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Bifurcation-Aware Optimal Control of Nonlinear Reactive Crystallization Systems for Enhanced Productivity and Dynamic Stability

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Abstract

Reactive crystallization systems are widely used in pharmaceutical manufacturing, specialty chemicals production, and advanced materials processing, where crystal quality, process stability, and productivity are critically important. However, the strong nonlinear coupling between nucleation, crystal growth, and thermal feedback can generate complex dynamic behavior, including steady-state multiplicity, oscillatory instability, and limit cycle oscillations, which significantly affect industrial operability and product consistency. In this work, a nonlinear reactive crystallization model incorporating Arrhenius-type thermal effects, nonlinear nucleation kinetics, and reduced-order crystal surface area closure is investigated using bifurcation analysis and dynamic optimal control.

Bifurcation analysis was performed using MATCONT with the Damköhler number as the continuation parameter. The analysis identified a limit point and two supercritical Hopf bifurcation points, indicating the existence of stable oscillatory operating regimes. An optimal control framework was subsequently developed in Pyomo, where the Damköhler number was treated as a time-dependent control variable and the cumulative crystal mass production was maximized over a finite operating horizon.

To improve reactor operability, a Hopf-bifurcation-avoidance constraint was incorporated into the optimization problem. The bifurcation-aware strategy produced higher overall productivity compared to unconstrained operation by preventing inefficient oscillatory behavior. The results demonstrate that integrating nonlinear dynamics with optimal control can significantly enhance process stability, crystal production, and operational efficiency in industrial reactive crystallization systems.

Keywords: Reactive Crystallization, Hopf Bifurcation, Optimal Control, Nonlinear Dynamics and Dynamic Optimization

1. Introduction

Reactive crystallization processes play a central role in a wide range of industrial applications, including pharmaceutical manufacturing, fine chemicals production, specialty materials synthesis, and separation technologies. In these systems, chemical reaction, nucleation, crystal growth, and heat transfer occur simultaneously within a continuously operated reactor, leading to highly coupled and nonlinear dynamics. The interaction between these phenomena determines key process outcomes such as product yield, crystal size distribution, and overall process stability. As a result, reactive crystallization systems are often characterized by complex transient behavior that cannot be adequately captured using simple steady-state modeling approaches.

A defining feature of reactive crystallization systems is the strong coupling between reaction kinetics and crystal population dynamics. Solute consumption through crystal growth is directly linked to nucleation rates, which in turn depend nonlinearly on supersaturation. Additionally, these kinetic processes are strongly influenced by temperature through Arrhenius-type dependencies.

This thermal coupling introduces feedback loops in which heat generation accelerates reaction rates, further increasing crystallization activity and solute depletion. Simultaneously, heat removal through cooling provides a stabilizing mechanism. The competition between these destabilizing and stabilizing effects can give rise to rich nonlinear dynamic phenomena, including steady-state multiplicity, oscillations, and bifurcations.

Among these nonlinear behaviors, Hopf bifurcations and the resulting limit cycle oscillations are of particular importance. Such oscillatory regimes arise when the system transitions from a stable steady state to periodic behavior due to changes in key operating parameters, such as residence time or Damköhler number. While these oscillations may be mathematically stable, they are often undesirable in industrial practice because they induce periodic fluctuations in supersaturation, temperature, and crystal growth rates. This leads to variability in product quality, including inconsistent crystal size distributions and reduced downstream process efficiency.

In addition to nonlinear dynamics, optimal operation of crystallization systems remains a challenging problem due to the presence of strong feedback mechanisms and multiple competing objectives. Traditional optimization approaches typically focus on maximizing steady-state productivity without explicitly considering dynamic stability. However, operating conditions that maximize yield often lie near bifurcation boundaries, where small perturbations can trigger oscillatory or unstable behavior. This highlights the need for a more integrated approach that combines nonlinear dynamical analysis with optimal control theory.

In this context, bifurcation analysis provides a powerful tool for understanding the qualitative behavior of nonlinear systems. By identifying critical points such as limit points and Hopf bifurcations, it becomes possible to map regions of stability and instability in the parameter space. This information is essential for designing robust operating strategies that avoid undesirable dynamic regimes. When combined with dynamic optimization methods, bifurcation analysis enables the development of control strategies that are both performance-oriented and stability-aware.

This work develops a comprehensive framework for the analysis and optimal control of a nonlinear reactive crystallization system. The model incorporates nonlinear nucleation and growth kinetics, Arrhenius-type thermal effects, and reduced-order moment closure for crystal surface area. Bifurcation analysis is performed using MATCONT with the Damköhler number as the continuation parameter to identify critical transition points, including Hopf bifurcations and limit points. Subsequently, an optimal control problem is formulated in which the Damköhler number is treated as the manipulated variable, and the objective is to maximize cumulative crystal mass production over a finite time horizon.

Furthermore, a bifurcation-aware optimal control strategy is introduced by incorporating a Hopf-bifurcation-avoidance constraint into the optimization framework. This allows the system to operate in dynamically stable regions while maintaining high productivity. The combined use of bifurcation analysis and optimal control provides a unified framework for improving both the performance and operability of reactive crystallization systems, with direct implications for industrial-scale implementation.

2. Literature Review

The study of crystallization dynamics and control has evolved significantly over the past several decades, driven by the increasing industrial importance of particulate processes in pharmaceuticals, specialty chemicals, food processing, and advanced materials manufacturing. Early research primarily focused on the fundamental understanding of nucleation, crystal growth, and population balance modeling, while more recent studies have incorporated nonlinear dynamics, bifurcation analysis, and optimal control strategies for improving process performance and stability.

One of the foundational contributions to particulate process modeling was provided by Dhananjay Ramkrishna in 1990 through the development of generalized population balance methodologies for particulate systems [1]. His work established a mathematical framework for describing the evolution of particle populations in terms of nucleation, growth, aggregation, and breakage processes. Around the same period, Jaroslav Nyvlt and Otakar Sohnel (1991) investigated the kinetics of industrial crystallization systems, emphasizing supersaturation-driven nucleation and crystal growth mechanisms[2]. Their work provided important insight into metastability and operating conditions in industrial crystallizers.

A major milestone in crystallization engineering was the publication of J. W. Mullin’s seminal book Crystallization in 1993, which became one of the most influential references in the field [3]. Mullin systematically described nucleation kinetics, crystal growth mechanisms, heat and mass transfer effects, and industrial crystallizer design. Shortly afterward, Alan Randolph and Maurice Larson (1994) further advanced particulate process theory by developing mathematical formulations for particle population dynamics and particulate reactor systems [4].

During the mid-1990s, increasing attention was directed toward the dynamic behavior of crystallization processes. George Stephanopoulos and Murray Moo-Young (1996) investigated the dynamics and control of crystallization systems and demonstrated that strong coupling between nucleation and growth kinetics could lead to highly nonlinear reactor behavior [5]. Their work highlighted the importance of process dynamics in maintaining stable crystallization operation.

