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Analysis and Control of The Methyl Isocyanate Hydrolysis Reaction

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Abstract
In this work, bifurcation analysis and Multiobjective Nonlinear Model Predictive Control is performed on Methyl isocyanate hydrolysis reaction in a CSTR. Bifurcation analysis is a powerful mathematical tool for studying the nonlinear dynamics of any process. Several factors must be considered, and multiple objectives must be met simultaneously. The MATLAB program MATCONT was used to perform the bifurcation analysis. The MNLMPC calculations were performed using the optimization language PYOMO in conjunction with the state-of-the-art global optimization solvers IPOPT and BARON. The bifurcation analysis revealed the existence of Hopf bifurcation points and limit points. The MNLMC converged on the Utopian solution. Hopf bifurcation points, which cause unwanted limit cycles, are eliminated using an activation function based on the tanh function. The most important significance of this work lies in the demonstration of a strategy that can eliminate the limit cycle causing Hopf bifurcations that are responsible for the Bhopal disaster when the hydrolysis of Methyl isocyanate is performed.

Keywords: Bifurcation, Optimization, Control, Methyl Isocyanate, CSTR

1. Background

The hydrolysis of Methyl isocyanate (MIC) is a fast, exothermic, and strongly nonlinear reactive process. due to the reactivity of the isocyanate functional group towards water. The formation of unstable compounds, the evolution of heat, and the formation of gases create conditions that can easily lead to imminent danger. Notably, this reaction is highly relevant to chemical safety, particularly in preventing industrial accidents involving highly reactive chemicals such as MIC.

The instability and hazardous nature of MIC largely stem from its exothermic, highly reactive hydrolysis. Methyl isocyanate, CH_3-N=C=O, is a highly toxic, small, and volatile molecule that is associated with the notorious Bhopal gas disaster. The chemical contains an isocyanate functional group (-N=C=O) that is highly susceptible to nucleophilic attack by water, making hydrolysis a major reaction pathway in the presence of moisture.

The hydrolysis of MIC proceeds with the attack of water molecules on the electrophilic carbon atom of the isocyanate group. This nucleophilic addition creates a carbamic acid intermediate, an unstable compound called methyl carbamic acid. Schematically, it could be described by the following reaction: MIC reacts with water to form methyl carbamic acid. This intermediate is thermodynamically unstable and rapidly decomposes into methylamine (CH_3NH_2) and carbon dioxide (CO_2). Thus, the general hydrolysis process converts MIC into less complex but still reactive and possibly hazardous products.

One of the defining features of hydrolysis in an MIC molecule is its strong exothermic nature. Large amounts of heat are generated during the synthesis and decomposition of the reactions. The reaction, when performed in a closed container with inadequate heat removal facilities, causes the temperature inside the container to rise rapidly, in turn increasing the rate of reaction.

The rate of MIC hydrolysis also depends on various conditions, such as temperature, water concentration, and the presence of catalysts or impurities. In fact, low concentrations of water are sufficient to cause the initiation of hydrolysis. On the other hand, an increase in temperature will significantly accelerate this reaction. In addition, basic and acidic impurities will act as catalysts and thus accelerate this reaction. In a factory setup, MIC is usually stored under carefully controlled conditions, refrigerated and kept dry.

In terms of safety, MIC hydrolysis is especially dangerous due to the possibility of runaway conditions. As a result of increasing temperature, not only will the rate of reaction increase, but other reactions such as polymerization of MIC may also happen. Due to the production of carbon dioxide, the reactor will experience an increase in internal pressure. If proper safety measures are not taken, venting or catastrophic failure can occur. Based on thermal, chemical, and pressure interactions, MIC Hydrolysis is a perfect example of a chemical instability problem.

The instability manifests as limit cycles, arising from Hopf bifurcations. This paper aims to demonstrate computational strategies to avoid the limit cycle causing Hopf bifurcations, which will ensure industrial safety for the of Methyl isocyanate (MIC) hydrolysis reaction.

2. Literature Review

Conducted studies of methyl isocyanate chemistry in the Bhopal incident [1]. Evaluated the kinetic parameters and critical runaway conditions in the hexamine-nitric acid reaction system to produce RDX in a non-isothermal batch reactor [2]. Analyzed the causes of the Bhopal disaster of conducted a review of clinical and experimental findings on the Bhopal gas tragedy 25 years after the tragedy [3,4]. Researched the variability of operating safety limits with a catalyst within a fixed-bed catalytic reactor for vapour-phase nitrobenzene hydrogenation [5].

Determined the connection between oscillatory thermal instability and the Bhopal disaster [6]. Determined optimal operating conditions for a catalytic reactor for butane oxidation using parametric sensitivity analysis and failure probability indicesp [7]. Researched the effects of thermal oscillations on the decomposition of organic peroxides, identifying hazard, utilization, and suppression [8]. Investigated the Thermal instability and runaway criteria and the dangers of disregarding dynamics [9]. Performed dynamic modeling and bifurcation analysis for the methylisocyanate hydrolysis reaction [10].

In this work, bifurcation analysis and multiobjective nonlinear model predictive control are performed for a methyl isocyanate hydrolysis reaction in a CSTR using the model described by [10]. The paper is organized as follows. First, the model equations are presented, followed by a discussion of the numerical techniques involving bifurcation analysis and multiobjective nonlinear model predictive control (MNLMPC). The results and discussion are then presented, followed by the conclusions.

