Abschlussbericht für die Max-Buchner-Forschungsstiftung

Thema: Optimaler Entwurf heterogen-katalytischer Reaktoren unter Berücksichtigung der Katalysatordesaktivierung

Optimal design of heterogeneous catalytic reactors considering catalyst deactivation

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Antragsteller: Prof. Dr.-Ing. Hannsjörg Freund

Doktorand: M.Sc. Mingquan Xie

Institution: Lehrstuhl für Chemische Reaktionstechnik

Friedrich-Alexander-Universität Erlangen-Nürnberg

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Abstract

Catalyst deactivation has been incorporated into an optimal reactor design methodology to develop tailor-made reactor concepts, with the goal of achieving the best overall performance over the whole catalyst lifetime. The reactor set-up and varying operating conditions were simultaneously optimized. Reactor configurations with different time and length scales were considered for reaction systems with different catalyst aging rates.

1. Overview

A good catalyst not only features high activity and selectivity, but also retains these features for a long time. In the same way, a good reactor design should allow to run the catalytic process at long-term high conversion, selectivity and space time yield. This work aims at reactor designs with overall optimality over the whole catalyst lifetime. According to the lifetime of catalysts, different configurations of heterogeneous catalytic reactors are applied in industry. Three configurations (see Table 1) are considered: a fixed bed reactor in the case study of the ethylene oxide synthesis (slow deactivation), and a fixed bed in swing mode as well as a moving bed in the case study of the methanol-to-gasoline process (fast deactivation).

Table 1: Comparison of three reactor configurations applied for catalyst deactivation.

Configuration	State	Solid operation	Regeneration	Catalyst lifetime
Fixed bed	Pseudo steady state	"Batch"	No regeneration	Months to years
Fixed bed (swing mode)	Dynamic	"Batch"	Every cycle	Days to weeks
Moving bed	Steady state	Continuous	Simultaneous	Seconds to weeks

For the identification of optimal reactor concepts a novel reactor design methodology [Freund2011] based on the elementary process functions concept [Freund2008] has recently been proposed. According to this methodology, a fluid element which travels through the reactor and changes its state within is tracked. By continuously providing the fluid element with optimal fluxes (e.g., heat flux, species fluxes) which are adjusted according to the state of the fluid element optimal reaction conditions in the reactor are achieved at any time. The optimal fluxes can be calculated by solving a dynamic optimization problem which is formulated based on the apparatus-independent model equations including balance equations, reaction and transfer kinetics, thermodynamics and design bounds. In this way, novel reactor concepts with optimal integration of different fluxes can be identified.

In the aforementioned reactor configurations for heterogeneous catalytic reaction systems with catalyst deactivation, a catalyst particle changes its state along the time on stream and/or the reactor length. This has to be accounted for in the design of optimal reactors for such applications. In this regard, the reactor design methodology described above has to be extended in order to consider the solid phase as well as the gas phase, both of which are represented as separate matter elements which are tracked along the reaction coordinate and/or time on stream (Figure 1). The fluxes can be provided dynamically according to the state at a certain location and a certain time on stream.

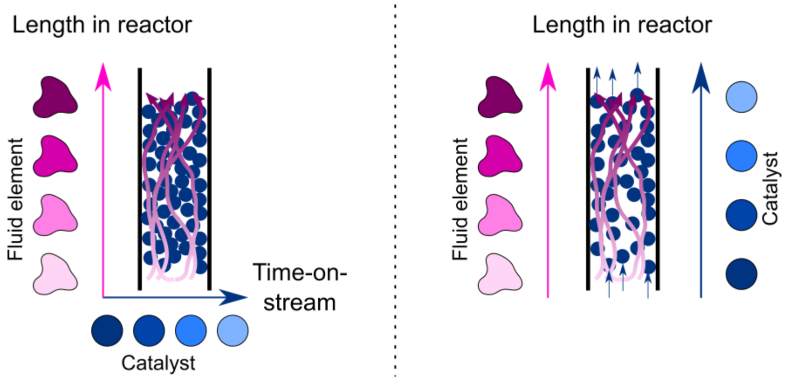


Figure 1: Illustration of the tracking of matter elements in typical reactors for heterogeneous catalytic reaction systems with fixed (left) and moving solid phase (right).

2. Results of the case studies

2.1. Ethylene oxide synthesis

In this reaction system, the lifetime of the catalyst is much longer than the residence time of the fluid element in the reactor. Thus, the lifetime span can be divided into different periods, and in each period the catalyst activity profile is dependent on the time-on-stream of the period. The performance of all periods together is maximized, with uniform reactor parameters and all operating conditions (Figure 2 Left) depending on the period simultaneously being optimized. Additionally, catalysts with different reaction and deactivation rates can be compared with respect to the overall performance by means of a sensitivity analysis (Figure 2 Right).

