PROCESS INTENSIFICATION BY MODEL-BASED DESIGN AND OPTIMAL OPERATION OF TAILOR-MADE REACTORS

Hannsjörg Freund
Technische Universität Dortmund, Lehrstuhl Reaction Engineering and Catalysis
44227 Dortmund, Germany

Abstract

This contribution provides an overview of our work on the model-based design and operation of catalytic reactors aimed at developing energy and resource efficient chemical processes. Special attention is given to the methodology of multi-level reactor design (MLRD) that follows the key idea to track a fluid element on its way through the – not yet specified – reactor and optimizing the reaction conditions along its way by providing the necessary material and energy fluxes. Based on the resulting flux profiles, optimal and novel reactor concepts tailored to the needs of the reaction system can be derived and analyzed. Such concepts include the use of structured catalyst supports in the form of additively manufactured periodic open cellular structures (POCS). The MLRD approach enables the predictive determination of the best reaction concept considering highly innovative process intensification options. Thereby, this method is a cornerstone for the development of more economical and more sustainable chemical reactors and processes for the future.

Keywords

Multi-Level Reactor Design, Structured Catalysts, Optimization

Introduction

Our work aims at the development of energy and resource efficient chemical processes with a special focus on the model-based design and optimal operation of catalytic reactors. In such a comprehensive approach innovative process-, reactor- and material concepts are considered, which includes the evaluation of different process intensification options. In contrast to incremental process improvements, process intensification aims at fundamental step changes in productivity and efficiency. To achieve this, an interdisciplinary, method- and model-based approach is essential. Supporting target-oriented key experiments are carried out for phenomenological elucidation, data retrieval and model validation. The combination of powerful simulation tools at different scales and at different times during the design process allows for linking the process-, reactor- and material design. That way it is possible to identify tailor-made solutions based on rigorous model-based optimization.

To achieve this goal, we conduct research in three main areas, all of them with the clear goal of enabling and achieving process intensification.

The central topic and methodological backbone of our work is the so-called multi-level reactor design (MLRD) methodology. Since this is a rigorous model-based design approach, one key aspect for success in its application is the availability and implementation of suitable reaction kinetic models to investigate the desired reaction conditions with necessary adequacy. Another component to realize these reaction conditions are optimized structured catalyst supports and reactors that can be fabricated using additive manufacturing techniques.

Multi-Level Reactor Design: Achieving the Optimum

As a central process unit, it is highly important that the reactor imposes the optimal reaction conditions along the

reaction route. The decisive facet of the MLRD methodology is the start from a fundamental and rigorous function-based investigation on the underlying principles and process tasks for reactors and process equipment (Freund and Sundmacher, 2008). From this more abstract point of view, process intensification can aim at obtaining the optimal integration and enhancement of individual fluxes (mass, energy, momentum and reaction fluxes). On the different levels of the approach, the optimal reaction conditions are first identified and then approximated by suitable reactor concepts (Freund et al., 2019, Peschel et al., 2010). With recent extensions of this approach, optimization under uncertainty, e.g. for variability in the feed composition and dynamic reactor operation with fluctuations in the feed flowrate can also be considered.

Model Selection: Adequacy is Key

In model-based optimization, feasible models with high accuracy and a large range of validity are necessary. Our particular interest here is to identify and use the most adequate level of detail for the model (Kaiser and Freund, 2019). This is especially true for the underlying intrinsic reaction kinetic model since it has a high impact on the optimal conditions determined in the first stage of the MLRD methodology. As one example, we developed a new model approach of lower complexity than the typical microkinetic approaches, yet able to capture the dynamics of the catalyst coverage in contrast to established steady-state kinetic model approaches and employed it for the optimization of reactors for the dynamic CO_2 methanation.

The aspect of model adequacy is further addressed for transport phenomena. As an example, a novel fixed-bed heat transport correlation was developed that is well suited for reactor optimization because it retains comparable accuracy to established models while achieving improved numerical manageability (Pietschak et al., 2020).

Additive Manufacturing: Form Follows Function

For the realization of the optimal reaction conditions identified using the MLRD approach, the effective transport properties of the catalytic bed have to be controlled and adjusted. In this context, the use of structured catalyst supports offers a high degree of freedom for a more efficient and flexible geometry design. A prime example is the intensification of the heat transport in catalytic fixed-bed reactors by utilizing optimized catalyst carrier structures. Busse et al. (2018) used periodic open-cellular structures (POCS) as catalyst carrier and benchmarked them with classic randomly packed fixed-bed reactors in terms of heat transfer. Due to the continuous solid matrix and the resulting enhanced heat conductivity on the reactor scale, they outperform classical packed bed systems and thus represent a promising option for process intensification with respect to the energy flux.

In this regard, additive manufacturing is an enabling tool for the technical realization of such optimized catalyst supports, which can be tailor-made to the specific needs of the reaction system at hand. Owing to the direct link between geometrical parameters and resulting properties that influence the reactor performance, various classes of structures can systematically be evaluated and even new structures for improved reactor performance can be derived.

Conclusions

Process intensification requires a holistic view of the process and a focus on process functions rather than on unit operations. The task is to find the optimal combination of fluxes (mass, energy, momentum and reaction fluxes) along the process route. The MLRD methodology for optimal reactor design utilizes such a function-based approach and thus addresses exactly the above-mentioned identification of the optimal combination of fluxes by means of dynamic optimization.

The MLRD methodology is perfectly suited to identify optimal reactor concepts, as demonstrated for a number of reaction systems of different level of complexity. An increase in complexity can arise from system specific and inherent requirements, e.g. the necessity to consider transport processes in more detail (mass transfer between phases, intraparticle processes, catalyst with deactivation etc.). Another source of complexity comes from external factors, in particular from the raw material shift towards the increased use of renewable feedstocks and energy from renewable sources, both of which are subject to fluctuations with respect to time. Ongoing and future work must address the identification of solutions that balance flexibility, robustness and performance of future processes in an optimal manner.

References

Busse, C., Freund, H., Schwieger, W. (2018). Intensification of heat transfer in catalytic reactors by additively manufactured periodic open cellular structures (POCS). *Chem. Eng. Process., 124,* 199-214.

Freund, H., Sundmacher, K. (2008). Towards a methodology for the systematic analysis and design of efficient chemical processes. From unit operations to elementary process functions. *Chem. Eng. Process., 47,* 2051-2060.

Freund, H., Maußner, J., Kaiser, M., Xie, M. (2019). Process intensification by model-based design of tailor-made reactors. *Curr. Opin. Chem. Eng., 26,* 46-57.

Kaiser, M., Freund, H. (2019). A multimodular pseudo-heterogeneous model framework for optimal design of catalytic reactors exemplified by methanol synthesis. *Chem. Eng. Sci., 206,* 401-423.

Peschel, A., Freund, H., Sundmacher, K. (2010). Methodology for the Design of Optimal Chemical Reactors Based on the Concept of Elementary Process Functions. *Ind. Eng. Chem. Res., 49*, 10535-10548.

Pietschak, A., Dixon, A. G., Freund, H. (2020). A new heat transfer correlation suited for the design of fixed-bed reactors via numerical optimization. *Chem. Eng. Sci., 220,* 115614.