3. Model Description

Methyl isocyanate (MIC) is an intermediate in the production of carbaryl and is commonly used as an insecticide. The hydrolysis of MIC is highly exothermic, generating methylamine and carbon dioxide as products. Additionally, MIC and methylamine can react to form dimethyl urea. The MIC hydrolysis reaction usually takes place in a large storage tank as a dynamic CSTR. Details of the model equations are briefly presented in this section. In this model, u represents the dimensionless MIC concentration, θ represents the dimensionless reactive temperature, τ the dimensionless time, f the dimensionless flow, θ_a the dimensionless cooling temperature, l, is the dimensionless heat transfer term, ε the dimensionless heat generated term, and γ The specific heat ratio.
The differential equations are

$$\begin{aligned} \frac{du}{dt} &= -ue^{(1/\theta_s - 1/\theta)} + f(1-u) \\ \frac{d\theta}{dt} &= ue^{(1/\theta_s - 1/\theta)} + f(\gamma\theta_a - \theta) - l(\theta - \theta_a) \end{aligned} \tag{1}$$

The base parameter values are
f = 1.7; γ = 1; ε = 10; θ_a = 0.0379; l =700; θ_s = 0.0371; (2)

4. Bifurcation Analysis

Bifurcation calculations are performed using the MATLAB software MATCONT. Bifurcation analysis explains the main causes for multiple steady-states and limit cycles. Branch points and limit points cause multiple steady-state solutions while limit cycles and oscillatory behavior are causd by Hopf bifurcation points [11,12]. The MATLAB program that effectively locates limit points, branch points, and Hopf bifurcation points is MATCONT. This program was developed and improved by several researhers This program is very effective in identifying Limit points(LP), branch points(BP), and Hopf bifurcation points(H) for an system of ordinary differential equations

$$\frac{dx}{dt} = f(x,\alpha) \tag{3}$$

$x \in R^n$ where the bifurcation parameter is α. The gradient vesctor is orthogonal to the tangent and hence the tangent plane at any point $w = [w_1, w_2, w_3, w_4, ...w_n]$ must satisfy

$$Aw = 0 \tag{4}$$

The matrix A is defined by

$$A = [\partial f / \partial x \quad | \partial f / \partial \alpha] \tag{5}$$

The sub-matrix $\partial f / \partial x$ is the Jacobian matrix. For both limit and branch points, the Jacobian matrix $J = (\partial f / \partial x)$ must have a determinant of 0.

At a limit point, there must exist only one tangent at the point of singularity. At this singular point, there is a one and only one non-zero vector, y, where $Jy = 0$. This vector is of dimension n. Since there is only one tangent, the vector $y = (y_1, y_2, y_3, y_4, ...y_n)$ must have the same direction with $\hat{w} = (w_1, w_2, w_3, w_4, ...w_n)$. Since

$$J\hat{w} = Aw = 0 \tag{6}$$

the n+1[th] component of the tangent vector $w_{n+1} = 0$. This is the necessary condition for the existence of a a limit point (LP).

For a branch point, two tangents must exist at the point of singularity. Let the two tangents be z and w. This implies that

$$\begin{aligned} Az &= 0 \\ Aw &= 0 \end{aligned} \tag{7}$$

Imagine a vector v that is orthogonal to one of the tangent w. v can be expressed as a linear combination of z and w ($v = \gamma z + \delta w$). Since $Az = Av$; $Av = 0$ and since w and v are orthogonal, $w^T v = 0$. Hence $Bv = \begin{bmatrix} A \\ w^T \end{bmatrix} v = 0$ which implies that B is singular. This implies that the necessary condition for the existence of a branch point is that the matrix $B = \begin{bmatrix} A \\ w^T \end{bmatrix}$ must be singular and have a determinant of 0.
At a Hopf bifurcation point,

$$\det(2f_x(x,\alpha)@I_n) = 0 \tag{8}$$

@ indicates the bialternate product while I_n is the n-square identity matrix. Hopf bifurcations cause limit cycles and should be eliminated because limit cycles make optimization and control tasks very difficult. More details can be found in respectively [13-15].

Hopf bifurcations lead to limit cycles, which can cause equipment damage and make control tasks more difficult. They also produce less desirable products. The tanh activation function (where a control value u is replaced by it) is used to eliminate spikes in the optimal control profiles. Several researchers have shown this explained, with multiple examples, how the same activation factor involving the tanh function also effectively removes the limit-cycle-causing Hopf bifurcation points [16-20].

