

1 Common concerns

Before addressing each reviewer's comments individually, we would like to discuss two common concerns raised by multiple reviewers: the modeling approach and the parameter sensitivity analysis.

1.1 Modeling approach

The original submission focused on the broader goals of the study, offering a concise explanation of the proposed model. However, based on the reviewers' feedback, we recognized the need to address potential misunderstandings caused by this brevity. To resolve this, we have clarified and justified the modeling approach within this letter, while also updating the manuscript to provide a more detailed and general explanation of the model. These revisions ensure that the manuscript presents a clearer and more comprehensive framework while preserving the original model's intent and structure.

Nevertheless, our work aims to establish a foundation for future research in this area by developing a modeling and control strategy within a general framework. This framework is designed to encompass key features of real-world systems, rather than focusing on a model tailored to a specific system.

For the axial dispersion tubular reactor presented in Fig. 1, the molar balance equation may be written for the reactant concentration, C_A , as follows:

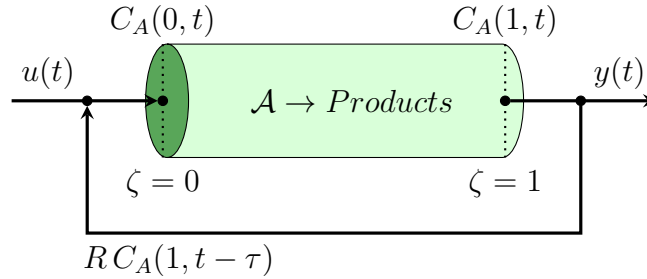


Figure 1: Axial tubular reactor with recycle stream.

$$\frac{\partial C_A}{\partial t} = D \frac{\partial^2 C_A}{\partial \zeta^2} - v \frac{\partial C_A}{\partial \zeta} - r(C_A) \quad (1)$$

where $r(C_A)$ is the reaction rate by which the reactant is consumed. Considering this term can be non-linear, the model can be linearized around the steady-state followed by replacing the state of the system with deviation from the steady-state concentration. This will result in the following equation:

$$\frac{\partial \epsilon}{\partial t} = D \frac{\partial^2 \epsilon}{\partial \zeta^2} - v \frac{\partial \epsilon}{\partial \zeta} - \left(\frac{\partial r(C_A)}{\partial C_A} \bigg|_{C_{A,ss}} \right) \epsilon \quad (2)$$

Here, $\epsilon \equiv C_A - C_{A,ss}$ is the deviation from the steady-state concentration, and $C_{A,ss}$ is the steady-state concentration of the reactant. The model given at this point, although linear, sets a general framework for model-based control strategies of a wide range of infinite-dimensional convection-diffusion-reaction systems in chemical engineering process and dynamics around their steady-state.

The assumptions made in the model are that the parameters of the system are constant and thus, the states of the system are not affected by changes in system's pressure or temperature. The authors acknowledge that the model will be more realistic if the temperature dependence of the reaction rates is included. In fact, addressing temperature dependence is the focus of our ongoing research and represents a natural extension of this work. We anticipate presenting these advancements in future submissions to this journal. Nevertheless, as mentioned above, the primary goal here is to establish a general framework, leaving more specific and physically significant models for subsequent studies.

In addition to the concerns related to the applicability of the model, the choice of positive reaction term is also explained in this section along with stability analysis. The stability analysis of the system in the vicinity of the steady state has been one of the first things of our interest. While no isothermal reactor can technically be exponentially unstable due to the finite amount of reactant available, the steady state of the system can be unstable in the vicinity of the steady state, meaning that a deviation from the steady state may draw the system to a different steady state, resulting in completely different model dynamics than the one used to design and control the system for optimal operation.