In 1998, S. K. Bhatia and Dhananjay Ramkrishna analyzed continuous precipitation systems and showed that nonlinear interactions between particle growth and supersaturation could generate dynamic instability [6]. These studies laid the groundwork for subsequent investigations into oscillatory crystallization phenomena and nonlinear bifurcation behavior.

The early 2000s marked a transition toward integrated crystallization process systems engineering. A. G. Jones (2002) introduced comprehensive process-systems approaches for crystallization modeling and control in his work Crystallization Process Systems [7]. During the same period, Hans Kramer and coworkers (2002) applied dynamic flowsheeting techniques to industrial crystallizers, demonstrating the importance of dynamic simulation in process optimization and operability studies. In parallel, Peter Daoutidis and Michael Henson (2003) investigated nonlinear control strategies for particulate processes, emphasizing the challenges associated with highly nonlinear reactor kinetics [8,9].

The importance of metastability and nucleation dynamics in industrial crystallizers was further investigated by Jens Ulrich and Jürgen Strege (2002), who studied the relationship between metastable zone width and crystal nucleation [10]. Their work provided important practical insights into crystallizer stability and product quality control.

In 2005, Allan Myerson published the Handbook of Industrial Crystallization, which consolidated modern developments in crystallization engineering, including nucleation theory, crystal growth modeling, process design, and industrial applications [11]. Around the same time, Masaaki Fujiwara (2005) explored crystallization engineering from the perspective of industrial process optimization and control [12].

As computational capabilities improved, advanced control strategies for crystallization systems became increasingly important. R. D. Braatz and collaborators made several major contributions in this area. D. Nagy and R. D. Braatz (2007) developed robust optimal control strategies for batch crystallization systems, demonstrating how dynamic optimization could improve crystal size distribution and productivity [13]. In 2008, Braatz reviewed advanced control approaches for crystallization systems and emphasized the growing importance of nonlinear model predictive control and optimization-based operation [14].

The integration of nonlinear control with particulate systems continued to evolve during the 2010s. Levent Özkan, Mayuresh Kothare, and Costas Georgakis (2011) investigated advanced control strategies for crystallization and particulate processes, highlighting the challenges associated with instability, uncertainty, and nonlinear process interactions [15]. In the same year, N. Mesbah and coworkers developed nonlinear model predictive control approaches for particulate systems with distributed particle populations [16].

Research attention then shifted toward optimal control of crystallization systems with explicit consideration of process dynamics. A. Rachah and coworkers (2013) developed optimal control strategies for alpha-lactose monohydrate crystallization, demonstrating that dynamic optimization could significantly improve crystal quality and process efficiency [17]. Around the same time, J. Ma, M. Baron, and R. D. Braatz (2013) investigated nonlinear control of crystal size distributions in batch crystallization systems [18].

The development of modern optimization tools further accelerated research in crystallization control. Lorenz Biegler (2014) introduced large-scale nonlinear programming methods for chemical processes, which became widely adopted for dynamic optimization and optimal control problems in process systems engineering [19]. These methods enabled the integration of differential-algebraic process models with advanced optimization algorithms such as IPOPT.

Recent research has increasingly focused on reactive crystallization systems and the interaction between reaction kinetics and crystallization dynamics. H. Y. Wang and J. D. Ward (2016) investigated the dynamics and control of continuous reactive crystallization processes and demonstrated the existence of strong nonlinear interactions between thermal feedback and crystal growth [20]. Their work highlighted the need for dynamic stability analysis in reactive crystallization systems.

More recently, R. K. Ramamoorthy and coworkers (2020) reviewed reactive crystallization mechanisms, emphasizing the importance of mixing, kinetics, and additive effects in controlling crystal formation [21]. In 2021, M. A. McDonald and coworkers presented a comprehensive review of reactive crystallization processes, identifying major challenges associated with process dynamics, scale-up, and control [22].

Current research trends increasingly emphasize the integration of machine learning, optimization, and nonlinear dynamics in crystallization systems. M. Sen, R. D. Braatz, and Panos Christofides (2022) investigated machine-learning-assisted optimization strategies for crystallization systems [23]. More recently, Y. Zhao, H. Wang, and R. D. Braatz (2025) proposed bifurcation-aware optimal control strategies for nonlinear crystallization systems, demonstrating the growing importance of integrating bifurcation analysis with process optimization [24].

Despite these advances, relatively limited work has been reported on the explicit incorporation of Hopf bifurcation constraints into optimal control formulations for reactive crystallization systems with strong thermal feedback and nonlinear nucleation kinetics. The present work addresses this gap by integrating MATCONT-based bifurcation analysis with Pyomo-based dynamic optimization for a nonlinear reactive crystallization model. The study explicitly incorporates Hopf-bifurcation avoidance into the optimal control framework and demonstrates that bifurcation-aware operation can improve both productivity and dynamic stability.

2.1 Main Objectives of This Study

The primary objective of this study is to develop a comprehensive nonlinear dynamical and optimal-control framework for a reactive crystallization system that exhibits strong coupling among reaction kinetics, crystal population dynamics, and thermal feedback effects. The system is characterized by nonlinear nucleation and growth mechanisms, Arrhenius-type temperature dependence, and moment-based closure approximations for crystal surface area, which together give rise to complex dynamic phenomena including steady-state multiplicity, Hopf bifurcations, and stable limit cycle oscillations.

A key objective is to systematically analyze the nonlinear dynamic behavior of the crystallization system using bifurcation theory. In particular, the study aims to identify critical operating regions where qualitative changes in system dynamics occur, such as limit points and Hopf bifurcation points, using the Damköhler number as the principal continuation parameter. The characterization of these bifurcations provides insight into the onset of instability and oscillatory behavior, which are highly relevant for industrial crystallization processes where stability and product consistency are essential.

Another major objective is to understand the impact of nonlinear oscillatory dynamics on reactor performance. The study investigates how stable limit cycles, arising from supercritical Hopf bifurcations, influence key process variables such as solute concentration, reactor temperature, crystal number density, and crystal mass density. This analysis is essential for linking nonlinear dynamic behavior with practical performance metrics such as productivity and product quality.

A further objective is to develop and implement a dynamic optimization framework that maximizes crystal production over a finite time horizon. The Damköhler number is treated as a time-dependent control variable, allowing manipulation of the relative timescales of reaction and transport processes. The optimization objective is formulated in terms of cumulative crystal mass, which represents the overall solid product yield.

In addition, a central objective of this work is to incorporate bifurcation information into the optimal control problem to improve process operability. Specifically, a Hopf-bifurcation-avoidance constraint is introduced to prevent the system from entering oscillatory regimes associated with limit cycle behavior. This bifurcation-aware control strategy is designed to ensure that optimal operation remains within dynamically stable regions of the state space while maintaining high productivity.

The study aims to demonstrate the practical value of integrating nonlinear dynamics with optimal control theory for industrial crystallization systems. By combining MATCONT-based bifurcation analysis with Pyomo-based dynamic optimization, the work provides a unified framework for understanding, predicting, and optimizing complex nonlinear process behavior. The overall goal is to enhance process performance, improve stability, and provide actionable insights for the design and operation of industrial crystallizers.

