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Suppression of chaos in a multicomponent chemical reaction via Optimal Linear Control*

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ABSTRACT: In this study, we address the problem of controlling chaotic behavior in chemical reactions involving four components occurring in a continuously stirred tank reactor. The controller developed in this study is based on the theory of optimal linear control, proposing a function to maximize the chemical reaction rate and suppress the chaotic behavior in the system. The numerical experiments performed demonstrated the ability to stabilize the chaotic behavior of the chemical reactions in this four-variable system. The chaotic oscillatory motion of the mass concentrations in the multicomponent system was driven towards a stable point. The control technique proved to be efficient for this problem and has the potential for application in chemical reactions involving multiple components.

Key Words: Optimal linear control; chemical reactions; chaos suppression.

Contents

1. Introduction

Reactors play a central role in numerous chemical processes in various industries, including petrochemical plants, the pharmaceutical industry, polymer synthesis, and fertilizer production. The design of a reactor entails calculation mass balance between reactants and products, understanding reaction kinetics, considering temperature, pressure, and other critical factors to achieve cost reduction and optimize modern industrial processes [1]. Recente advances in mathematical modeling, computation, and control theory have facilitated the development of more sophisticated kinetic models, enabling the construction and operation of highly efficient reactors [2].

Chemical reactions frequently exhibit characteristics of nonlinear dynamical systems due to the time-varying nature of the variables involved, with component rates often displaying irregular variations throughout the process. Additionally, experimental evidence supports the existence of chaotic behavior in chemical reaction systems, particularly in reversible and autocatalytic processes involving reactive intermediates and products [3]. This behavior is analogous to that observed in mechanical systems [4]. As a result, the development of control and stabilization systems for these processes becomes crucial in the context of chemical reactors.

The stabilization of nonlinear systems exhibiting complex or chaotic behavior has been the subject of analysis in recent years. Specifically, the use of nonlinear controllers to regulate chemical reactors is a classical task for process engineers. Reactor control has been successfully achieved using input-output linearizers [5], predictive controllers [6], adaptive controllers [7], fuzzy controllers [8], linear PID controllers [9], neural networks, and optimal controllers [10].

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A widely employed control strategy pertains to the theory of optimal control, considering either external constraint-free scenarios. The enhancement of reactor operation and the requirement to implement optimal operating trajectories, characterized by maximum productivity and reduced operating costs, have given rise to the trajectory tracking problem [13], in which optimal control approaches have demonstrated success [11]. In the optimal control approach, Hamiltonian techniques have been applied to nonlinear processes [12], requiring the development of equations to incorporate nonlinear constraints and formulate an appropriate objective function. To obtain a control for the desired task, optimal control theory commonly employs Pontryagin's maximum (or minimum) principle to find the most favorable control strategy [14]. This approach presents a viable strategy for transitioning a dynamical system from one state to another, particularly when constraints on state variables and input controls are present.

Control designs encompass strategies aimed at stabilizing an unstable system at an equilibrium point, a periodic orbit, or, more broadly, enabling the convergence of trajectories towards stable equilibrium points. In recent years, significant interest has been observed in the control of nonlinear systems exhibiting unstable behavior, leading to the discussion of numerous techniques. Among the feedback control strategies, one of the most popular is the Ott-Grebogi-York (OGY) method, introduced in the 1990s by Ott, Grebogi, and York [15]. The OGY method is based on the principle that small perturbations can exert a significant impact on the chaotic dynamics of a system. The central idea involves introducing a small controlled perturbation, through a feedback signal, to stabilize or control the desired chaotic orbit. Unstable periodic motions are identified by examining nearby points between two successive iterations in the Poincaré map of the system.

Another method for stabilizing chaotic systems was proposed by Sinha et al. [16], which utilizes the Lyapunov Floquet transform. This method allows the chaotic motion to be directed to a desired periodic trajectory or fixed point by linearizing the equations describing the error between the actual and desired trajectories. Another technique, proposed by Rafikov and Balthazar [17], aims at finding conditions that ensure the application of linear control in nonlinear systems. In this approach, control and synchronization of chaotic systems are achieved through the utilization of a linear feedback controller. The proposal explicitly outlines a methodology for minimizing the functional for the Hamilton-Jacobi-Bellman equation using a suitable Lyapunov function.