5. Multiobjective Nonlinear Model Predictive Control(MNLMPC)

Originally developed a rigorous multiobjective nonlinear model predictive control(MNLMPC) strategy [21]. This procedure is used for performing the MNLMPC calculations. used. In a problem for which the variables $\sum_{t_i=0}^{t_i=t_f} q_j(t_i); j = 1, 2..., n$ have to be optimized simultaneously, and the dynamic model is given by

$$\frac{dx}{dt} = F(x,u) \tag{9}$$

t_f being the final time value, and n the total number of objective variables and u the control parameter. The single objective optimal control problem is solved independently for each of the variables $\sum_{t_i=0}^{t_i=t_f} q_j(t_i)$ and produces the values q_j^*. Then, the multiobjective optimal control (MOOC) problem that will be solved is

$$\min(\sum_{j=1}^{n}(\sum_{t_{i=0}}^{t_f} q_j(t_i) - q_j^*))^2$$
$$subject\ to\ \frac{dx}{dt} = F(x,u); \tag{10}$$

This will provide the values of u at different times. The first control value of u obtained is used, and the remaining values are discarded. This procedure repeats until the implemented value matches the initial one, or the Utopia point ($\sum_{t_{i=0}}^{t_f} q_j(t_i) = q_j^*; j = 1,2...,n$) is reached.

is used for these calculations. Here, the differential equations are converted to a Nonlinear Program (NLP) using the orthogonal collocation method. The NLP is solved using IPOPT and confirmed as a global solution with BARON [22-24].

6. Results and Discussion

Bifurcation Results

θ_a, θ_s, γ, f are the bifurcation parameters.

- When bifurcation analysis is performed with θ_a, as the bifurcation parameter, two Hopf bifurcation points were found at label (u, θ, θ_a) values of (0.307421 0.039045 0.037403)
- and (0.093443 0.041406 0.039256) (Figure. 1a). Figure. 1b and Figure. 1c show the limit cycles produced by these Hopf bifurcations. The Hopf bifurcations disappear when θ_a, is modified to $\theta_a \tanh(\theta_a)/2000$
- When bifurcation analysis is performed with θ_s, as the bifurcation parameter, one Hopf bifurcation point was found at (u, θ, θ_s) values of (0.283275 0.039599 0.037437) (Figure. 2a). Figure. 2b shows the limit cycle produced by this Hopf bifurcation. This Hopf bifurcation disappears when θ_s, is modified to $\theta_s \tanh(\theta_s)/0.002$ (Figure. 2c)
- When γ is the bifurcation parameter, two Hopf bifurcation points were found at label (u, θ, γ) values (0.307421 0.039045 0.447029) and (0.093443 0.041406 2.509320). (Figure. 3a). Figure. 3b and Figure. 3c show the limit cycles produced by these Hopf bifurcations. The Hopf bifurcations disappear when γ, is modified to $\gamma \tanh(\gamma)/10$ (Figure. 3d).
- When bifurcation analysis is performed with f as the bifurcation parameter, one Hopf bifurcation points were found at (u, θ, f) values of (0.221632 0.039628 1.588926) (Figure 4a). Figure. 4b shows the limit cycle produced by this Hopf bifurcation. This Hopf bifurcation disappears when f, is modified to $f \tanh(f)/2$ (Figure. 4c)

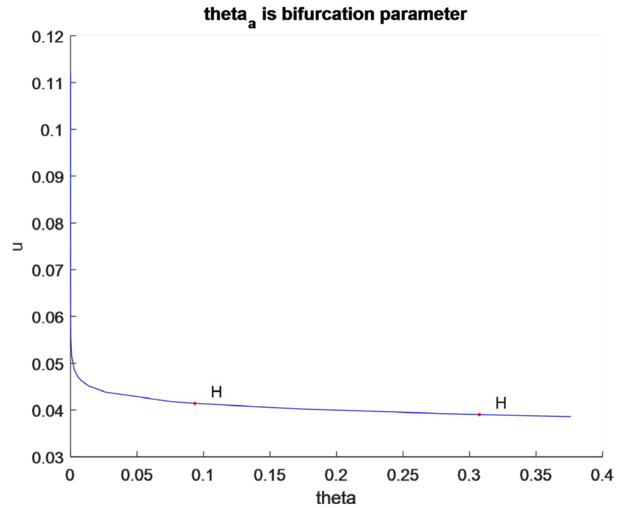

Figure 1a: θ_a, is the bifurcation parameter

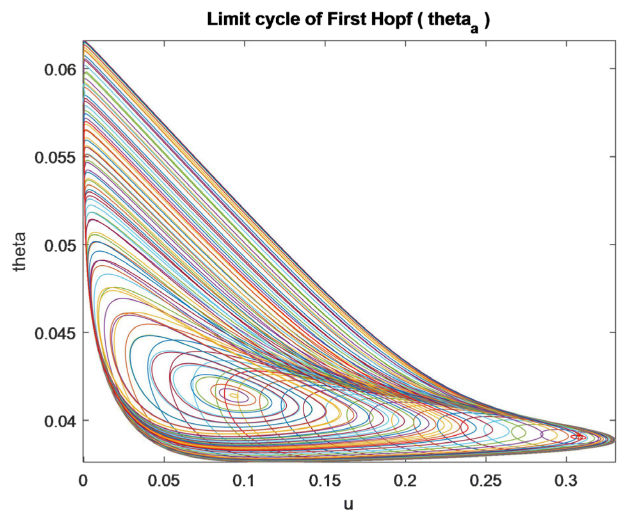

Figure 1b: Limit Cycle due to the first Hopf (θ_a, is the bifurcation parameter)