The present work introduces a novel integrated framework that combines nonlinear bifurcation analysis with dynamic optimal control for a reactive crystallization system exhibiting strong thermal and kinetic nonlinearities. While reactive crystallization processes have been extensively studied in the context of population balances, crystallization kinetics, and process optimization, relatively few studies have explicitly incorporated nonlinear dynamical behavior into the optimal control formulation. The novelty of this research lies in the simultaneous consideration of bifurcation structure, oscillatory instability, and productivity optimization within a unified computational framework.

A major contribution of this study is the integration of MATCONT-based bifurcation analysis with Pyomo-based dynamic optimization. The nonlinear reactive crystallization model exhibits limit points and Hopf bifurcations arising from the coupling between Arrhenius-type thermal feedback, nonlinear nucleation kinetics, and crystal growth dynamics. Unlike conventional crystallization studies that focus primarily on steady-state operation or trajectory optimization, the present work explicitly identifies and characterizes the onset of oscillatory instability through continuation and bifurcation techniques. The determination of supercritical Hopf bifurcations and stable limit cycle behavior provides deeper insight into the operability limits of the crystallization process.

Another important novelty is the development of a bifurcation-aware optimal control strategy. In most existing optimal control studies, the optimization problem is solved without considering whether the resulting operating trajectory enters dynamically unstable or oscillatory regions. In contrast, this work incorporates a Hopf-bifurcation-avoidance constraint directly into the dynamic optimization framework. This allows the optimizer to systematically avoid operating regions associated with sustained oscillations while still maximizing crystal production. The results demonstrate that avoiding oscillatory regimes can improve overall productivity, highlighting the importance of coupling nonlinear stability analysis with process optimization.

The study also introduces the use of the Damköhler number as a time-dependent manipulated variable in the optimal control problem. Since the Damköhler number represents the ratio between kinetic and transport timescales, its dynamic manipulation provides a physically meaningful mechanism for regulating crystallization behavior. This approach enables the control strategy to directly influence the balance between reaction intensity, crystal growth, and thermal effects, thereby improving reactor performance. From a modeling perspective, the work employs a reduced-order moment-closure approximation for crystal surface area, thereby retaining the essential nonlinear interactions between crystal number density and crystal mass density without requiring computationally expensive full population-balance formulations. This reduced-order formulation preserves the dominant instability mechanisms responsible for oscillatory behavior while remaining suitable for dynamic optimization and bifurcation analysis.

The present research contributes to the broader field of nonlinear process systems engineering by demonstrating how bifurcation theory can be embedded into optimal control formulations for complex chemical systems. The integrated methodology developed in this work provides a new pathway for designing productivity-enhancing yet dynamically stable operating policies for industrial crystallization processes. This combination of nonlinear dynamics, bifurcation-aware optimization, and reactive crystallization modeling represents a significant advancement beyond traditional steady-state or purely optimization-based approaches.

The rest of the paper is organized as follows. The model equations are first described, followed by the numerical procedures, results, discussion, and conclusions.

2.2 Model Equations

The reactive crystallization system considered in this work represents a strongly nonlinear physicochemical process in which solute consumption, crystal nucleation, crystal growth, and thermal feedback interact dynamically within a continuous reactor environment. Such systems are commonly encountered in industrial crystallization operations involving fine chemical production, pharmaceutical manufacturing, specialty materials processing, and precipitation-based separation systems, where the interplay between reaction kinetics and crystal population dynamics can lead to complex transient behavior. The present model is formulated to capture the essential mechanisms responsible for nonlinear dynamic phenomena including steady-state multiplicity, oscillatory instability, and self-sustained periodic crystallization behavior.

The model assumes that crystal formation occurs through simultaneous nucleation and growth processes, both of which are strongly influenced by the local thermal environment. Crystal growth consumes dissolved solute while simultaneously altering the available crystal surface area, thereby creating nonlinear coupling between concentration depletion and crystal population evolution. In addition, nucleation kinetics are modeled using a highly nonlinear power-law dependence on concentration, which introduces strong sensitivity to supersaturation levels. This nonlinear nucleation mechanism is one of the primary sources of dynamic amplification within the system.

Thermal effects play an equally important role in determining the overall reactor dynamics. The crystallization and reaction processes are thermally activated through an Arrhenius-type exponential dependence on temperature, resulting in strong positive feedback between temperature rise and kinetic acceleration. As the reactor temperature increases, both nucleation and growth rates intensify, leading to enhanced solute consumption and further heat generation. At the same time, thermal relaxation toward the coolant temperature introduces a stabilizing mechanism through heat removal.

The competition between destabilizing thermal activation and stabilizing heat dissipation creates the conditions necessary for nonlinear oscillatory behavior and bifurcation phenomena.

To reduce the computational complexity associated with detailed population balance formulations, a reduced-order moment closure approximation is employed for the crystal surface area. The surface area is represented as a nonlinear function of the crystal number density and crystal mass density, thereby preserving the essential coupling between crystal population dynamics and growth kinetics while maintaining a tractable low-dimensional dynamic model. This reduced formulation retains the dominant nonlinear mechanisms responsible for dynamic instability and oscillatory crystallization.

The resulting system consists of four coupled nonlinear ordinary differential equations describing the temporal evolution of the dimensionless solute concentration, reactor temperature, crystal number density, and crystal mass density. Due to the strong interactions among thermal feedback, nonlinear nucleation, and crystal growth kinetics, the model exhibits a rich bifurcation structure, including limit points, Hopf bifurcations, and stable periodic oscillations. These nonlinear dynamic features make the model particularly suitable for investigating operability limits, instability mechanisms, and oscillatory regimes in industrial reactive crystallization systems.

The original reactive crystallization model describes the coupled evolution of solute concentration, reactor temperature, crystal number density, and crystal mass density in a continuous crystallization system with nonlinear nucleation and growth kinetics. The model incorporates Arrhenius-type thermal feedback and crystal surface area effects responsible for complex nonlinear dynamic behavior including steady-state multiplicity, oscillatory instability, and limit cycle oscillations.

The dimensional solute balance is expressed as

$$\frac{dC}{dt} = \frac{C_f - C}{\tau} - k_g A C \exp\left(\frac{-E}{RT}\right) \tag{1}$$

where the first term represents feed replenishment and the second term corresponds to crystal growth consumption. The dimensional thermal balance is given by

$$\frac{dT}{dt} = \frac{T_f - T}{\tau} + \beta_r \exp\left(\frac{-E}{RT}\right) + \delta_r k_g AC - U\left(T - T_c\right) \tag{2}$$

in which the first term accounts for thermal exchange with the feed stream, the second term represents temperature-dependent thermal generation, the third term represents crystallization heat release, and the final term corresponds to heat removal through cooling. The crystal nucleation balance is represented by

$$\frac{dN}{dt} = k_b C^b \exp\left(\frac{-E}{RT}\right) - \frac{N}{\tau} \tag{3}$$

where nonlinear nucleation kinetics depend strongly on concentration through the power-law exponent *b*. The crystal mass balance is expressed as

$$\frac{dM}{dt} = 3k_g AC\exp\left(\frac{-E}{RT}\right) - \frac{M}{\tau} \tag{4}$$

where crystal mass accumulation is governed by nonlinear crystal growth kinetics.The crystal surface area is approximated using a reduced-order moment closure relation given by $A = (NM)^{1/3}$ which couples the crystal number density and crystal mass density through an effective surface area approximation. To reduce the governing equations into dimensionless form, the following scaling transformations are introduced: $s = \frac{C}{C_f}; \theta = \frac{T - T_f}{T_f}; n = \frac{N}{N_r}; m = \frac{M}{M_r}; t^* = \frac{t}{\tau};$

where C_f is the feed concentration, T_f is the feed temperature, N_r is the reference crystal number density, M_r is the reference crystal mass density, and τ is the characteristic residence time.