There are two types of problems in control theory. In the first, the control function $u(t)$ is determined to be a function of time. In other words, the optimal control function defines an optimal trajectory corresponding to a specific initial condition of the system. In the second type, the control function $u(t, x)$ depends on both time and state variables. This type of control is referred to as feedback control or control synthesis and can be applied to any initial condition. When the system variables deviate from the desired regime, the optimal control stabilizes around the desired trajectory and minimizes the function that represents the squared deviations from that trajectory. This control technique has been successfully applied in various areas, including transportation systems [18], nonlinear vibration control [19], and others [20].

The objective of this study is to propose the application of optimal linear control to stabilize and control the chaotic behavior of chemical reactions occurring in a continuously stirred tank reactor in a four-component system.

2. Dynamic model

Analysis of chemical reactions in a reactor is essential for determining steady-state multiplicity, output instabilities, and other behaviors present in the desired process. To select optimal operating ranges that maximize reactor productivity, process safety, and operating costs while considering thermodynamic and kinetic constraints, it is often necessary to employ a closed system that maintains variables at selected set points. The operation of a continuous stirred tank reactor (CSTR), depicted in Figure 1, involves a multicomponent input stream F that supplies the chemical reactions within the reactor and a multicomponent output stream comprising a set of state variables (X_i). These variables can be measured to generate control actions u, which, in this case, operate the control valve C.

In Figure 1, when the controller u compares the concentrations of the multicomponent at the output with a predetermined value and actuates the control valve C to make adjustments at the input, a closed system, also known as a feedback system, is formed. The primary function of the control valve C is to

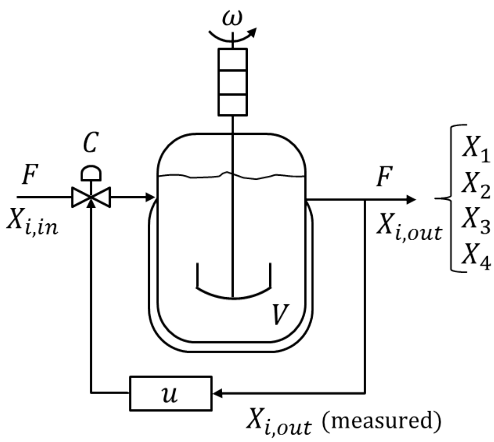

Figure 1: Model of a continuous stirred tank reactor with feedback control. Source: Authors' own elaboration.

compensate the residence time of the reactants in the chemical reactor by manipulating the input flow rate to achieve the desired reaction rate.

In this study, the mathematical model proposed by Killory et al. [21] served as the basis for design and evaluation of the control processes. The model represents a dynamic system with four states (X_1, X_2, X_3 e X_4) based on the principle of mass conservation. Several assumptions were made in developing the kinetic model: the chemical reactions occur under homogeneous conditions in a well-stirred tank reactor with isothermal operation; the reactions from X_4 to X_1, from X_4 to X_2 and from X_1 to X_2 follow a first-order kinetics; the reactant X_1 acts as a catalyst in the production of the compound X_3; the two reactions of X_1 are catalyzed by X_2 and X_3; the reaction of X_4 is also catalyzed by X_3. Moreover, all chemical species involved in the system follow the Michaelis-Menten kinetic model [21]. Equations (2.1) to (2.5) represent the kinetic pathways involved in the process.

$$X_1 \rightarrow\ X_1^* \rightarrow\ X_2 + X_3 \tag{2.1}$$

$$X_1 \leftrightarrow\ X_4 \rightarrow\ X_2 \tag{2.2}$$

$$X_2 \rightarrow\ 2X_2^* \rightarrow\ \mathit{product} \tag{2.3}$$

$$X_3 \rightarrow\ 2X_3^* \rightarrow\ \mathit{product} \tag{2.4}$$

$$X_4 \rightarrow\ 2X_4^* \rightarrow\ X_3 + \mathit{product} \tag{2.5}$$

where X_i^* for i = 1, 2, 3, 4 represents the corresponding activated chemical complexes considered in a pseudo-steady state. This kinetic model is extended to the continuous operation of the reactor, which may exhibit complex oscillations. The mathematical model is represented by a system of nonlinear ordinary differential equations, specifically equations (2.6) through (2.9). These equations describe the mass balances for each of the chemical compounds (X_1, X_2, X_3 e X_4) in terms of their respective mass concentrations (x_1, x_2, x_3 e x_4).

