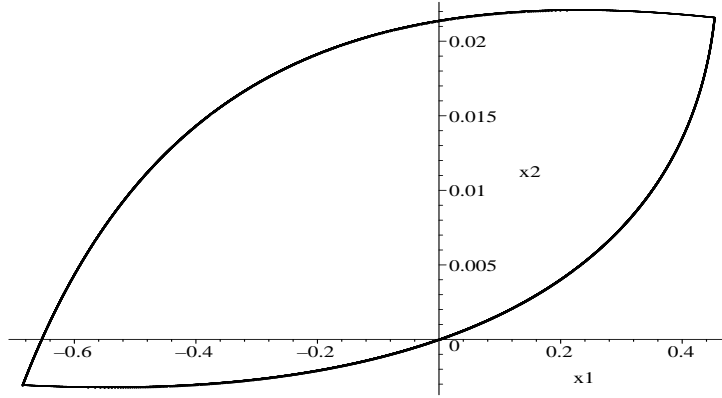

Figure 3: Trajectory of nonlinear system (19) for $x(0) = x^0$ and $u = \tilde{u}(\tau)$.

The control $\tilde{u}(\tau)$ defined by formula (32) and the corresponding trajectory $x(\tau)$ of the nonlinear system (19) is shown in Fig. 2 and Fig. 3, respectively. As we observe in Fig. 2, the chosen control scenario corresponds to the simultaneous swiching of $\tilde{u}_1(t)$ and $\tilde{u}_2(t)$ at $t = \tau_1$ and $t = \tau_2$. The mean value of the control $\tilde{u}_2(\tau)$ is $\bar{u}_2 = \frac{1}{\tau_f} \int_0^{\tau_f} \tilde{u}_2(\tau)d\tau \approx 0.00153$. The control system (19) admits the following equilibrium state for $(u_1, u_2) = (\bar{u}_1, \bar{u}_2)$: $\bar{x}_1 \approx 0.0649$, $\bar{x}_2 \approx 0.0075$. We compute the cost corresponding to the control $u = \tilde{u}(\tau)$:

$$J = \frac{1}{\tau_f} \int_0^{\tau_f} x_1(\tau)d\tau \approx -0.10672.$$

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Number of switchings	Mean controls $(\bar{u}_1, \bar{u}_2)$	Steady-state $(\bar{x}_1, \bar{x}_2)$	Cost $J = \frac{1}{\tau_f} \int_0^{\tau_f} x_1(\tau)d\tau$
$N = 2$	$(0.107, 0.0015)$	$(0.0649, 0.0075)$	$-0.1067 < \bar{x}_1$
$N = 3$	$(0.560, -0.012)$	$(-0.0445, 0.0077)$	$-0.2252 > \bar{x}_1$
$N = 4$	$(-0.312, 0.0082)$	$(-0.0707, -0.0004)$	$-0.0698 > \bar{x}_1$

Table 1: Simulation results for system (19) with bang-bang controls (32).

To compare this result to the bang-bang controls with a higher number of switchings N, we present trajectories of system (19) with $N = 3$ (Fig. 4) and $N = 4$ (Fig. 5). The numerical data for these simulations are summarized in Table 1. All the computations are performed by using the fourth order Runge–Kutta method with the integration step size of $3 \cdot 10^{-5}$ to numerically solve system (19) with controls of the form (32).

As $J < \bar{x}_1$ for the case $N = 2$, we conclude that bang-bang inputs (32) ensure better performance of the conversion of acetic acid anhydride to acetic acid in comparison to the steady-state operation with the same mean values of the controls $(\bar{u}_1, \bar{u}_2)$. To estimate this improvement in absolute units, we observe that the dimensionless values $\bar{x}_1$ and J correspond to the following outlet concentrations of A by formulas (18):

$$C_{\bar{x}_1} = (1 + \bar{x}_1)\bar{C}_A \approx 0.8099 \, \frac{mol}{l}, \ C_J = (1 + J)\bar{C}_A \approx 0.7737 \, \frac{mol}{l}.$$

Thus, in the non-isothermal adiabatic case imposed to two inlet perturbations, the absolute improvement with respect to the steady-state operation of the first-order reaction is

$$\Delta C = C_J - C_{\bar{x}_1} \approx -0.0362 \, \frac{mol}{l},$$

and the relative improvement is

$$\Delta C / C_{\bar{x}_1} \approx -4.47\% \tag{41}$$

for the control with $N = 2$ switchings. This result can be compared with the response of system (19) for two trigonometric inputs of the same frequency and zero phase shift:

$$u_i^\sharp(\tau) = \bar{u}_i + \frac{u_i^{max} - u_i^{min}}{2} \cos(\omega\tau), \quad \tau \in [0, \tau_f], \ \omega = 2\pi/\tau_f, \ i = 1, 2. \tag{42}$$

The cost functional evaluated along the solution $x^\sharp(t) = (x_1^\sharp(t), x_2^\sharp(t))$ of system (19) with controls (42) and the initial data $x^\sharp(0) = x^0$ is

$$J^\sharp = \frac{1}{\tau_f} \int_0^{\tau_f} x_1^\sharp(\tau)d\tau \approx -0.0842.$$