Using these scaling relations, the following dimensionless groups are obtained:

$$Da = k_g \tau A_r;\ \gamma = \frac{E}{RT_f};\ \lambda = U\tau;\ \beta = \beta_r \tau;\ \delta = \delta_r \tau$$

The dimensionless Arrhenius activation term becomes $\phi = \exp(\gamma\theta)$; and the dimensionless surface area closure relation is $a = (nm)^{1/3}$ .The resulting scaled equations used in the bifurcation analysis are therefore

$$\begin{aligned}
\frac{ds}{dt} &= Da(1-s) - k_g sa\phi \\
\frac{d\theta}{dt} &= -\theta + \beta Da\phi + \delta k_g sa - \lambda(\theta - \theta_c) \\
\frac{dn}{dt} &= k_b s^b \phi - n \\
\frac{dm}{dt} &= 3k_g sa\phi - m
\end{aligned} \tag{5}$$

The nonlinear coupling between thermal activation, crystal growth, and nucleation introduces strong feedback mechanisms capable of generating limit points, Hopf bifurcations, and stable periodic oscillations under appropriate operating conditions. Tables 1 and 2 summarize details about model variables and parameters.

Symbol	Variable Description
C	Solute concentration
T	Reactor temperature
N	Crystal number density
M	Crystal mass density
A	Crystal surface area
C_f	Feed concentration
T_f	Feed temperature
T_c	Coolant temperature
s	Dimensionless solute concentration
θ	Dimensionless temperature
n	Dimensionless crystal number density
m	Dimensionless crystal mass density
a	Dimensionless crystal surface area
ϕ	Arrhenius activation term
t	Time
t^*	Dimensionless time

Table 1: Model Variables

Symbol	Parameter Description	Value
k_g	Crystal growth coefficient	1.50
k_b	Crystal nucleation coefficient	1.50
b	Nucleation nonlinearity exponent	13.0
β	Dimensionless thermal generation coefficient	1.20
δ	Crystallization heat coupling parameter	70.5
λ	Thermal relaxation coefficient	4.80
γ	Arrhenius sensitivity parameter	0.543997
θ_c	Dimensionless coolant temperature	0.05
Da	Damköhler number	1.50
E	Activation energy	—
R	Universal gas constant	8.314 J mol^{-1} K^{-1}
U	Heat transfer coefficient	—
τ	Residence time	—
A_r	Reference surface area	—
N_r	Reference crystal number density	—
M_r	Reference crystal mass density	—

Table 2: Model Parameters

2.3 Bifurcation Analysis and Optimal Control

2.3.1 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 caused by Hopf bifurcation points. The MATLAB program that effectively locates limit points, branch points, and Hopf bifurcation points is MATCONT. This program was developed and improved by several researchers (Dhooge Govearts, and Kuznetsov, 2003) [25]. This program is very effective in identifying Limit points (LP), branch points (BP), and Hopf bifurcation points(H) for a system of ordinary differential equations

$$\frac{dx}{dt} = f(x,\alpha) \tag{6}$$

$x \in R^n$ where the bifurcation parameter is α. The gradient vector is orthogonal to the tangent and hence the tangent plane at any point $w = [w_1, w_2, w_3, w_4, ...w_{n+1}]$ must satisfy

$$Aw = 0 \tag{7}$$

The matrix A is defined by

$$A = [\partial f / \partial x \quad | \partial f / \partial \alpha] \tag{8}$$

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, the $n+1^{th}$ component of the tangent vector w_{n+1} = 0 . For a branch point,

the matrix $B = \begin{bmatrix} A \\ w^T \end{bmatrix}$ must be singular and have a determinant of 0.

At a Hopf bifurcation point,

$$\det(2 f_x(x,\alpha) @ I_n) = 0 \tag{9}$$

$@$ indicates the bialternate product while I_n is the n-square identity matrix. A Hopf bifurcation occurs when a complex conjugate pair of eigenvalues of the Jacobian matrix crosses the imaginary axis, resulting in the loss of stability of an equilibrium and the emergence of a limit cycle. Numerically, this is detected in MATCONT when the real part of a pair of complex eigenvalues changes sign while remaining nonzero in the imaginary direction. 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 Kuznetsov (1998)) and Govaerts [2000] respectively [26,27].

2.3.2 Optimal Control

Pyomo.dae (Hart et al, 2017) is used for the Optimal Control calculations. Pyomo.DAE is a powerful extension of the Pyomo optimization modeling framework, which is well-suited for solving dynamic systems of differential and algebraic equations [28]. It is a symbolic environment for solving differential-algebraic equation systems in the context of optimization problems. This is very important in process systems engineering, chemical kinetics, and control systems, where the dynamic response of systems is of prime interest.

At its heart, Pyomo. DAE enables users to define time-varying variables, derivatives, and constraints symbolically, which can be easily integrated into a Pyomo model. Users can easily define continuous sets for time or other continuous variables, which can be used to define their derivatives over those sets. This symbolic approach enables users to easily discretize continuous differential-algebraic equation systems using finite difference, collocation, or orthogonal collocation methods, thereby transforming continuous differential equations into algebraic equations that can be solved with standard solvers. The framework can handle both initial-value problems and dynamic optimization problems. In dynamic optimization, Pyomo.DAE allows the formulation of time-dependent objective functions and constraints, which is particularly useful in optimal control, energy systems, and chemical process scheduling problems.

One of the major advantages of Pyomo.DAE is that it is compatible with the Pyomo ecosystem. This allows users to leverage existing solver interfaces, variable bounds, nonlinear constraints, and objective functions within a combined static and dynamic modeling framework. Furthermore, the symbolic framework makes it easier to perform model verification, automatic differentiation, and sensitivity analysis. Pyomo.DAE provides a flexible, extensible, and open-source environment for modeling, simulation, and optimization of dynamic systems. By integrating symbolic modeling of DAEs with powerful discretization and optimization capabilities, it provides a unique framework for solving complex time-dependent problems. Its tight integration with Pyomo enables the efficient solution of both simple and complex dynamic optimization problems, making it a cornerstone of modern computational modeling of dynamic systems. In Pyomo. DAE , the differential equations are converted to a Nonlinear Program (NLP) using the orthogonal collocation method. The NLP is solved using IPOPT (Wächter & Biegler, 2006) [29].