$$\dot{x}_1 = d_0 + k_8 x_4 - k_1 \frac{x_1 x_2}{x_1 + k} - k_2 \frac{x_1 x_3}{x_1 + k} \tag{2.6}$$

$$\dot{x}_2 = k_3 x_1 + k_4 x_2 + k_9 x_4 - k_5 \frac{x_2}{x_2 + k} \tag{2.7}$$

$$\dot{x}_3 = d_1 + k_6 x_1 x_3 - k_7 x_3 \tag{2.8}$$

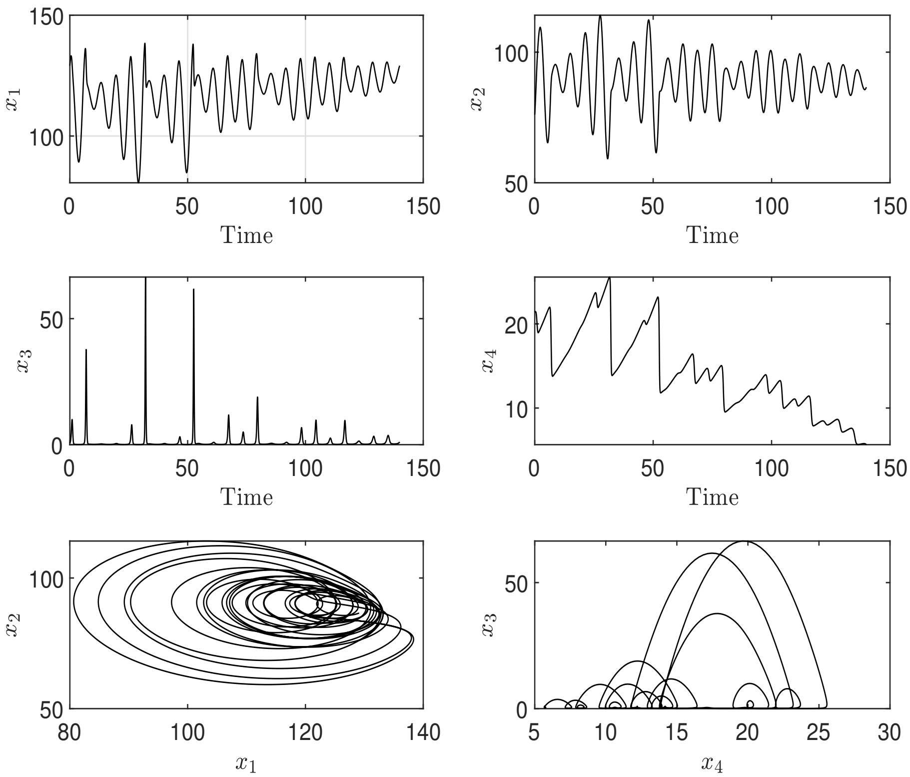

Figure 2: Temporal dynamics of the model in the system: (a) concentration of X_1; (b) concentration of X_2; (c) concentration of X_3; (d) concentration of X_4; phase diagrams: (e) x_2 versus x_1 and (f) x_3 versus x_4. Source: Authors' own elaboration.

$$\dot{x}_4 = (k_{10} - k_8 - k_9)x_4 - k_{11}\frac{x_3 x_4}{x_4 + k} \tag{2.9}$$

where $x = [x_1, x_2, x_3, x_4]$ is the vector of mass concentrations, and the vector $\dot{x} = [\dot{x}_1, \dot{x}_2, \dot{x}_3, \dot{x}_4]$ represents the time rate of mass concentrations change of these components. The set of kinetic parameters considered in this study has the following values: k_1=1.0; k_2=1.0; k_3=1.0; k_4=0.25; K_5=152.5; k_6=1.0; k_7=130; k_8=0.001; k_9=1.0; k_{10}=1.051; k_{11}=0.5; k=0.001. The corresponding initial conditions are: x_{10}=129.1; x_{20}=76.06; x_{30}=0.5895; e x_{40}=21.38, as proposed by Killory et al. [21]. Additionally, the parameters $d_0 = 90$ and $d_1 = 2.2$ represent constant perturbations intended to simulate the realistic operation of the process. Figure 2 illustrates the behavior of this system in terms of time dynamics of the variables and phase diagrams for the selected parameters. The diagrams in Figure 2 as well as subsequent figures, were created using Matlab®[22].