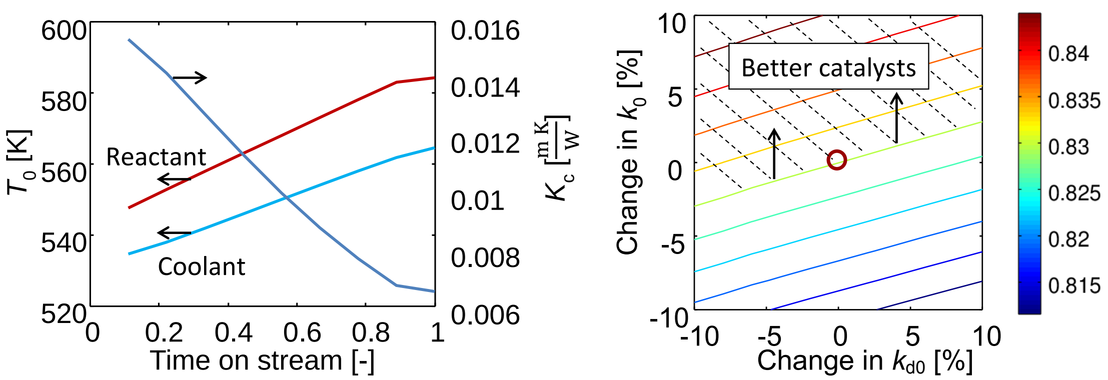


Figure 2 Left: Optimal profiles of inlet reactant temperature, inlet coolant temperature and coolant characteristics. Right: Comparison of resulting average selectivity over the whole catalyst lifetime from catalysts with different initial selectivities and deactivation rates.

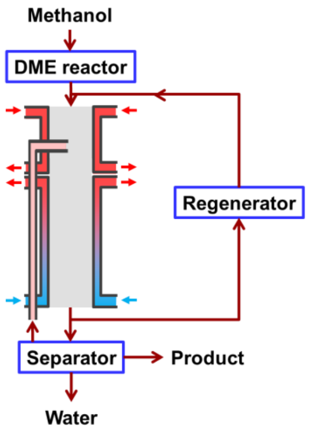
2.2. Methanol-to-gasoline process

2.2.1. Moving bed

In this reactor configuration, the gas phase and the solid phase pass through the reactor with different velocities, achieving steady state. A profile of catalyst activity influenced by the temperature and composition of the gas phase evolves in the reactor. A balance between reaction rates and deactivation rates can be obtained via rigorous optimization, resulting in an optimal reactor concept (Figure 3 Left).

2.2.2. Fixed bed in swing mode

The catalyst lifetime in this reaction system is in the order of days. A distinct aspect of this reactor configuration is the dynamic behavior. The optimal profiles, depending not only on the location but also on the time on stream, can be achieved by controlling operating conditions, e.g. inlet flow rate (Figure 3 Right), optimally along time on stream.



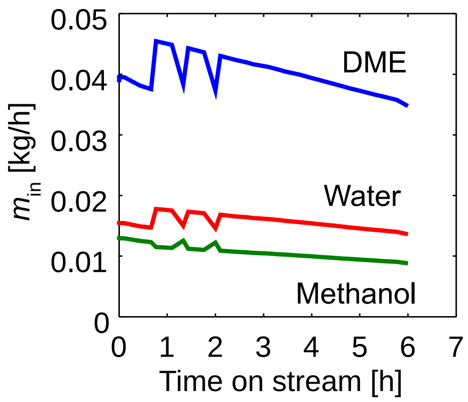


Figure 3 Left: An optimal reactor concept for moving bed configuration. Right: Dynamic inlet flow rate for fixed bed in swing mode.

Conference contributions

M. Xie, H. Freund, *Design and Operation of Heterogeneous Catalytic Reactors to Achieve Overall Optimality over the Whole Catalyst Lifetime*, Oral presentation, Jahrestreffen Reaktionstechnik 2016, 2. – 4. Mai 2016, Würzburg.

M. Xie, H. Freund, *Model-based identification of optimal integrated reactor concepts for heterogeneous catalytic reaction systems with rapidly deactivating catalysts*, Oral presentation, ProcessNet-Jahrestagung, 12. – 15. September 2016, Aachen.

References

H. Freund, A. Peschel, and K. Sundmacher, *Model-based reactor design based on the optimal reaction route*, Chemie-Ingenieur-Technik, 83(4):420–426, 2011.

H. Freund and K. Sundmacher, *Towards a methodology for the systematic analysis and design of efficient chemical processes. Part 1. From unit operations to elementary process functions*, Chemical Engineering and Processing: Process Intensification, 47(12):2051–2060, 2008.