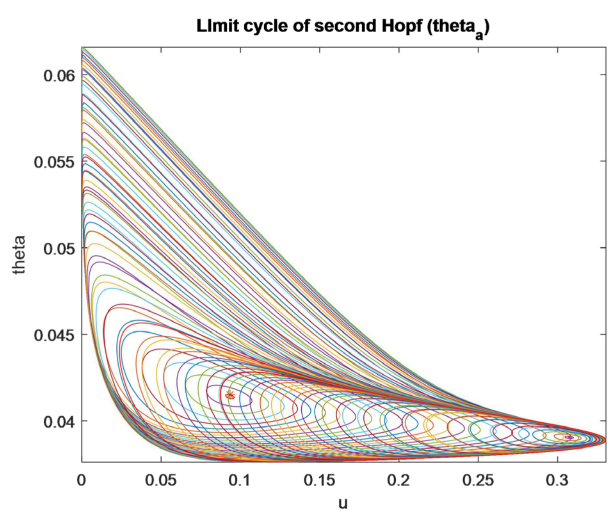

Figure 1c: Limit Cycle due to the second Hopf (θ_a, is the bifurcation parameter)

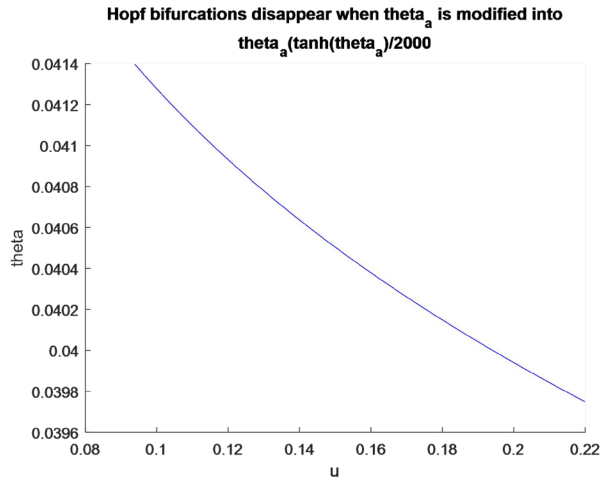

Figure 1d: Hopf Bifurcation Disappears when θ_a, is modified to θ_a tanh(θ_a)/2000

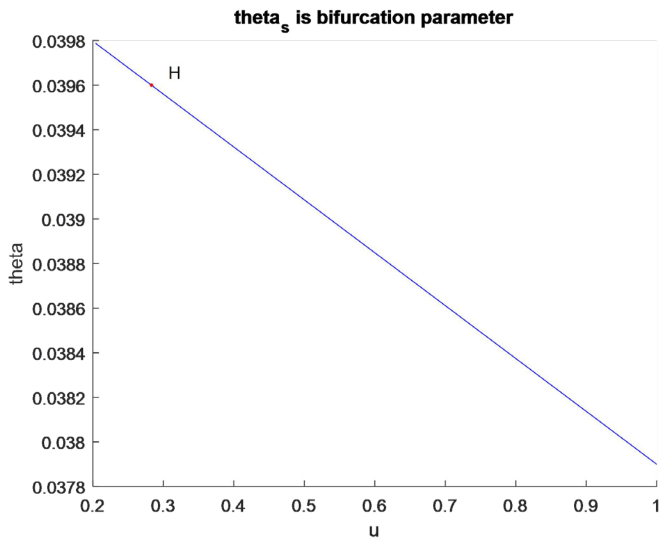

Figure 2a: θ_s, is the bifurcation parameter

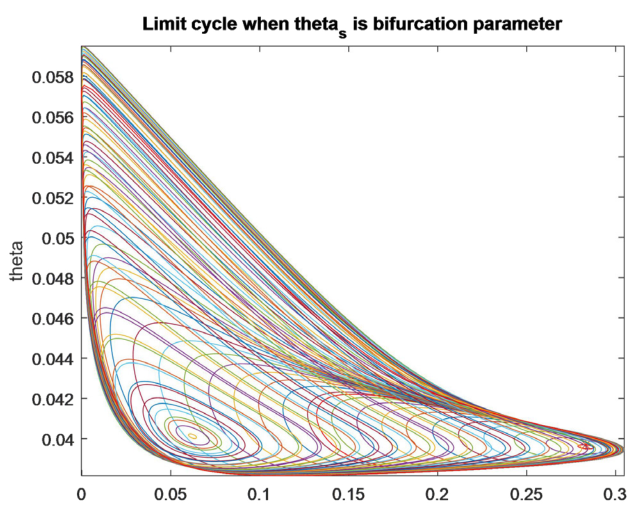

Figure 2b: Limit cycle When θ_s, is the bifurcation parameter

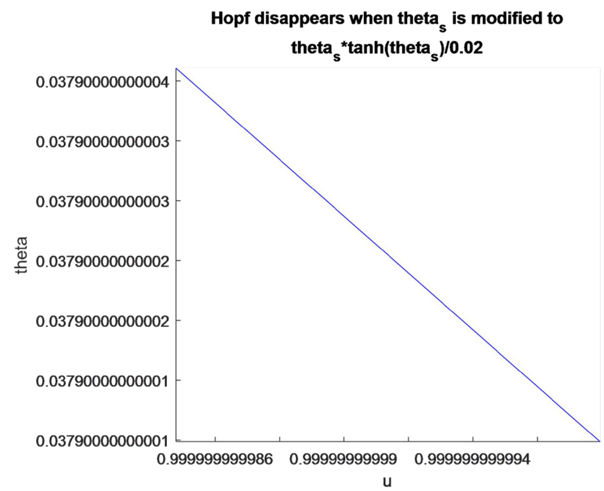

Figure 2c: Hopf Bifurcation Disappears when θ_s, is modified to $\theta_s \tanh(\theta_s)/0.002$