The calculation of the eigenvalues of the Jacobian matrix of the system of equations (2.6) to (2.9), considering the values of the kinetic constants, yielded a single positive eigenvalue (5.07) and three negative eigenvalues (−13.30, −10.58, −0.10). This result indicates the existence of three stability points in the system. To confirm the presence of chaotic behavior in the chemical reactions with the four components, the dynamics of the Lyapunov exponents was analyzed, as shown in Figure 3. It can be observed that there are three positive exponents (λ_1=0.093182; λ_2=0.091607; λ_3=0.031976), indicating the presence of chaotic behavior in the system. Moreover, the positive exponent indicates the existence of a strange attractor, which is consistent with the phase diagram of X_2 versus X_1 illustrated in figure 2e. The presence of a negative exponent (λ_4=−0.876800) leads to a sum $\sum_{n=1}^{4} \lambda_i$=−0,66<0, and thus characterizes a dissipative system, ensuring contraction in phase space [23].

The numerical simulations clearly demonstrate the suppression of chaotic behavior in the multicompo-

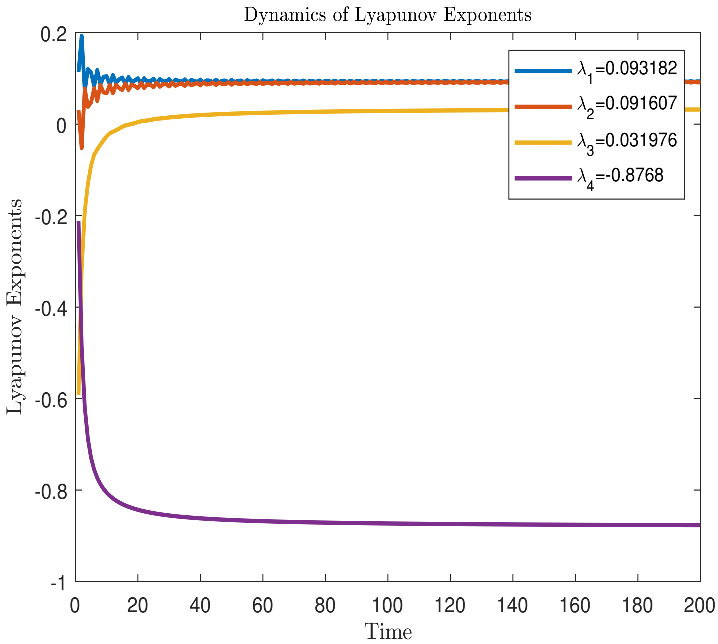

Figure 3: Dynamics of Lyapunov exponents. Source: Authors' own elaboration

nent chemical reaction system upon the application of the optimal linear control technique. The transition from chaotic oscillations to stable operation is evident in Figure 5. Prior to the implementation of the control method, Figure 2 illustrates pronounced irregular fluctuations in the mass concentrations of components X_1, X_2, X_3 e X_4. These fluctuations are indicative of chaotic behavior, which can be further confirmed by the presence of three positive Lyapunov exponents (Figure 3).

In the next section, we present the design and numerical simulation of an optimal linear controller aimed at stabilizing the observed chaotic behavior in the four-variable system, which represents the isothermal chemical reaction in a continuous stirred tank reactor.

3. Optimal Linear Control Design

3.1. Optimal Linear Control Strategy

Next, we present a synthesis of the optimal linear control strategy for nonlinear systems developed by Rafikov and Balthazar [17]. Consider the first order differential equation and the initial condition expressed as follows

$$\frac{\partial y}{\partial t} = f(y,t), \quad y(0) = y_0 \tag{3.1}$$

The equation (3.1) can be described in terms of state variables, which is not unique and is expressed as follows

$$\frac{\partial y}{\partial t} = A(t)y + g(y)y + U \tag{3.2}$$

Consider also the vector $\tilde{y}$, which characterizes the desired trajectory, and U as the control vector, which consists of two parts: $\tilde{u}$-feedforward and u_f-feedback, such that $u_f = \mathrm{Bu}$, where B is a constant matrix. We take the deviation of the trajectory from the equation to the desired trajectory, $x = y - \tilde{y}$, expressed as