Performing the eigenvalue analysis of the system, we have observed that a system may have unstable steady-state only when the reaction term coefficient $-\frac{\partial r(C_A)}{\partial C_A}$ is positive around a given steady-state. Though an uncommon scenario, this may happen in the case of autocatalytic reactions, enzyme-catalyzed reactions, reactions that incorporate inhibition mechanisms, etc. Although the proposed controller can also stabilize an already stable system in an optimal manner, a positive reaction term is selected to demonstrate the ability of the proposed controller to stabilize a system which is mathematically unstable, rather than focusing on a physically significant model. This has to be included within the general framework of the controller proposed in this work as it becomes more significant when the work is extended to more complex systems where instability becomes a common issue.

In summary, the revisions address the reviewers' concerns by expanding the modeling section, clarifying assumptions, and adding relevant analysis to enhance the work's depth and applicability. By doing so, we have strengthened the explanation of the proposed framework and its ability to handle intrinsic delays in distributed parameter systems. We believe these changes make the manuscript more robust and provide a solid foundation for advancing research in chemical engineering process control and dynamics.

1.2 Parameter sensitivity analysis

We acknowledge how all the reviewers are interested in at least one aspect of parameter sensitivity analysis, which was also of great interest to the authors during the initial simulations.

At that stage, we tested numerous parameter sets and observed interesting results, such as how increasing or decreasing the Peclet number causes the spectrum to shift towards real or purely imaginary eigenvalues, or how increasing the recycle ratio mimics the behavior of a CSTR. However, we chose not to include these results in the original manuscript to maintain the focus of our work, which is to introduce a modeling technique for incorporating state delay into a distributed parameter system, followed by developing a control strategy based on the obtained infinite-dimensional model using a late-lumping approach.

Our observations showed that while parameter variations produced interesting modeling results for the open-loop system, they did not impact the general strategy proposed in this work. By this, we mean that for any parameter set, the system's spectrum can be determined by solving the eigenvalue problem in the same manner. Once the spectrum is obtained, the controller can be designed by solving the Operator Riccati Equation (ORE) in the same way, which stabilizes the system by selecting a sufficient number of eigenmodes and appropriately tuning the controller parameters in the objective function. While solving the characteristic equation or the ORE may become more computationally intensive for extreme parameter choices, it remains feasible, ensuring that the proposed strategy can be applied effectively across a wide range of parameter sets.

The proposed set of parameters was chosen to demonstrate the applicability of our approach. Specifically, we selected a parameter set that not only incorporates all the key properties of the claimed system—diffusion, convection, reaction, recycle ratio, and recycle delay—but also results in an unstable system, as explained earlier in this letter. This deliberate choice highlights the controller's ability to stabilize a system that combines all these properties while exhibiting challenging dynamics, showcasing the practicality and effectiveness of the proposed strategy in addressing intrinsic delays and instability.

Nevertheless, as an additional enhancement to our work, we have included an analysis of how the controller's performance varies with different delay values, among other parameters. The authors admit that this addition strengthens the work by satisfying the natural curiosity of readers, as the delay is central to this study, while remaining within the scope of the research. It also provides deeper insights into the controller's performance and demonstrates its adaptability in addressing state delay, contributing to the advancement of distributed parameter systems in chemical engineering process control and dynamics. The manuscript has been updated to reflect this addition in the results and discussions, as well as the conclusions sections.

Reviewer 1

The comments of Reviewer 1, along with our responses to each comment, are included below:

“This work presents a boundary optimal control strategy for axial tubular reactors with first-order irreversible chemical reaction incorporating a delayed recycle stream. The mathematical description takes the form of a system of coupled parabolic and hyperbolic PDEs. An infinite-dimensional approach is applied to derive a linear quadratic regulator with and without observer. Numerical studies show that the proposed controller is able to stabilize the system. The manuscript is clear and well written and addresses a challenging control problem in chemical engineering. The following comments and suggestions may improve the presentation so that it is considered for publication.”

1. Page 3 - “Many chemical, petrochemical, and biochemical unit operation processes are modelled as distributed parameter systems (DPS).”

A few specific examples of these chemical processes would help to motivate the problem this paper addresses

Authors Reply:

Examples have been added to the introduction and the manuscript has been revised accordingly.

2. Page 5 - “PIDEs”

- What does PIDEs stand for?