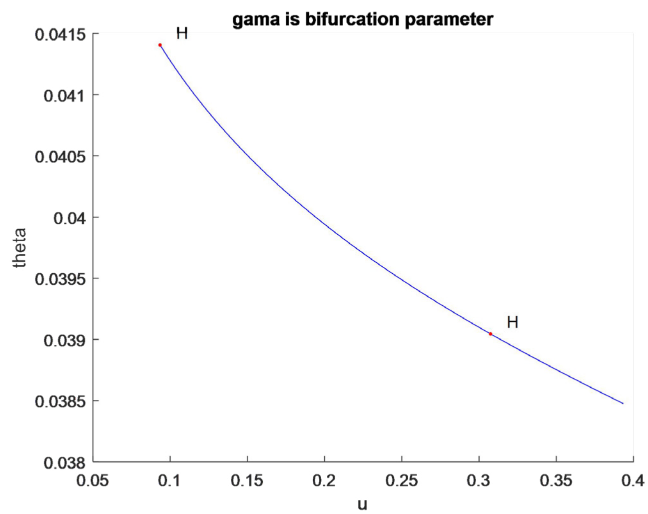

Figure 3a: γ is the bifurcation parameter

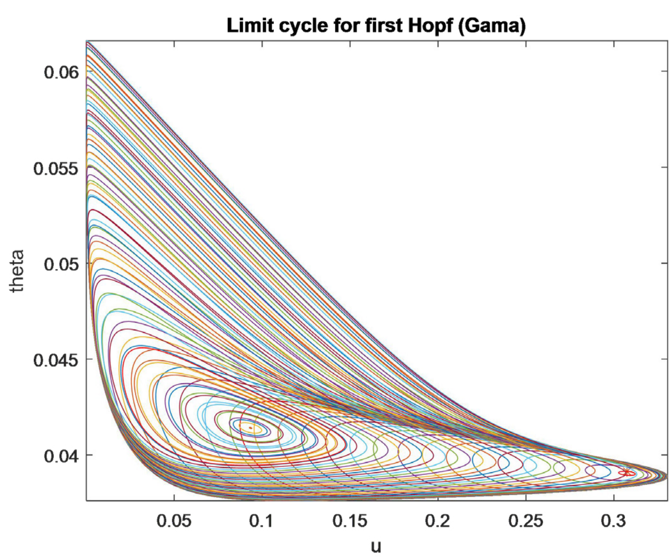

Figure 3b: Limit Cycle for first Hopf γ is the bifurcation parameter

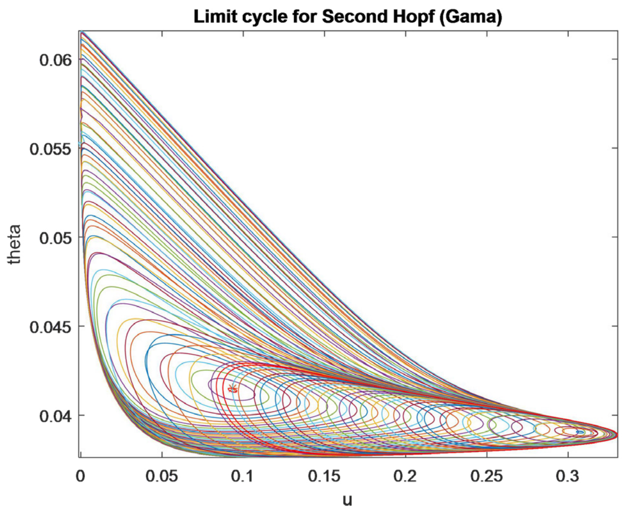

Figure 3c: Limit Cycle for second Hopf γ is the bifurcation parameter

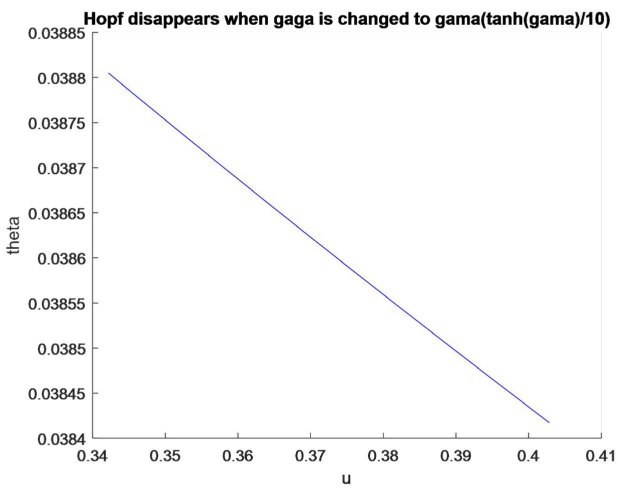

Figure 3d: The Hopf bifurcations disappear when γ, is modified to γ tanh(γ)/10

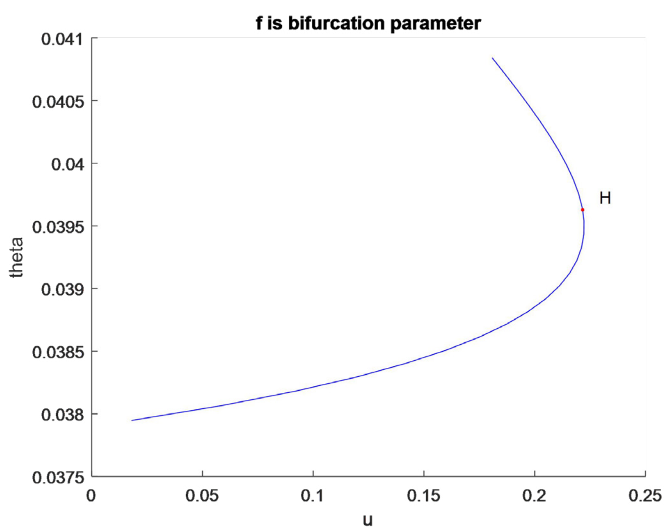

Figure 4a: f is the bifurcation parameter

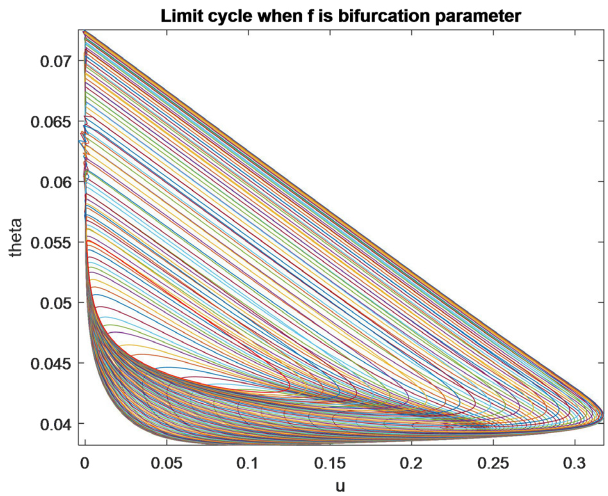

Figure 4b: Limit cycle when f is the bifurcation parameter

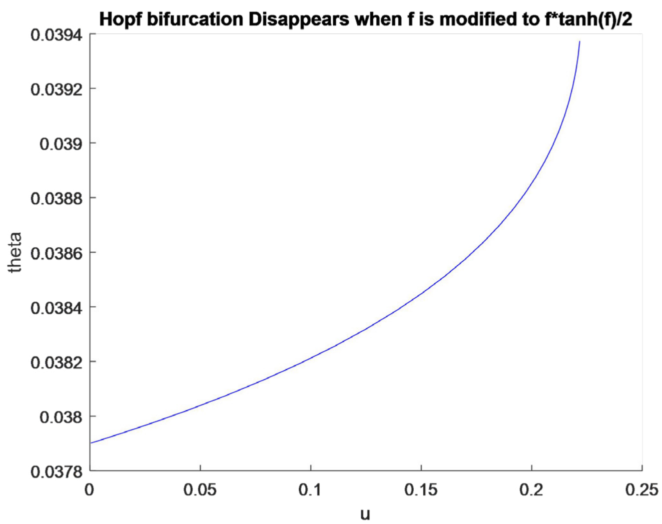

Figure 4c: Hopf disappears when *f*, is modified to *f* tanh(*f*)/2

The use of the activation factor involving the tanh function causes the Hopf bifurcations to disappear, thereby validating the analysis in Sridhar (2024) [20].

The Bhopal gas tragedy in December 1984 was caused by an uncontrolled chemical reaction resulting from the storage of Methyl Isocyanate (MIC). However, a more complex systems approach can place the event within the context of nonlinear dynamics, which explicitly refers to the occurrence of the Hopf bifurcation due to hydrolysis in MIC. The event explains how a gradual change in operational parameters can drive a chemical process from a stable state into self-sustained oscillations, ultimately leading to catastrophic instabilities. Methyl isocyanate reacts vigorously with water, producing methylamine, carbon dioxide, and heat in a largely exothermic process. Storage of MIC under ideal conditions involves keeping it dry, cold, and free from contaminants to prevent this reaction. In the case of the Bhopal tragedy, water contamination of a storage tank for MIC initiated this reaction process since the reaction rate is higher with increased temperatures, thereby having positive feedback related to temperatures and rates of reaction, which is a nonlinear behavior of a chemical system. In nonlinear systems, the Hopf bifurcation occurs when a steady operating point becomes unstable as a control parameter crosses a critical value. In MIC storage, control numbers include things like temperature, water level, and heat escape capacity. At first, the system might be in a steady state, with heat from slow hydrolysis balanced by heat loss to the surrounding air. But then, as the water contamination increases, the cooling systems stop working. The control number in the system shifts more. It reaches a point where the steady point can no longer hold, and the system changes into an oscillating mode. These oscillations cause substantial variations in the heat and pressure inside the storage tank. Each heat bump accelerates hydrolysis, producing more heat and gas. The increased pressure causes relief valves to release gases, which cools parts of the tank, but then the reaction speeds up again.