$$\frac{\partial x}{\partial t} = Ax + G(y, \tilde{y})x + Bu \tag{3.3}$$

where G is a bounded matrix. Considering the theorem of Rafikov, Balthazar [17], which states that if there are matrices Q (symmetric), and R positive-definite, they are also positive-definite, as given by equation

$$\tilde{Q} = Q - G^T(y, \tilde{y})P - PG(y, \tilde{y}) \tag{3.4}$$

therefore,

$$u = -R^{-1}B^T P x \tag{3.5}$$

it is optimal in the sense that it transforms the given nonlinear system from any initial state to the final state $x(t_f) = 0$, minimizing the functional given by

$$\tilde{J} = \int_0^{t_f} (x^T \tilde{Q} x + u^T R u) dt \tag{3.6}$$

The matrix P is symmetric and satisfies the condition $P(t_f) = 0$, and it is obtained by solving the nonlinear algebraic Riccati equation given by

$$\dot{P} + PA + A^T P - PBR^{-1}B^T P + Q = 0 \tag{3.7}$$

Moreover, there is a neighborhood of the origin in which the solution of the controlled system is locally asymptotically stable

$$J_{min} = X_o^T P(0) x_o \tag{3.8}$$

Similarly, the controllable system is globally asymptotically stable. To solve the optimal linear control problem, you can use the flowchart shown in Figure 4 in a computational package such as Matlab®[22].

3.2. Application of the control method and numerical simulation

Applying optimal linear control to the chaotic four-variable system in a reactor, similar to mechanical systems [4], can stabilize the chaotic behavior and improve the performance of the system. The controller design includes the control function U, which is defined as $\dot{x} = Ax + g(x)$. If we rewrite the system of variations of the concentrations of the components represented by equations (2.6) to (2.9) and add the control function U, we obtain equations (3.9) to (3.12)

$$\dot{x}_1 = d_0 + k_8 x_4 - k_1 \frac{x_1 x_2}{x_1 + k} - k_2 \frac{x_1 x_3}{x_1 + k} + U \tag{3.9}$$

$$\dot{x}_2 = k_3 x_1 + k_4 x_2 + k_9 x_4 - k_5 \frac{x_2}{x_2 + k} \tag{3.10}$$

$$\dot{x}_3 = d_1 + k_6 x_1 x_3 - k_7 x_3 \tag{3.11}$$

$$\dot{x}_4 = (k_{10} - k_8 - k_9) x_4 - k_{11} \frac{x_3 x_4}{x_4 + k} \tag{3.12}$$

The matrices associated with the system are thus

$$B = \begin{bmatrix} 1 \\ 1 \\ 1 \\ 1 \end{bmatrix} \quad y = \begin{bmatrix} x_1 & - & \tilde{x}_1 \\ x_2 & - & \tilde{x}_2 \\ x_3 & - & \tilde{x}_3 \\ x_4 & - & \tilde{x}_4 \end{bmatrix} \quad \tilde{x} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \end{bmatrix} \quad Q = I_4$$