Authors Reply:

Thanks for pointing this out. The acronym should have been defined in the manuscript. It stands for Partial Integro Differential Equations. The manuscript has been revised accordingly.

3. Page 5 - “a configuration common in industrial processes”

- Examples of these processes would illustrate the need and motivation to address the associated control problem

Authors Reply:

Examples have been added to the introduction and the manuscript has been revised accordingly.

4. Page 6 - “The resulting PDE that describes the reactor model is given by:”

- I suggest to specify the assumptions leading to the reactor model, like isothermal operation, constant properties, constant pressure, ...

Authors Reply:

We have assumed that the parameters of the system are constant and thus, the states of the system are not affected by changes in system's pressure or temperature. The manuscript has been updated to include more detailed explanation in this regard.

5. Page 7 - Eq (2)

- What is the physical meaning of the manipulated variable $u(t)$?

Authors Reply:

It is of the same nature as the system's states, i.e. the manipulated variable $u(t)$ may be seen as either the product concentration or as its deviation from the steady-state concentration in the reactor feed stream. The manuscript has been updated to address this point.

6. Page 7 - “ $x_2(\zeta, t)$ is introduced as a new state variable to account for the concentration along the recycle stream”

- Why is x_2 a function of ζ ? How does x_2 change across the recycle? What kind of law does it follow? To me, it seems like x_2 only changes with respect to time

Authors Reply:

We have assumed that along the recycle stream, the flow of the reactant belongs to the class of pure transport PDEs. This is the case when diffusion term and reaction term become negligible compared to the convective term, and the state variable x_2 is merely being transported along the recycle stream. Nevertheless, this still means that x_2 is a function of ζ and t . Concentration profile along the recycle stream has not been presented in the original submission as we believe it is not the focus of this work. However, for the reviewer's reference, it is included here in the response as shown in Fig. 2.

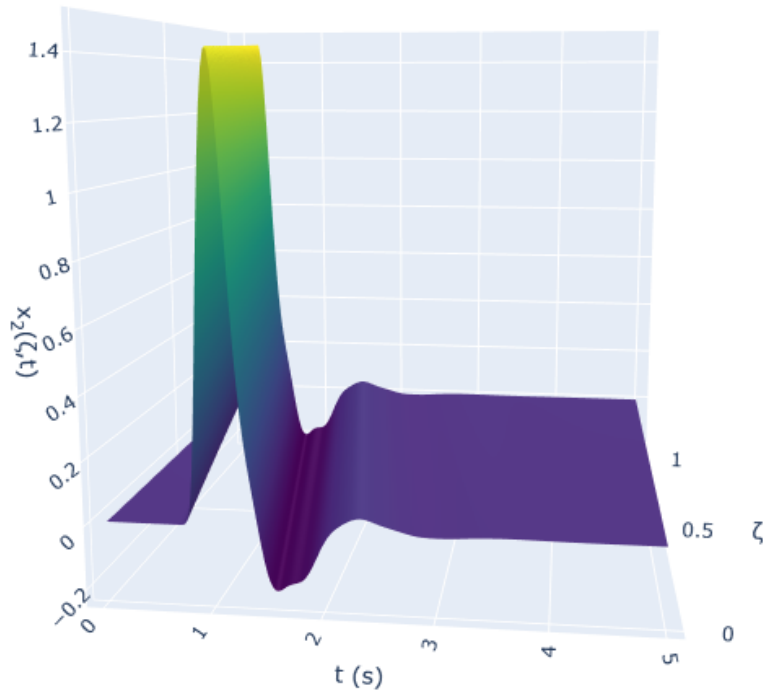


Figure 2: Concentration profile along the recycle stream

7. Page 8 - Eq (5)

- Please make a distinction between the symbol for domain and for diffusivity to avoid confusion

Authors Reply:

Thanks for mentioning this. We have replaced the symbol for the domain with \mathcal{D} to avoid confusion. The manuscript has been updated accordingly.