Near a Hopf bifurcation, these waves grow rather than shrink, pushing the system into a runaway state. This meant the heat rose quickly and the pressure built up until the tank burst, forcing the gases into the air, carrying the harmful MIC and other reaction products. This was what happened in Bhopal. The Bhopal disaster didn't come from one big failure. The whole storage system just lost its grip on stability. Little things—like a bit of water getting in or a small bump in temperature—used to get handled naturally. But not this time. Instead of smoothing things out, the system actually made those problems worse. Once things crossed a certain point, disaster wasn't just possible—it was going to happen. That's why the usual, neat-and-tidy safety checks didn't cut it. They expect small problems to get small reactions, but real life isn't always that simple. Looking at the Bhopal tragedy through the lens of Hopf bifurcation really drives home the crucial role of nonlinear dynamics in chemical safety. Staying safe isn't just about keeping everything "normal." You also have to make sure you're not creeping up on one of those tipping points where control slips away. In Bhopal, poor cooling, inadequate monitoring, and neglected maintenance pushed the MIC storage system right to the edge. Once they crossed that line, things started spiraling—oscillations, runaway reactions, and, finally, the release of toxic gas. The application of the tanh function to the manipulated variables will provide a strategy to avoid situations in both small- and large-scale operations that involve MIC. To avoid dangerous limit cycles, the activation factor involving the tanh function should be incorporated into the manipulated variables when the process is controlled.

7. Multiobjective Nonlinear Model Predictive Control (MNLMPC)

For the MNLMPC, the procedure described is followed. f, θ_a are chosen as the control parameters. $\sum_{t_i=0}^{t_i=t_f} u(t_i)$ is maximized and $\sum_{t_i=0}^{t_i=t_f} \theta_a(t_i)$ is minimized individually, and led to values of 2 and 0.0002. The overall optimal control problem will involve the minimization of was $(\sum_{t_i=0}^{t_i=t_f} u(t_i) - 2)^2 + (\sum_{t_i=0}^{t_i=t_f} \theta_c(t_i) - 0.0002)^2$ minimized subject to the model's equations. This led to a value of zero (the Utopia point). The MNLMPC values of the control variables f, θ_a are 1.494 and 0.0387. The control variables f, θ_a exhibit spikes, which are remedied using the Savitzky_Golay filter and result in the smooth profiles fsg, $\theta_a sg$. Figures 5a-5d show the MNLMPC profiles.