2.4 Formation of Stability Dataset from MATCONT Results

A stability dataset was developed from numerical continuation calculations performed in MATCONT. The stability dataset consists of rows, each representing a continuation point from an equilibrium branch. Each row contains the state variables, the bifurcation parameter, and a stability measure [30]. The stability measure is a numerical value derived from the Jacobian matrix. The Jacobian matrix is computed numerically at each equilibrium point. The eigenvalues are then computed automatically using MATLAB. The maximum value of the real part of these eigenvalues is then computed as a scalar stability measure [31].

The stability measure is computed using "eig_real_max = max(real(eigvals));" in MATLAB. The stability measure is a quantitative metric in which negative values indicate locally asymptotically stable equilibria, positive values indicate instability, and a zero crossing indicates a Hopf bifurcation. The stability dataset is then saved as a CSV file. The dataset can then be used in subsequent computational calculations to perform classification or regression to identify stability boundaries or approximate bifurcations [32].

2.5 Neural Network Surrogate for Stability Prediction

Direct embedding of eigenvalue calculations into IPOPT-based optimal control is impractical for several reasons: (i) computing eigenvalues at each time step is computationally expensive, (ii) the mapping from states to the maximum eigenvalue is non-smooth near eigenvalue crossings, and (iii) symbolic differentiation of eigenvalues is challenging.

Prior to neural-network training, all input variables were standardized to improve numerical conditioning and training stability. Let x_{raw} denote the vector of state variables and bifurcation parameters= obtained from the stability dataset. For each input variable j, the training mean μ_j and training standard deviation σ_j were computed over all training samples. = The training mean for the input variable j is defined as the arithmetic average over all training samples as $\mu_j = \frac{1}{N}\sum_{k=1}^{N} x_j^{(k)}$ and the training standard deviation for the input variable j is defined as: $\sigma_j = \frac{1}{N}(\sum_{k=1}^{N} x_j^{(k)} - \mu_j)^2$

The standardized inputs were defined as

$$x_j = \frac{x_{raw,j} - \mu_j}{\max(\sigma_j, \varepsilon_{scale})} \tag{10}$$

where ε_{scale} is a small positive regularization parameter introduced to prevent numerical singularities associated with extremely small variances. This transformation ensures that all inputs remain properly scaled while avoiding excessively large neural-network activation arguments during optimization. The normalization procedure improves neural-network conditioning and enhances the robustness of gradient-based optimization. The vectors μand σ computed during training were stored and embedded identically within the Pyomo optimal-control formulation to ensure consistency between neural-network training and deployment.

To overcome these limitations, a feedforward neural network is trained to approximate the maximum eigenvalue as a smooth function of the system state and bifurcation parameter. A typical architecture employs the hyperbolic tangent (tanh) as a smooth activation function. If the input vector is denoted by x, which represents the scaled variables, then the network is defined as:

The vectors μ,σ computed during training were stored and embedded identically within the Pyomo optimal control formulation to ensure consistency between neural network training and deployment.

To avoid repeated eigenvalue computations during optimization, a feedforward neural network is trained to approximate the maximum real eigenvalue as a smooth function of the system states and bifurcation parameter. Using hyperbolic tangent activation functions ensures smooth differentiability required by IPOPT. If the input vector is denoted by x, which are the scaled variables, the network architecture is defined as

$$\begin{aligned} z1 &= tanh\left(W1\,x + b1\right) \\ z2 &= tanh\left(W2\,z1 + b2\right) \\ \lambda\_max\_NN &= W3\,z2 + b3 \end{aligned} \tag{11}$$

where W_i and b_i denote the weight matrices and bias vectors, respectively. The hidden-layer variables z_1 and z_2 represent nonlinear transformations of the input variables and intermediate features. The final output $\hat{\lambda}_{max}$ provides a smooth approximation of the spectral abscissa. Because tanh is infinitely differentiable, the network is fully smooth, guaranteeing the availability of first and second derivatives required by IPOPT. Without biases, the network output would be constrained to pass through the origin, limiting flexibility.

The hidden-layer outputs z1, z2, represent nonlinear combinations of the inputs and previous-layer features, respectively. Each element of z_1 is a smoothed combination of the original inputs, while each element of $z2$ encodes more abstract patterns extracted from z1. The final output λ_max_NN provides a smooth approximation of the maximum real eigenvalue, enabling efficient and differentiable stability evaluation within the optimal control problem. The integration into optimal control is done using a soft penalty formulation where we use a smooth_max function that converts λ into a smooth, nonnegative penalty that only "activates" when the system is unstable:

$$s_{\max}(\lambda+\varepsilon_1) = smooth - \max(\lambda+\varepsilon_1) = \frac{(\lambda+\varepsilon_1)+\sqrt{(\lambda+\varepsilon_1)^2+\varepsilon_1}}{2} \tag{12}$$

λ is the neural network's predicted maximum eigenvalue at the current state and parameter, while ε is a small positive safety margin to ensure differentiability. The soft penalty formulation involves the new objective function, where the original objective function $\sum optv(t) = \sum (m(t)$ is modified to $\sum (optv(t) + \alpha_{hopf}.s_{\max}(\lambda+\varepsilon_1))^2$ α_{Hopf} controls how aggressively instability is penalized, and ε_1 prevents numerical issues at exactly λ = 0 and slightly shifts the stability boundary. The goal is to maximize the objective function value while ensuring differentiability for IPOPT and avoiding non-smooth optimization. This approach avoids repeated eigenvalue computations and provides a smooth, differentiable surrogate suitable for gradient-based optimization. Furthermore, this enables the incorporation of stability constraints into optimal control without explicitly computing eigenvalues during optimization.

To facilitate integration into optimal control, a stability dataset is constructed from MATCONT continuation results. At each equilibrium point, the Jacobian is evaluated and its eigenvalues computed. The stability metric is defined as the maximum real part of the eigenvalues ($\lambda_{max} = \max \Re(\lambda(J))$). Negative values indicate stability, positive values indicate instability, and zero crossings correspond to Hopf bifurcations.

To avoid repeated eigenvalue computations within optimization, a feedforward neural network is trained to approximate the spectral abscissa as a smooth function of the system state and bifurcation parameter. Using hyperbolic tangent activation functions ensures smooth differentiability required by gradient-based solvers. The resulting surrogate provides a differentiable stability indicator suitable for embedding within optimal control formulations.

3. Results

Bifurcation analysis was performed using MATCONT with Da as the bifurcation parameter. A limit point was obtained at $[s,\theta,n,m,Da]$ values of (0.480566 3.942655 0.000934 2.425681 1.556618). Two Hopf bifurcation points were obtained at $[s,\theta,n,m,Da]$ values of (0.489281 3.792130 0.001087 2.377679 1.551852) with the first Lyapunov coefficient = -3.044928e+00 and at $[s,\theta,n,m,Da]$ values of (0.492892 3.728609 0.001155 2.353341 1.546904) with first Lyapunov coefficient = -7.939978e-02.