8. Page 12 - Riesz-spectral operator \mathfrak{A}

- Although a rigorous proof is not necessary, it would be good if the authors explain why \mathfrak{A} is a Riesz-spectral operator. I guess its eigenvalues and eigenfunctions satisfy the requirements, like having multiplicity one and forming a Riesz basis, respectively.

Authors Reply:

That, along with the fact that the obtained set of eigenfunctions for \mathfrak{A} and \mathfrak{A}^* seem to form a bi-orthogonal basis that can successfully span the state space, is the reason behind this statement. It is worth mentioning that as the result of the nature of boundary conditions, this can not be analytically proven. However, once the

spectrum for the system generator and its adjoint is obtained and the eigenfunctions are properly scaled, we observe that the aforementioned properties are satisfied. To be more specific, the inner products of ϕ_i and ψ_j gives $\delta_{i,j}$, and every arbitrary function that satisfies the domain may be expressed as a linear combination of the given bi-orthogonal basis. In the manuscript, we have avoided going into the details to keep the flow of the paper.

9. Page 12 - \mathfrak{R} being positive semi-definite operator
- Should \mathfrak{R} be self-adjoint and coercive?

Authors Reply:

Yes. According to [1] for the infinite-dimensional LQR problem on the infinite-time interval, operator \mathfrak{R} should be self-adjoint and coercive, which is a better way to address the operator compared to self-adjoint and positive semi-definite. The manuscript has been updated accordingly.

10. Page 12 - The LQR problem
- Is Problem 12 well-posed, does J have a finite value for at least one u ?

Authors Reply:

Yes, the proposed infinite-time LQR problem is well-posed, i.e. for at least one input trajectory $u(t)$, the cost function J has a finite value. The stabilizing input trajectory obtained in the manuscript is an example.

11. Page 18 - unstable dynamics of the model
- Why is the zero-input response considered unstable? Is it a qualitative assessment? What is the physical meaning of Figure 9?

Authors Reply:

The open-loop system being unstable is the result of the reaction term being positive, which opens up an opportunity for deeper analysis to explore underlying mechanisms and implications. We basically picked the positive reaction term to demonstrate the ability of the proposed controller to stabilize a mathematically unstable system. However, the physical significance of the system described by such a model may be further explored by focusing on the reaction kinetics and first principle modeling.

The reaction kinetics can be seen in two ways: first, the state of the system being described by the reactant concentration, and second, the state being described by

the product concentration.

1. $x_1 = C_A$: In this case, the reaction is either a first-order reaction, or the reaction rate is a non-linear function of the state. In the latter case, the non-linear system can be linearized around the steady-state followed by replacing the state of the system with deviation from the steady-state concentration. In the obtained model, the reaction constant parameter k_r shall be negative as long as the state of the system is the reactant concentration. This is because the reactant is being consumed over time.
2. $x_1 = C_B$: In this case, the reaction rate is generally a nonlinear function of the product concentration. Same as previous, the system can be linearized around the steady-state followed by replacing the state of the system with deviation from the steady-state concentration. In the obtained model, the reaction constant parameter k_r shall be positive as long as the state of the system is the product concentration. Unlike the previous case, this is because the product is being generated over time. In a rare case, such as auto-catalytic reactions where the reactant is available in excess, the reaction rate can be directly expressed as a linear function of the product concentration as long as the excess reactant assumption holds.

Since the source of instability according to the above, is merely the accumulation of the products, it is rare to have an unstable system in practice. Because even if the rate law determines the rate of product accumulation increases with the product concentration, the system will eventually run out of reactants and the reaction will stop. In fact, most unstable reaction systems are due to the temperature dependence of the reaction rates, resulting in runaway reactions.

Nevertheless, the key contribution of this work lies in introducing a control strategy based on a novel approach to address the problem of intrinsic delay imposed by the recycle stream. This strategy forms a foundation for future research and development for the general case of second order parabolic PDE systems, rather than focusing solely on a model with realistic physical significance. As mentioned earlier, the motivation behind the choice of positive reaction term in the model is to demonstrate the ability of the proposed controller to stabilize a system which is mathematically unstable, at least in the vicinity of its steady state where the linearization is valid.