The above cost corresponds to the absolute improvement of the performance

$$\Delta C^\sharp = (1 + J^\sharp)\bar{C}_A - C_{\bar{x}_1} \approx -0.0166 \frac{mol}{l},$$

and the relative improvement with respect to the steady-state operation by using the trigonometric inputs is

$$\Delta C^\sharp / C_{\bar{x}_1} \approx -2.05\%. \tag{43}$$

In this case, as $\Delta C < \Delta C^\sharp$ in formulas (41) and (43), the bang-bang controls ensure better performance than the sinusoidal controls of the form (42) for the first-order reaction. It should be emphasized that the periodic boundary condition $x(0) = x(\tau_f)$ does not hold for sinusoidal inputs in general, while our control design scheme guarantees that the conditions (22) of Problem 2 are satisfied.
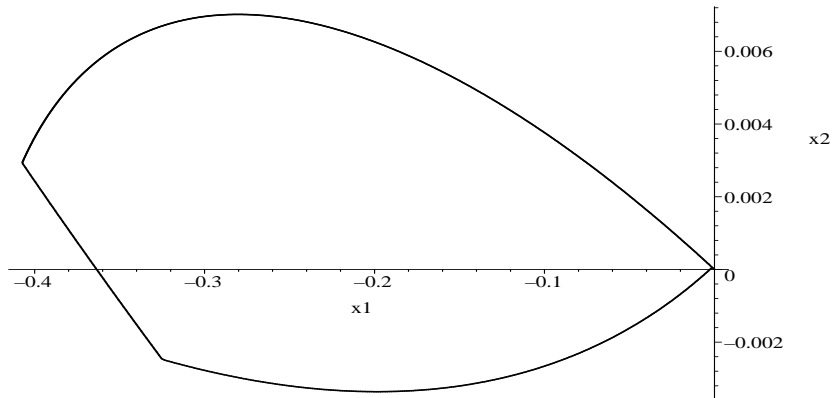

Figure 4: Trajectory $x(\tau)$ with $N = 3$ switchings.

Note that a higher number of switchings $N > 2$ in controls of the form (32) does not necessarily lead to the improvement of the performance, according to Table 1. The cost J evaluated along each of the trajectories, shown in Fig. 4 and Fig. 5, is larger than the corresponding steady-state value.

A crucial point of our control design is that the trajectories of the continuous-time nonlinear system (19) satisfy the periodic boundary conditions, which is proved rigorously by the derivation of equations (33)–(34) and illustrated by
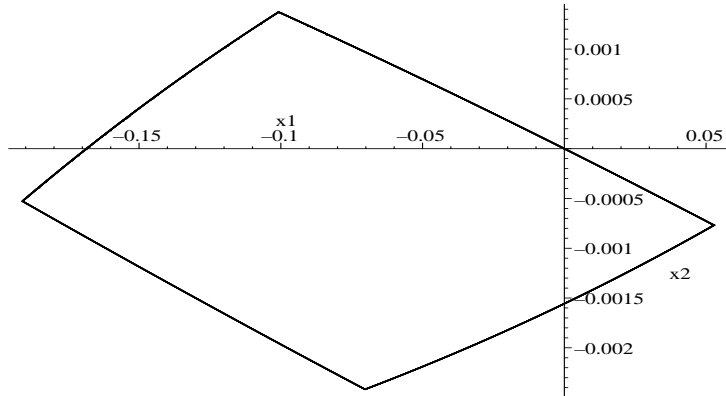
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Figure 5: Trajectory $x(\tau)$ with $N = 4$ switchings.

Figs. 4–5. It should be noted that we do not use any kind of sampling or optimization software packages in comparison to model predictive control approaches.

6. Conclusions

In this paper, we have proposed a control design scheme for maximizing the performance of a non-isothermal CSTR imposed to two forced inlet modulations over a given period of time τ_f. In contrast to the publications dealing with nonlinear frequency response functions, we apply bang-bang controls resulting from the Pontryagin maximum principle with the periodic boundary conditions $x(0) = x(\tau_f)$. The controls and the solution $x(\tau)$ can be extended periodically on each segment $\tau \in [k\tau_f, (k+1)\tau_f]$, $k = 1, 2, \ldots$, so that the moving average values $\frac{1}{\tau_f} \int_\tau^{\tau + \tau_f} x_i(s)\,ds$ do not depend on τ. The basic feature of our construction relies on the computation of the switching times from equations (33) and (34). As the example in Section 5 shows, the switching times (39) for the linearized control system (25) may be considered as a good approximation of the switching times (40) for system (19). The sensitivity of our control design scheme to uncertainties still remains to be analyzed. This question is closely related to the stability of periodic solutions $x(\tau)$ to the open-loop system (19) with controls (32). The stability analysis for this class of nonlinear time-varying systems with discontinuous right-hand sides (see, e.g., [19, 20]) appears to be

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more challenging than just a question of stability for the equilibrium $x = 0$ with $u = 0$.

We do not consider the robustness issues and the question of optimizing the period τ_f (or, equivalently, the dimensionless frequency $\omega = 2\pi/\tau_f$) in this paper, leaving these problems for future work.

Acknowledgements

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