The negative Lyapunov coefficients indicate that both Hopf bifurcations are **supercritical**, leading to stable limit cycle oscillations [33]. The bifurcation diagram is shown in Fig. 1a. The limit cycles corresponding to the two Hopf bifurcation points were shown in Figs 1b and 1c.

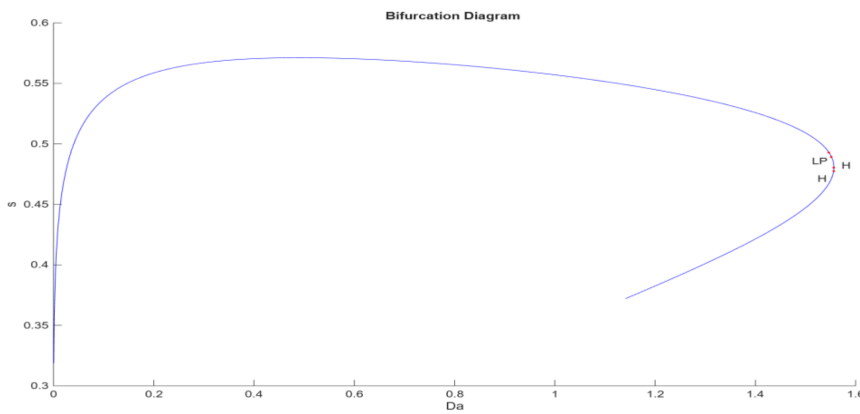

Figure 1a: Bifurcation Diagram Demonstrating one Limit point and 2 Hopf Bifurcation Points

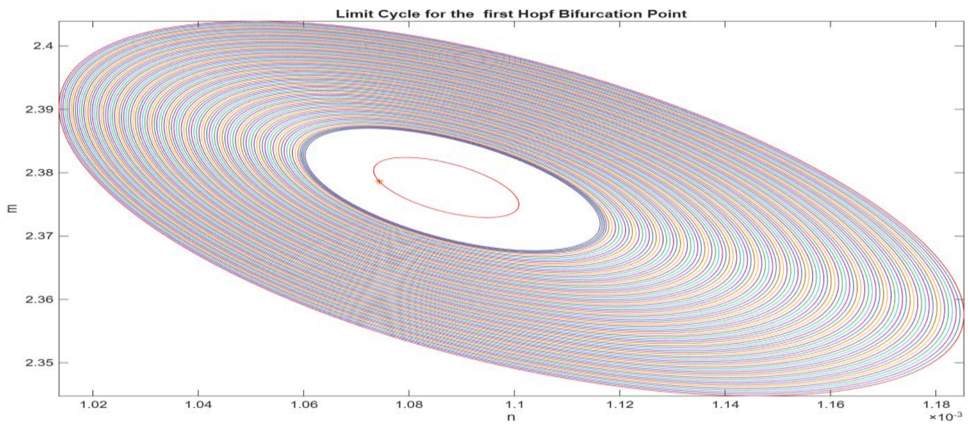

Figure 1b: Limit Cycle for the First Hopf Bifurcation Point

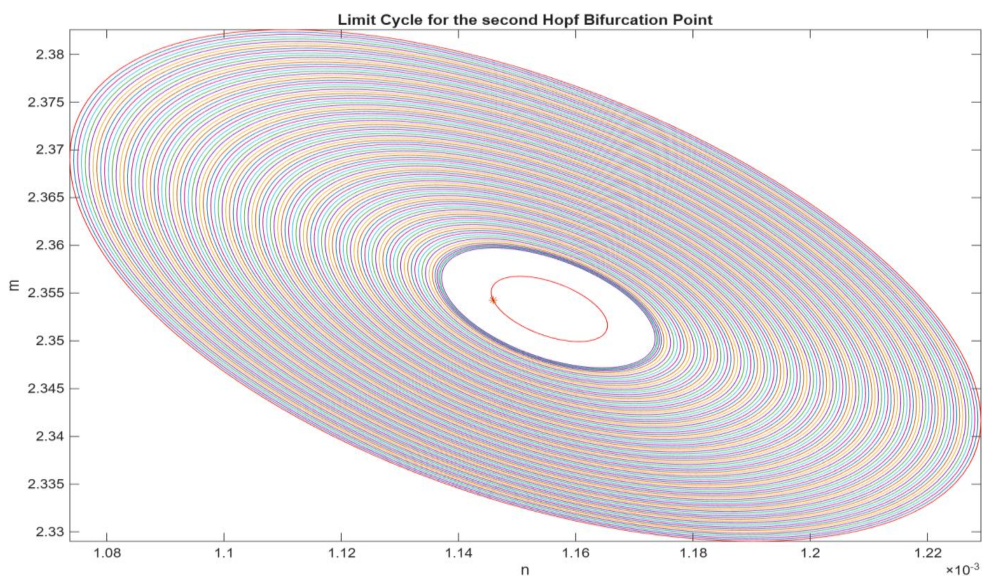

Figure 1c: Limit Cycle for the Second Hopf Bifurcation Point

To investigate the effect of bifurcation-aware operation, an optimal control problem was solved both without and with a Hopf-bifurcation-avoidance constraint. PYOMO. DAE with IPOPT was used. The objective was to maximize the crystal mass density m(t) using the objective function $\sum((optv(t)=m(t))+\alpha_{hopf}.s_{\max}(\lambda+\varepsilon_1))^2$, over the specified operating horizon. The corresponding objective function was formulated to maximize the overall reactor conversion while maintaining feasible reactor operation. In the absence of the Hopf bifurcation constraint α_{Hopf} = 0, the obtained optimal objective value of $\sum((optv(t)=m(t))$ was 49.62 . When the Hopf bifurcation avoidance constraint was activated with α_{Hopf} = 0.011, the optimal objective value increased to 50.08. Figures 2a-2d show the profiles without and with the Hopf Bifurcation constraint. This improvement indicates that operating the system outside the oscillatory regime can enhance overall productivity by preventing dynamic losses associated with sustained limit-cycle behavior.