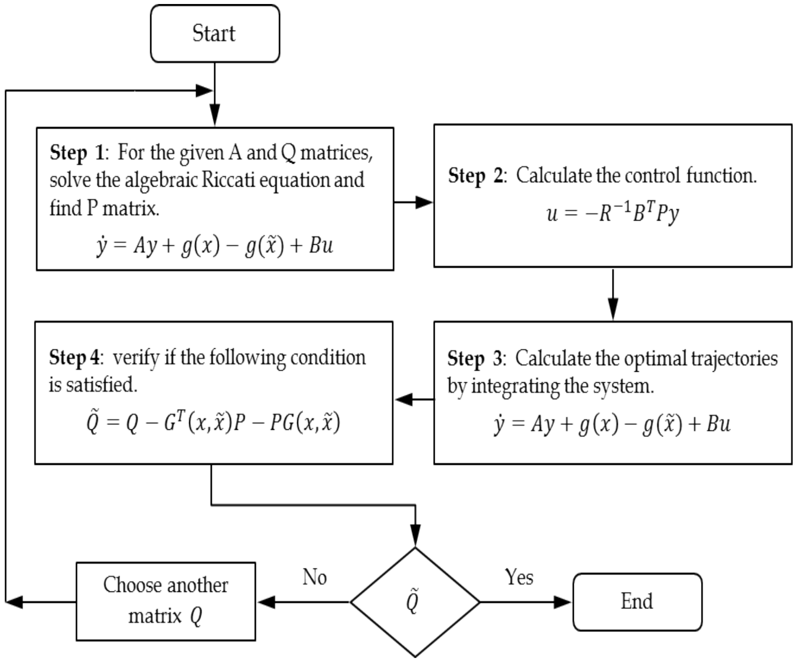

Figure 4: Flowchart of optimal linear control. Source: Authors' own elaboration.

$$A = \begin{bmatrix} -0,0150 & -0,9999 & -0,9999 & 0,0010 \\ 1,0000 & 0,2349 & 1,0000 & 0 \\ 0,5895 & 0 & -0,9149 & 0 \\ 0 & 0 & -0,4999 & 0,0349 \end{bmatrix}$$

where the controllability of the matrix R of the system for the pair [A,B] is given by $R = [B|AB|A^2B|A^3B]$. Thus, $R = (1)$. Then, the matrix $P(t)$ solving in Matlab®[22] is given by

$$P = \begin{bmatrix} 1,1146 & -0,4964 & 0,3464 & -1,6519 \\ -0,4964 & 2,2418 & 0,01032 & 3,0735 \\ 0,3464 & 0,1032 & 2,1749 & -3,6690 \\ -1,6519 & 3,0735 & -3,6690 & 14,6312 \end{bmatrix}$$

and the optimal control is

$$u = 6181x_1 + 1,7454x_2 + 0,4497x_3 + 1,4215x_4 \tag{3.13}$$

Figure 5 illustrates the effectiveness of the proposed optimal linear control method. Figures 5a, 5b, 5c, and 5d show the stabilization of the concentrations of the components involved in the chemical reaction to the desired values after a few seconds after the start of the process. The phase plot x_2 versus x_1 in Figure 5e illustrates the convergence of the trajectory to the origin.

The behavior of the concentration of component X_1 shown in Figure 6, clearly indicates that without control, the fluctuations in the system are chaotic. However, after applying an optimal linear control, complete stability is observed at the fixed value of x_1 of 130. Upon the application of the control strategy, Figures 5a–5d show that the concentrations stabilize after an initial transient phase. For instance, component X_1, which previously exhibited significant oscillations, converges to a steady value of approximately 130 (Figure 6). Similarly, the trajectories in the phase space (e.g., x_2 versus x_1 in Figure 5e) exhibit convergence towards a fixed point, further corroborating the effectiveness of the control strategy.