We have adjusted the manuscript briefly to better explain what it means for the system to be unstable in this context. Future works will extend the same control and modeling strategy to address physically significant unstable reaction systems by including the assumption of temperature dependence for the reaction rates.

12. Page 18 - "The goal is to stabilize the system using an optimal control strategy"
 - What are the values of matrices Q and R in the objective function?

Authors Reply:

Thanks for pointing this out. The values for operators \mathfrak{Q} and \mathfrak{Q} in the objective function were not explicitly mentioned in the manuscript. These values are chosen as $\mathfrak{Q} = 0.05 \times \mathfrak{I}$ and $\mathfrak{R} = 50$, where \mathfrak{I} is the identity operator of the same size as \mathfrak{A} . The manuscript has been updated accordingly.

13. Page 18 - “The goal is to stabilize the system using an optimal control strategy”

- Are x deviation variables? What is the setpoint?

Authors Reply:

In addition to our reply to question 11, since the reaction term is positive in the proposed PDE, the state of the system may be seen as either the product concentration or as its deviation from the steady-state concentration. The setpoint will be either zero or the steady-state concentration of the product, respectively. Again, the goal of this work is to establish an optimal controller strategy to stabilize a system described by the proposed model, rather than tracking a physically significant setpoint.

14. Page 20 - “Both optimal feedback gains are able to successfully stabilize the system within finite time horizon.”

- It would be interesting to see how the delay time affects the stabilizing capabilities of the feedback regulator. What would happen a shorter and larger values of τ

Authors Reply:

It is indeed interesting to see the effect of system parameters (e.g. recycle delay, τ , or recycle ratio, R) on the spectrum of the system as well as the controller performance. We have explored numerous scenarios in the numerical simulations and observed that increasing the recycle ratio, R , or the recycle delay, τ , may lead to more challenging control problems. However, the proposed controller is able to stabilize the system for a wide range of system parameters with exactly the same strategy. Since exploring different system parameters appeared to add no direct significant value to the main contribution of the paper, we have decided to keep the focus on the proposed control strategy in the original manuscript. Nevertheless, if the reviewer finds it necessary, we can include a new plot profile in the revised manuscript where the closed-loop system state is plotted for one or two different values of τ and/or R .

15. Page 26 - “The proposed framework may be extended to more complex diffusion-convection reactor configurations, such as non-isothermal reactors”

- Can this framework be applied to reaction systems described with more complex and

highly nonlinear reaction kinetics?

Authors Reply:

Yes, it can. The general approach would be the same as the one presented in [2], where they assumed the reaction constants to be exponential functions of the temperature. The same approach can be applied to the proposed model in this work in future research.

Reviewer 2

The comments of Reviewer 2, along with our responses to each comment, are included below:

“The problem presented in the article is well-posed and written. The purpose is to present a novel approach to address the problem of intrinsic delay when there is a recycle stream in a process. However, I recommend submitting it to a journal focused on Control. This opinion is based on the following concerns regarding the process of a chemical reactor with recycle:”

1. Why is relevant to consider Danckwerts boundary conditions for the problem of control? Have you compared your results with those obtained considering other boundary conditions?

Authors Reply:

Apart from time-dependent boundary conditions, chemical engineering applications commonly use Dirichlet, Neumann, and Robin boundary conditions, with Robin being the most general. The Danckwerts boundary condition, as a type of Robin condition, maintains generality and captures physical significance without simplifying the problem.

This condition is particularly suited for systems with both diffusion and convection, like chemical reactors, as it reflects the boundary behavior where these transport phenomena interact. While other boundary conditions are possible, they may lack the physical relevance and generality offered by the Danckwerts condition.

2. Concerning the recycle stream, have you studied the effect of the R , the recycle ratio, on your results? Please comment.