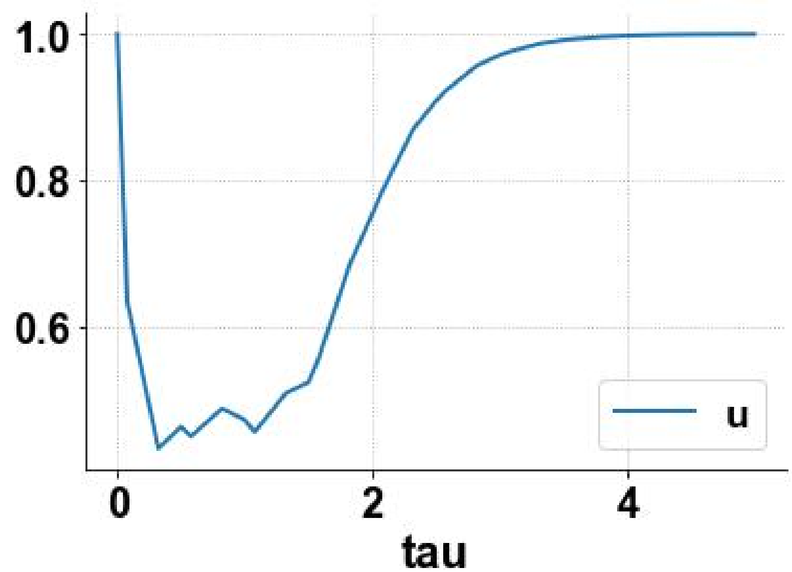

Figure 5a: MNLMPC profile of u

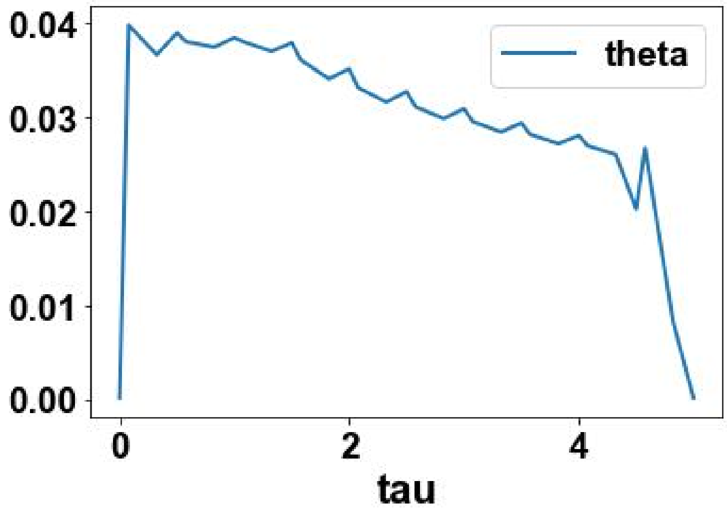

Figure 5b: MNLMPC profile of θ

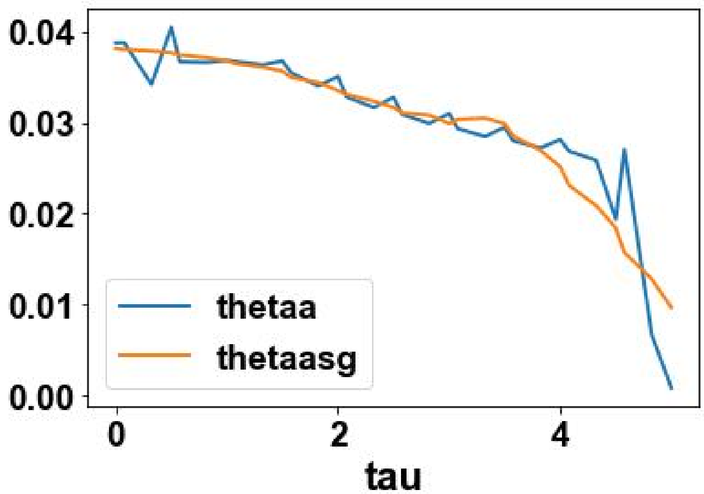

Figure 5c: MNLMPC profile of θ_a, $\theta_a sg$

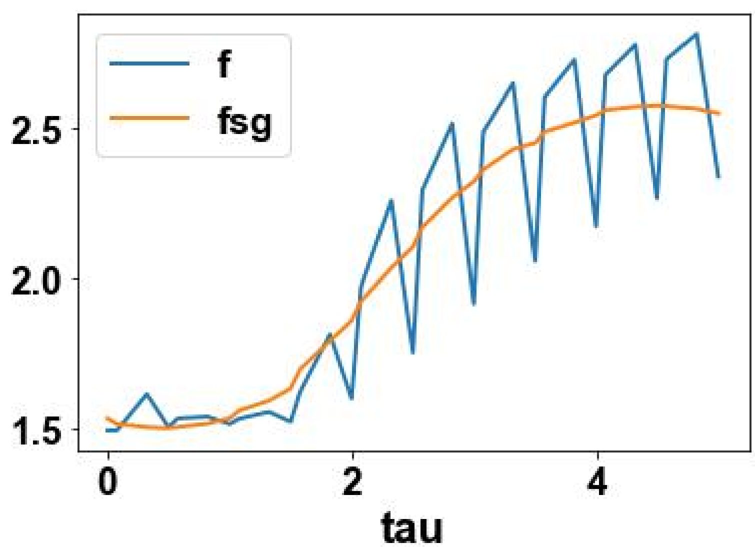

Figure 5d: MNLMPC profile of f and fsg

8. Importance of This Research

The issue of the avoidance of limit cycles in the hydrolysis reaction of methyl isocyanate, commonly denoted as MIC, is very essential when considering the safety of chemical processes. The reactant, MIC, undergoes a highly exothermic hydrolysis, which is prone to nonlinear behavior because the reaction rates are largely affected by concentration and temperature. It is important to note that in some operational conditions, such a nonlinear behavior can cause limit cycles in some of the essential system states, such as temperature.

Limit cycles in MIC Hydrolysis reactions can cause temperature oscillation. Each oscillation cycle can raise the reactor temperature to potentially damaging levels. Even if the oscillations do not cause the temperatures to exceed safe limits with each cycle, the cumulative effects of temperature stress can weaken the material used in the reactor's construction.

For control theorists, oscillations in a system reduce the efficacy of common feedback control techniques. Oscillations mean that the system cannot come to rest at a stable fixed point. For this reason, it is not possible to control temperature or concentration very well. In these cases, controllers might oscillate even further due to constant overcorrection. For MIC, this is particularly problematic. Here, small disturbances, such as contaminants, variations in cooling ability, or slight variations in the feeding stream, can combine with potential oscillations to cause very rapid growth.

Omitting limit cycles also plays a significant role in preventing dangerous emissions. The oscillating behavior could intermittently increase vapor pressure, leading to the emission of harmful MIC compounds and intermediates. Such cycles tend to emit intermittently, and prompt measures can barely be taken to restrict the exposure of workers and the adjacent community compared to continuous gas emissions.

Hence, it is imperative that reactor designs and operations actively avoid parameter spaces conducive to the existence of limit cycles during MIC hydrolysis reactions. This is because, with accurate models based on kinetics and bifurcation theory, it is possible to guarantee stable reactor behavior.

This work, in which a tanh activation factor is incorporated into the manipulated variable, is the first in the open literature to present a strategy to effectively eliminate the Hopf bifurcations that cause limit cycles.

9. Conclusions

Bifurcation analysis and multiobjective nonlinear control (MNLMPC) studies on a methyl isocyanate hydrolysis reaction in a CSTR. The bifurcation analysis revealed the existence of Hopf bifurcation points which cause unwanted limit cycles. These Hopf bifurcation points are eliminated using an activation function based on the tanh function. The Multiobjective Nonlinear Model Predictive Control (MNLMPC) calculations converge to the Utopia solution. A combination of bifurcation analysis and Multiobjective Nonlinear Model Predictive Control (MNLMPC) for the acetic anhydride hydrolysis reaction in a CSTR is the main contribution of this paper.

Data Availability Statement

All data used is presented in the paper

Conflict of interest

The author, Dr. Lakshmi N Sridhar, has no conflict of interest.

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