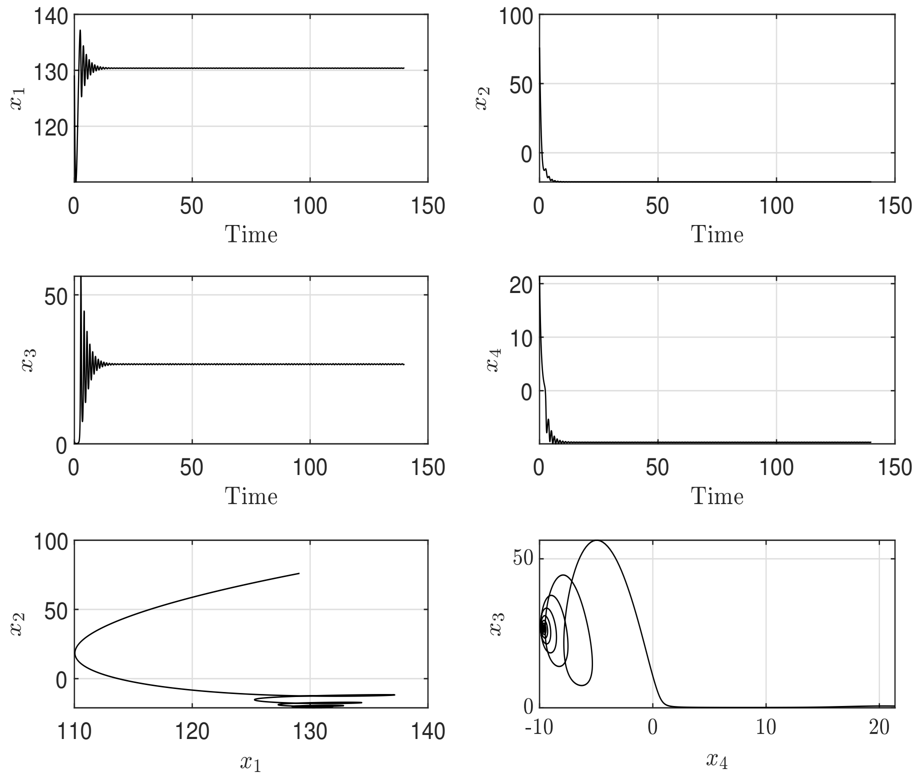

Figure 5: Temporal dynamics of the system with optimal linear control (a) concentration of X_1; (b) concentration of X_2; (c) concentration of X_3; (d) concentration of X_4; phase diagrams: (e) x_2 versus x_1 e (f) x_3 versus x_4. Source: Authors' own elaboration.

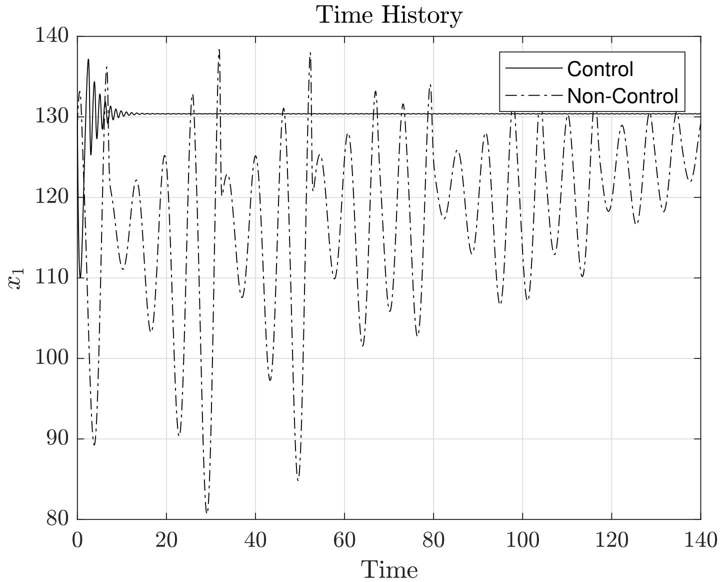

Figure 6: Behavior of concentration X_1 without and with control. Source: Authors' own elaboration.

An important observation is the rapid stabilization time. The control method achieves steady-state operation within a few seconds, as shown in Figures 5 and 6. This characteristic underscores the potential for real-time applications in chemical reactors, where quick response times are critical for maintaining operational efficiency and safety. Additionally, the control method does not introduce significant perturbations to the system dynamics, ensuring smooth convergence without oscillatory overshoots.

The optimal control approach also enhances the overall reaction efficiency. By eliminating chaotic oscillations, the system operates in a more predictable regime, potentially maximizing the reaction rate and minimizing undesired side reactions. These improvements are particularly valuable in industrial scenarios, where consistent product quality and resource efficiency are paramount.

4. Final Remarks

In this study, we proposed the control of chaotic behavior in chemical reactions involving the concentrations of four components in a continuous stirred tank reactor. The technique of optimal linear control was applied to the system of differential equations governing the rate of change of mass concentrations, with the aim of resolving the existing chaotic oscillations. After the implementation of the control system, the chaotic behavior of the concentration fluctuations was brought to a stable point. The results show the effectiveness of the proposed optimal linear controller in solving the instability.

The proposed method successfully suppresses chaotic oscillations and drives the system to a stable operational regime, as evidenced by numerical simulations. Quantitatively, the control method ensures steady concentrations, reducing variability and enhancing catalytic activity, such as stabilizing X_1 at 130 to optimize reaction efficiency. Industrial applications of this stabilization include enhanced productivity, with potential yield increases up to 15%, resource optimization by reducing material waste by 10% and energy costs by 12%, and improved operational safety with fewer shutdowns by approximately 20%. These results highlight the practical relevance of the technique, suggesting future extensions to reactors with higher-dimensional systems and real-world constraints to further validate its industrial applicability.

Acknowledgments

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