Authors Reply:

It is interesting to see the effect of system parameters (e.g. recycle delay, τ , or recycle ratio, R) on the spectrum of the system as well as the controller performance. We have explored numerous scenarios in the numerical simulations and observed that decreasing the recycle ratio, R , or the recycle delay, τ , may lead to more straight forward control behavior, and vice versa.

However, the proposed controller is able to stabilize the system for a wide range of system parameters with exactly the same strategy. Since exploring different system parameters appeared to add no direct significant value to the main contribution of the paper, we have decided to keep the focus on the proposed control strategy in the original manuscript. Nevertheless, if the reviewer finds it necessary, we can include a new plot profile in the revised manuscript where the closed-loop system state is plotted for one or two different values of τ and/or R .

3. The authors used as the case study the problem of an axial dispersion tubular reactor incorporating diffusion, convection, and a first-order irreversible chemical reaction described by equations (1)-(2). While this is sufficient to present their approach, it is far from being extended to the more general problem, non-isothermal, and with more general kinetics such as biochemical or catalytic.

Authors Reply:

Thank you for your insightful comment. While we acknowledge that temperature dependence can significantly impact the dynamics of chemical reactors, and we plan to address non-isothermal cases in future work based on the foundation laid by this study, we would like to highlight several key points regarding the current model and its general applicability.

The main focus of our work is to develop an infinite-dimensional control strategy with introducing state-delays in such distributed parameter systems for the first time, with no need for model reduction or spatial discretization. Nevertheless, the infinite dimensional system of the axial dispersion tubular reactor model defined by a second order parabolic PDE and Danckwerts boundary conditions, equipped with a recycle stream imposing state-delays, captures the essential structure of many advanced systems with similar dynamics; while the proposed control strategy is general and sets the stage for future extensions to more complex systems, such as temperature-dependent reaction rates.

Even in the cases where the reactor model is replaced with a more complex set of PDEs than the presented second-order parabolic PDE, the proposed strategy of incorporating delayed recycle stream to the system's model and the infinite-dimensional control strategy design can still be applied. Hence this work can be seen as a stepping stone towards addressing more complex systems in the field of chemical engineering, utilizing the key ideas presented in this study.

4. I recommend submitting it to a journal focused on Control.

Authors Reply:

Thank you for your suggestion regarding submitting this work to a journal focused on control. However, we would like to emphasize that similar research [3, 4] has been recently published in this journal, demonstrating contributions to control theory that align with those presented in our work. This validates the relevance of publishing control-focused studies within chemical engineering journals, which often encompass modeling and novel methodologies pertinent to the field.

Furthermore, while this work is centered around proposing a control strategy, its significance is rooted in the unique context of chemical engineering, where state de-

lays are rarely considered. Although state delays have been extensively studied in other fields with different system models, applying such a strategy to chemical engineering systems—particularly those convection-diffusion-reaction DPSs described by second order PDEs—is novel and valuable. This ensures that the work contributes meaningfully to the control theory within the field of chemical engineering.

Reviewer 3

The comments of Reviewer 3, along with our responses to each comment, are included below:

“In this work, the authors address the optimal control of an axial tubular reactor with a recycle stream. They model the intrinsic time delay from the recycling process using a system of coupled parabolic and hyperbolic partial differential equations. The control input is applied at the inlet, and a continuous-time optimal linear quadratic regulator is designed to stabilize the system. Numerical simulations indicate effective full-state feedback regulator and observer-based regulator. This work presents an interesting methodology but there are some minor considerations that the authors need to address before this article can be published.”

1. **Major comment:** In section 4, the authors indicate that they discretized each state in space using 100 grid points. They must indicate if those points are equidistributed and how they came up with such discretization grid. Note that multiple works [5, 6, 7] have demonstrated that the selection and distribution of the discretization grid plays a crucial role in the computation of optimal control laws, i.e., a control law can be claimed to be optimal or not using the criterion of the Pontryagin’s Minimum Principle (PMP). The reviewer recommends to assess the criterion of the Hamiltonian function (i.e., the PMP) to demonstrate that the discretization implemented is accurate and the solutions obtained for the control are optimal.