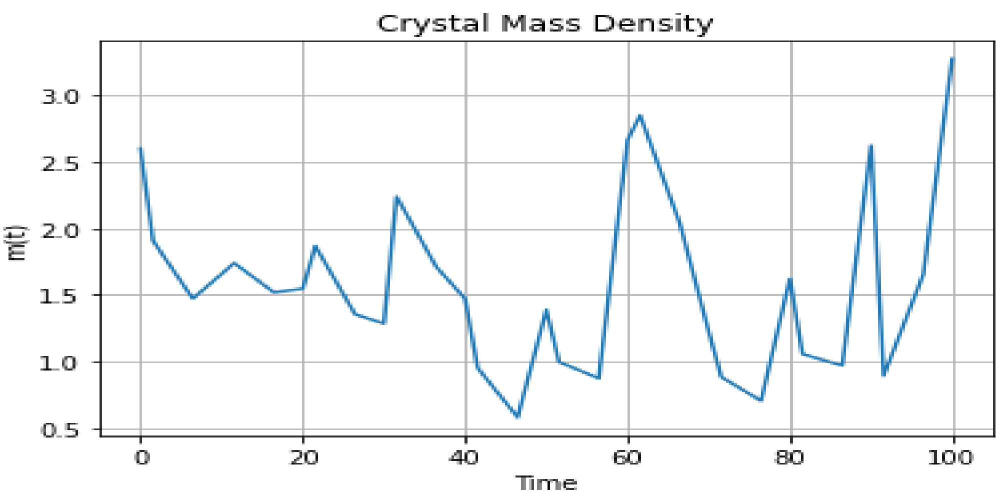

Figure 2a: m(t) vs t no Hopf Constraint

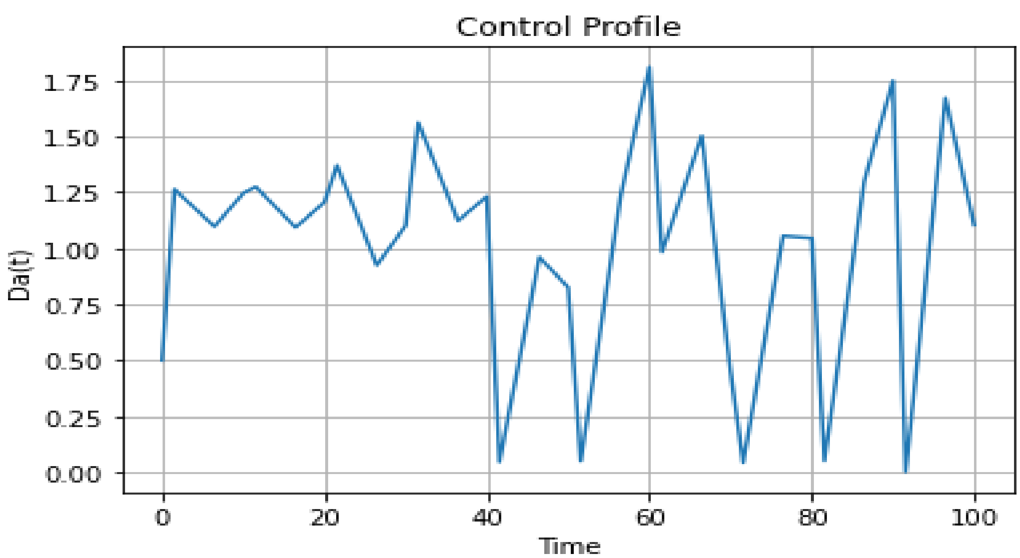

Figure 2b: Da vs t no Hopf Constraint

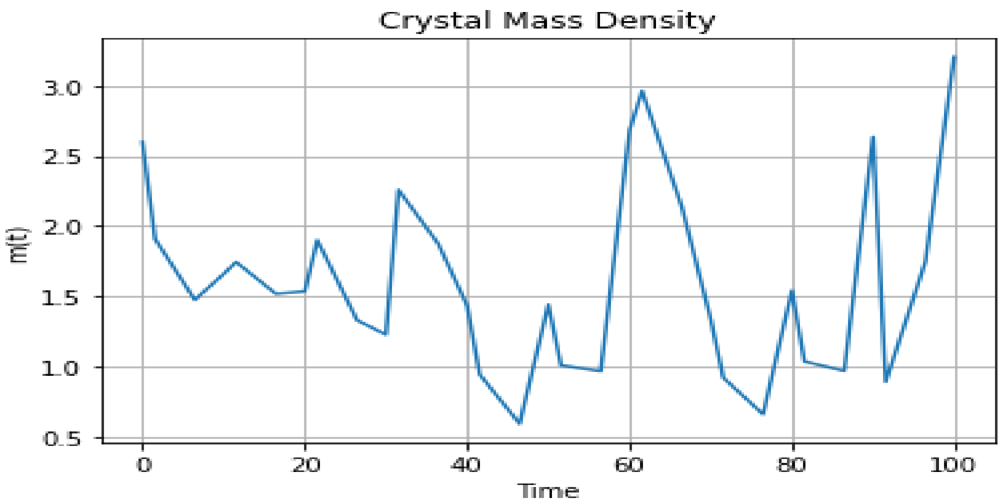

Figure 2c: m(t) vs t with Hopf constraint

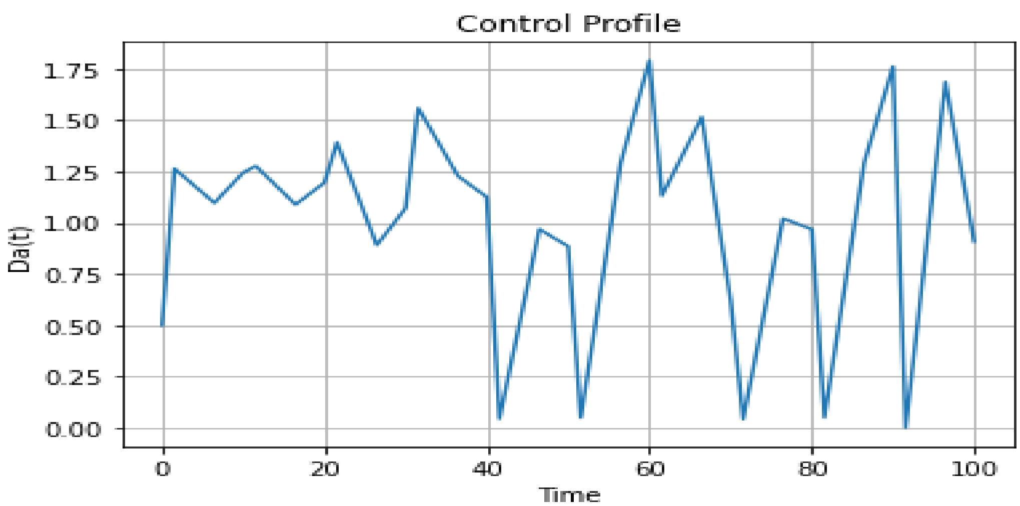

Figure 2d: Da vs t with Hopf Constraint

4. Discussion of Results

The results obtained from the bifurcation analysis and optimal control study provide important insights into the nonlinear behavior and operational performance of reactive crystallization systems under strong coupling between kinetics, transport, and thermal feedback. The combined MATCONT-based bifurcation analysis and Pyomo-based optimal control framework reveal not only the presence of complex dynamical regimes—including steady-state multiplicity and oscillatory behavior—but also how these regimes can be systematically exploited or avoided to enhance industrial performance.

The bifurcation analysis, performed with the Damköhler number (Da) as the continuation parameter, demonstrates that the crystallization system exhibits a rich nonlinear structure [34]. The detection of a limit point indicates the presence of steady-state multiplicity, which is a well-known challenge in industrial crystallizers, as it can lead to abrupt transitions between operating regimes under small perturbations in feed conditions or heat removal rates. More critically, the identification of two Hopf bifurcation points confirms the existence of self-sustained oscillatory behavior in the reactor [35]. These oscillations arise from the strong positive feedback loop between temperature-dependent kinetics and crystal growth, which amplifies perturbations in solute concentration and temperature.

