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Stabilizing Unstable Operation Regimes of Cooled Exothermic Fixed-Bed Reactors

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Highlights

- Detailed dynamic two-dimensional model used for fixed-bed reactor analysis.
- Model-based investigation of unstable operation regimes induced by parametric sensitivities.
- NMPC methodology to uncover and stabilize unstable operation regimes.
- Operating in unstable regimes significantly improves catalyst lifetime.

1. Introduction

Since more than 50 years, the large variety of different applications for exothermic fixed-bed reactors reported in literature gives rise to a continuously ongoing interest in improving their performance and reliability [1]. So far, the major objective was to guarantee steady, safe, and efficient operation. Non-steady (e.g., periodic) operation concepts have been investigated, but found minor attention for real commercial applications due to increased technical requirements. However, the current trend towards a more flexible production to react on markets as well as on volatile inputs (e.g., coming from renewable sources), reveals many new opportunities for non-steady reactor operation. Furthermore, modern high performance catalysts, each tailored for specific reactions, became very active and selective, but also very sensitive in terms of temperature resistance. Thus, the interest in detailed predictions on the reactor dynamics has become more important than ever before. Computational models are capable to deliver this level of detail in a reasonable amount of time and allow for estimating first general trends and potentials of dynamic reactor operation [2].

This work exemplarily deals with a fixed-bed reactor for CO_2 methanation. This reaction is strongly exothermic such that distinct hot-spots are formed that can strongly influence the catalyst durability and process safety. In addition, this process is a promising route for storage of electrical surplus energy harvested from volatile renewables (e.g., wind, solar). In this context, looking at the reactor dynamics and controllability allows to identify unconventional operation regimes, as illustrated below.

2. Methods

Our study follows up the two-dimensional, pseudo-homogenous, tubular reactor model for CO_2 methanation described in [2]. The reactor operation is mainly controlled by the adjacent temperature of the cooling medium, which is further on denoted as cooling temperature T_{cool}. All calculations are implemented and performed in MATLAB to ensure a flexible and efficient access to advanced control strategies (e.g., Nonlinear Model Predictive Control) later on. The results are evaluated in terms of CO_2 conversion and CH_4 selectivity at the reactor outlet as well as the spatial temperature distribution inside the reactor.

3. Results and discussion

In the first step, we characterize the reactor by cooling temperature step changes with subsequent simulation into the resulting steady-states. Depending on the reactor's history multiple steady-states are observable which give rise to hysteresis behaviour (see Fig. 1), similar to the observations reported in [1,3,4]. Depending on the operation regimes the derived detailed reactor model also allows the prediction of the temperature distribution in two space dimensions, namley radial and axial (see Fig. 2). We found the local displacement of the main reaction zone and investigated its sensitivity with respect to essential model parameters (e.g., heat transfer coefficients, feed composition, GHSV).

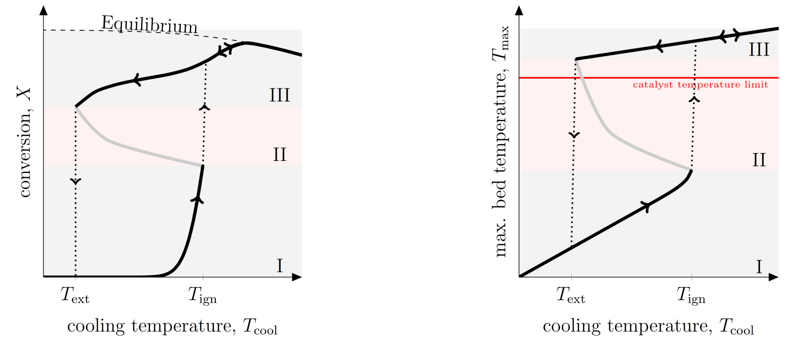

Figure 1. Hysteresis behaviour and operation regimes for conversion (left) and maximum bed temperature (right); (I) **stable** low conversion regime; (II) **unstable** intermediate regime; (III) **stable** high conversion regime; Arrows indicate the reactor's history.

As shown in Fig. 1, the reactor ignition and extinction points limit the range of stable operation regimes (I and III), such that under some scenarios (e.g., in case of a methanation reactor) the productive regime III might not be feasible due to insufficient catalyst temperature resistance. At this point, dynamic control becomes vital to stabilize regime II and, thus, to increase the amount of practical operation regimes. However, using conventional control strategies (e.g., PID control) might fail, due to the strong nonlinear nature of the reactor. The remedy to address this issue is Nonlinear Model Predictive Control (NMPC), which effectively stabilizes regime II, and, thus, will be explored in more detail in this contribution.

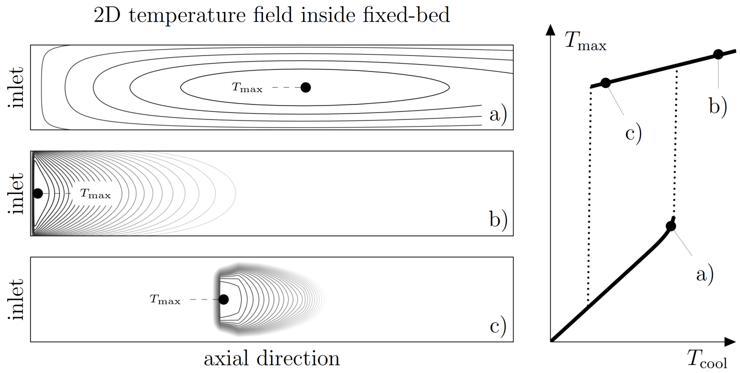

Figure 2. Temperature field at different stable operation regimes; contours decrease in 1%-steps of T_{max}.

4. Conclusions

In the near future, more flexible chemical production scenarios will demand for dynamic reactor operation. In this context, the key requirement is to combine well established knowledge on reactor dynamics and modern tools of computational mathematics and control. Following this idea, we demonstrate how to uncover and stabilize unstable operation regimes of exothermic fixed bed reactors to widen the operation range and enhance the durability of catalysts in these reactors.

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Keywords fixed-bed reactor, steady-state multiplicity, nonlinear model predictive control, methanation