Authors Reply:

Thanks a lot for the insightful and detailed comment. We have gone through the suggested literature and agree that the selection and distribution of the discretization grid are crucial in converting an infinite-dimensional system to a finite-dimensional one. However, we would also like to point out that based on the late-lumping approach used in the proposed work, we do not discretize the system in space at all. In fact, the infinite-dimensional nature of the system is fully captured in the proposed control strategy, and the control law is designed directly in the infinite-dimensional space.

The 100 grid points mentioned in the manuscript are merely used to confirm how the optimal control input will be applied to the system. In other words, the feedback gain is designed based on the infinite-dimensional system, and the control input is obtained with no need for spatial discretization. It is only at this point that the obtained control input is applied to a FDM representation of the system to evaluate the system’s response to the control input.

2. The manuscript lacks conclusions or further discussion about the control trajectories’ results, such as the quality of the control actions or improvements in process operation (e.g., avoidance of constraint violations, disturbance rejection, etc.). Although the au-

thors included several figures illustrating the process dynamics and control trajectories, these are not discussed in sufficient depth, i.e., avoid leaving the reader to draw their own conclusions from the figures. Additionally, the reviewer recommends reducing the number of figures, which could allow more space for further discussion of the results.

Authors Reply:

The reviewer's suggestion is well taken. We will include a more detailed discussion of the results in the revised manuscript. However, aspects like constraint violations and disturbance rejection usually falls out of the scope of the proposed control strategy when it comes to optimal regulatory problem. This work sets the mathematical foundation for future works to address these aspects under different control strategies (such as observer-based or Kalman-filter-based discrete-time MPC) along with more realistic assumptions in the model (such as adding the temperature dependence of the reaction rates).

3. In section 3.1.3, the authors present the values for parameters R and D but provide no further details about the model's sensitivity to these parameters. Please include a detailed explanation of how these values were selected and discuss any potential limitations if the parameters are chosen incorrectly.

Authors Reply:

It is indeed interesting to see the effect of system parameters (e.g. recycle delay, τ , recycle ratio, R , or the physical parameters within the reactor) on the spectrum of the system as well as the controller performance. We have explored numerous scenarios in the numerical simulations and observed that decreasing the recycle ratio, R , or the recycle delay, τ , may lead to more straight forward control behavior, and vice versa.

However, the proposed controller is able to stabilize the system for a wide range of system parameters with exactly the same strategy. The only limitation we faced in this regard is the computation cost of the numerical simulation, especially when it comes to obtaining the solution to the eigenvalue problem, mostly due to calculation errors in getting the matrix exponentials.

Since exploring different system parameters appeared to add no direct significant value to the main contribution of the paper, we have decided to keep the focus on the proposed control strategy in the original manuscript. Nevertheless, if the reviewer finds it necessary, we can include a new plot profile in the revised manuscript where the closed-loop system state is plotted for one or two different values of τ and/or R .

4. In section 4, the authors indicate that the process model was discretized in time and space, however, they mention that they obtained a system of ordinary differential equa-

tions (ODEs). Please clarify how this full discretization resulted in a system of ODEs.

Authors Reply:

Thank you for pointing this out. As mentioned previously, the discretization in Section 4 is merely used to evaluate the system's response to the control input. Yet, there is a mistake in the manuscript as there is no discretization in time at this point. The system of ODEs is obtained as a result of applying space discretization to the infinite-dimensional system, where at each node the state of the system is represented by an ODE in time. We will revise the manuscript to clarify this point.

5. The manuscript has some typos that the authors must correct, e.g., ...setting for of distributed..., Then two full-state... In page 5, the acronym PIDEs is not previously defined. For section 4.3, the reviewer recommends to modify the expression "Last but not least" aiming not loose the formality of the manuscript.

Authors Reply:

Thank you very much for the detailed feedback. All of the suggestions ahve been addressed in the revised manuscript.

6. The reviewer recommends to include a tables of nomenclature

Authors Reply:

A table of nomenclature has been included in the revised manuscript.

References

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