The negative first Lyapunov coefficients associated with both Hopf points confirm that the bifurcations are supercritical, leading to stable limit cycles rather than diverging oscillations [36]. From an industrial perspective, this result is particularly important because it implies that once the system enters the oscillatory regime, it will settle into sustained periodic behavior rather than chaotic instability. While such oscillations are mathematically stable, they are generally undesirable in industrial crystallization processes due to their impact on product consistency, particle size distribution, and downstream separation efficiency.

In practical terms, limit cycle oscillations in crystallization systems lead to periodic fluctuations in supersaturation, which directly affect nucleation and growth rates. This results in oscillatory production of fine and coarse particles, reducing overall product quality and increasing variability in crystal size distribution. In addition, oscillatory thermal behavior increases energy consumption due to repeated heating and cooling cycles, thereby reducing overall process efficiency. These factors highlight the importance of understanding not only steady-state behavior but also dynamic stability characteristics when designing and operating industrial crystallizers [37].

To address these challenges, an optimal control framework was implemented using the Damköhler number as a manipulated variable. Two cases were considered: an unconstrained optimization and a bifurcation-aware optimization incorporating a Hopf-avoidance constraint. In both cases, the objective function was formulated to maximize the cumulative crystal mass density over the operating horizon, which serves as a direct measure of product yield and reactor productivity.

In the unconstrained case, the optimal solution drives the system close to the nonlinear regime where productivity is high but dynamic stability is not guaranteed. This is a common feature in industrial optimization problems, where operating conditions that maximize yield often lie near instability boundaries. However, operation near the Hopf bifurcation region introduces oscillatory behavior that can reduce effective productivity due to periodic depletion and regeneration cycles in solute concentration and temperature. Although instantaneous production rates may be high during certain phases of the oscillation, the time-averaged yield is not necessarily optimal.

The introduction of a Hopf-bifurcation-avoidance constraint significantly alters the optimal operating trajectory. By restricting the system from entering the oscillatory regime, the optimizer is forced to select operating conditions that maintain dynamic stability while still achieving high productivity. Interestingly, the results show that this bifurcation-aware strategy leads to a higher overall objective value compared to the unconstrained case [38]. This improvement highlights an important industrial insight: maximum instantaneous reaction rates do not necessarily correspond to maximum integrated productivity when nonlinear dynamics are considered.

From an industrial perspective, this result has several important implications. First, it demonstrates that dynamic stability constraints can enhance rather than restrict process performance. This contradicts the traditional view in process optimization, where stability constraints are often seen as conservative limitations that reduce achievable performance [39]. In contrast, the present results show that avoiding oscillatory regimes can improve average production efficiency by eliminating inefficient cyclic behavior in the crystallization process.

Second, the results highlight the importance of bifurcation-aware process design. In many industrial crystallization systems, operating conditions are selected based on steady-state optimization without explicit consideration of dynamic instabilities [40]. However, this study shows that such an approach can lead to suboptimal operation, particularly when the system operates near Hopf bifurcation boundaries. Incorporating bifurcation information into the optimization framework allows for the identification of safer and more efficient operating regions that would not be apparent from steady-state analysis alone.

Third, the use of the Damköhler number as a control parameter provides a practical interpretation for industrial implementation. The Damköhler number effectively represents the ratio of reaction kinetics to transport timescales, which can be manipulated in practice through changes in residence time, feed concentration, or flow rate. This makes the proposed optimal control strategy not only theoretically meaningful but also operationally implementable in real crystallization units.

Finally, the study demonstrates the broader value of integrating nonlinear dynamics tools with optimal control methods. The combination of MATCONT-based bifurcation analysis and Pyomo-based dynamic optimization provides a powerful framework for understanding and improving complex chemical processes. This integrated approach enables simultaneous consideration of productivity, stability, and operability, which are all critical factors in industrial crystallization systems.

The results clearly show that nonlinear dynamic phenomena such as Hopf bifurcations and limit cycles are not merely mathematical curiosities but have direct and significant implications for industrial crystallization performance. By incorporating bifurcation-aware constraints into the optimal control formulation, it is possible to enhance product yield, improve operational stability, and reduce inefficiencies associated with oscillatory behavior. This highlights the importance of embedding nonlinear dynamical analysis into the design and optimization of advanced chemical engineering systems.

5. Conclusions

This work investigated the nonlinear dynamics and optimal operation of a reactive crystallization system characterized by strong coupling between solute concentration, crystal population dynamics, and thermal feedback. The model captures key physicochemical mechanisms including nonlinear nucleation, growth kinetics, and Arrhenius-type temperature dependence, which together give rise to rich dynamical behavior such as steady-state multiplicity, Hopf bifurcations, and stable limit cycle oscillations.

Bifurcation analysis performed using MATCONT with the

Damköhler number as the continuation parameter revealed the existence of a limit point and two Hopf bifurcation points. The negative first Lyapunov coefficients associated with both Hopf bifurcations confirmed that the system undergoes supercritical Hopf bifurcations, leading to stable periodic oscillations in reactor states. These oscillations arise from strong nonlinear feedback between temperature-dependent reaction kinetics and crystal growth processes, which amplify perturbations in concentration and thermal states. From an industrial perspective, such oscillatory regimes are generally undesirable as they induce periodic variations in supersaturation, nucleation rate, and crystal growth, ultimately affecting product quality and process consistency.

To address these nonlinear dynamic effects, an optimal control framework was developed with the Damköhler number treated as the manipulated input. The objective was formulated to maximize the cumulative crystal mass production over a finite time horizon, representing overall reactor productivity. Two scenarios were considered: an unconstrained optimal operation and a bifurcation-aware optimal operation incorporating a Hopf-bifurcation-avoidance constraint. The results demonstrated that while the unconstrained case tends to operate the system near the boundary of oscillatory instability, the inclusion of the bifurcation constraint shifts the optimal trajectory toward dynamically stable operating regions.

Interestingly, the bifurcation-aware strategy resulted in a higher overall objective value compared to the unconstrained case. This indicates that avoiding oscillatory regimes can enhance time-averaged productivity by eliminating inefficiencies associated with periodic depletion and regeneration cycles in solute concentration and temperature. The results therefore highlight that dynamic instability, even when mathematically stable in the form of limit cycles, can reduce effective industrial performance.

From an industrial standpoint, the findings emphasize the importance of integrating nonlinear dynamical analysis into process design and optimization. Traditional steady-state optimization approaches may inadvertently drive the system toward instability boundaries, resulting in suboptimal performance when dynamic effects are present. In contrast, the proposed bifurcation-aware optimal control framework provides a systematic methodology to identify and avoid such regimes while still achieving high productivity.

This study demonstrates that explicit consideration of bifurcation structure in reactive crystallization systems can significantly improve operational outcomes. The integration of MATCONT-based bifurcation analysis with dynamic optimization in Pyomo provides a powerful tool for understanding, predicting, and optimizing complex nonlinear chemical processes. The results highlight the potential for improved yield, enhanced operational stability, and more robust industrial crystallizer design by incorporating nonlinear dynamics into control and optimization strategies.

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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