

METHODOLOGY FOR THE DESIGN OF OPTIMAL CHEMICAL REACTORS BASED ON THE CONCEPT OF ELEMENTARY PROCESS FUNCTIONS

Andreas Peschel¹, Hannsjörg Freund^{*,1} and Kai Sundmacher^{1,2}

¹Max Planck Institute for Dynamics of Complex Technical Systems,
Sandtorstraße 1, 39106 Magdeburg

²Process Systems Engineering, Otto-von-Guericke University Magdeburg,
Universitätsplatz 2, 39106 Magdeburg

Summary

A new reactor design methodology based on the optimal trajectory of a reaction system in state space is developed. The general idea is to track a fluid element on its way through the process and adjust the fluxes into the element at every time to optimize a certain objective. Afterwards, the most suitable control variables to obtain these flux profiles are determined.

This model based approach allows a systematic design of new innovative reactor concepts. Additionally, different process integration concepts can be compared and the potential of process intensification measures can be quantified. The method is illustrated for the SO₂-oxidation.

Keyword

Process Intensification, Novel Reactor Technologies, Dynamics and Control of Chemical Reacting Systems

Introduction

Along the path of a fluid element on its way through a chemical reactor the optimal values of the temperature, pressure and educt/product concentration change due the change in the composition of the element. Such optimal profiles can be calculated in state space and represents the optimal route of the reaction system.

However, such optimal profiles can usually not be realized in standard reactors, since the control variables to achieve these profiles like the cooling temperature are constant. Likewise reactor optimization using heuristics, attainable region methods¹ or rigorous optimization methods such as superstructure optimization^{2,3} cannot always guarantee to find the best profiles in state space for the reaction system, since the solution space is limited by using a set of predefined reactors. Even if profiles, e.g. for the cooling temperature, are obtained from these methods, the reactor networks consist usually of several connected ideal standard reactors and can hardly be built in practice. In addition, not all process intensification measures such as distributed dosing of reactants or increasing the heat transfer area can easily be investigated using the aforementioned methods.

Methodological Approach

In this contribution, we propose a new methodology for the analysis and optimal design of chemical reactors based on the best reaction route in the thermodynamic state space. The idea is to determine the best reaction route in state space by tracking a fluid element on its way through the process (Lagrangian approach) and manipulate the

fluxes into this element.⁴ This yields a dynamic optimization problem constrained by the balance equations with given reaction kinetics, initial and final conditions. Instead of choosing a priori a reactor design and optimize the free parameters of the chosen reactor set up, a new innovative reactor design is developed based on the best route in state space. The methodology is intended for the investigation of all kind of process intensification measures such as integration of reaction, cooling and separation in one apparatus or the application of high interface area for heat and mass transfer. To achieve the goal of reactor design based on the optimal route in state space of the reaction system, a multi-level approach is developed.

On the first level, the dynamic optimization problem is constrained by the balance equations, the reaction kinetics, inlet and outlet conditions as well as system inherent boundaries, e.g. a maximum temperature for the catalyst. However, the fluxes into the fluid element are not bounded and they are actually optimized. This gives rise to the maximum possible performance of the reaction system.

$$\begin{array}{ll} \max_{j_A(t)} (\text{Objective}) & \text{Notation:} \\ \text{s.t. } \mathbf{C} \cdot \frac{d\mathbf{x}}{dt} = \mathbf{S} \cdot \begin{bmatrix} \mathbf{j}_A \\ \mathbf{r}_V \end{bmatrix} & \mathbf{x}: \text{ State vector} \\ \mathbf{g}(\mathbf{x}) = \mathbf{0}, \text{ e.g. } \mathbf{r}_V = \mathbf{r}_V(\mathbf{x}) & \mathbf{x}_0, \mathbf{x}_f: \text{ Initial, final conditions} \\ \mathbf{h}(\mathbf{x}) \leq \mathbf{0}, \text{ e.g. } x_i^L \leq x_i \leq x_i^U & \mathbf{C}: \text{ Capacity matrix} \\ \mathbf{x}(t_0) = \mathbf{x}_0, \mathbf{x}(t_f) = \mathbf{x}_f & \mathbf{S}: \text{ Coupling matrix} \\ & \mathbf{j}_A: \text{ Vector of outer fluxes} \\ & \mathbf{r}_V: \text{ Vector of reaction rates} \end{array}$$

* To whom correspondence should be addressed. E-mail: freund@mpi-magdeburg.mpg.de

On the second level, the influence of limited fluxes is investigated and optimal profiles for suitable control variables are obtained. The model is extended by the kinetic expression for all fluxes. Instead of optimizing the fluxes directly, control variables, which can be adjusted over the residence time of the fluid element, are optimized. The control variables are obtained from the kinetic expressions of the fluxes and are either the kinetic coefficients, the driving forces or the exchange areas. This gives rise to various possibilities for controlling the fluxes and several of the below stated optimization problems must be solved. On this level, the best control variables are obtained by applying a systematic decision structure.

$$\begin{aligned} & \max_{\mathbf{K}_A(t), \mathbf{x}_e(t)} (\text{Objective}) & \text{Notation :} \\ \text{s.t. } & \mathbf{C} \cdot \frac{d\mathbf{x}}{dt} = \mathbf{S} \cdot \begin{bmatrix} \mathbf{j}_A \\ \mathbf{r}_V \end{bmatrix} & \mathbf{x}_e : \quad \text{Equilibrium} \\ & \mathbf{j}_A = \mathbf{K}_A(\mathbf{x} - \mathbf{x}_e) & \text{state vector} \\ & k_{ij,A}^L \leq k_{ij,A} \leq k_{ij,A}^U & \mathbf{K}_A : \quad \text{Matrix of} \\ & x_{i,e}^L \leq x_{i,e} \leq x_{i,e}^U & \text{kinetic coefficients} \\ & \mathbf{g}(\mathbf{x}) = \mathbf{0}, \text{ e.g. } \mathbf{r}_V = \mathbf{r}_V(\mathbf{x}) \\ & \mathbf{h}(\mathbf{x}) \leq \mathbf{0}, \text{ e.g. } x_i^L \leq x_i \leq x_i^U \\ & \mathbf{x}(t_0) = \mathbf{x}_0, \mathbf{x}(t_f) = \mathbf{x}_f \end{aligned}$$

Based on the profiles of the best control variables, a technical approximation is developed on the third level of the proposed methodology. The developed reactor represents the technically optimal reactor and approaches the optimal state space trajectory of the reaction system. On the third level the structural shape of the reactor is determined and effects coming with non-uniform field for temperature, concentration and velocity are taken into account.

Besides the dynamic states of the problem a number of inequality and equality constraints exist. In order to solve such dynamic optimization problems properly, the simultaneous approach, which is based on complete discretization of the dynamic optimization problem, is chosen. As discretization method orthogonal collocation on finite elements is used.⁵ The discretized problem is efficiently solved using state-of-the-art NLP solvers such as *IPOPT* and *CONOPT*.

Example: SO₂-Oxidation

In order to illustrate the proposed methodology, the gas phase oxidation of SO₂ to SO₃ is investigated. Besides the economical importance of sulfuric acid the SO₂-oxidation is a well suited example for the development of the methodology. It represents the whole class of exothermic equilibrium reactions, can be modeled with a pseudo-homogeneous model and the required data is available in the literature.

In general, the energy efficiency and the reduction of investment costs are the most important aspects for the economy of the process. The results of the optimization of the minimal residence time problem, which corresponds to

the minimal reactor size or accordingly to the maximum space time yield, are shown in Fig. 1.

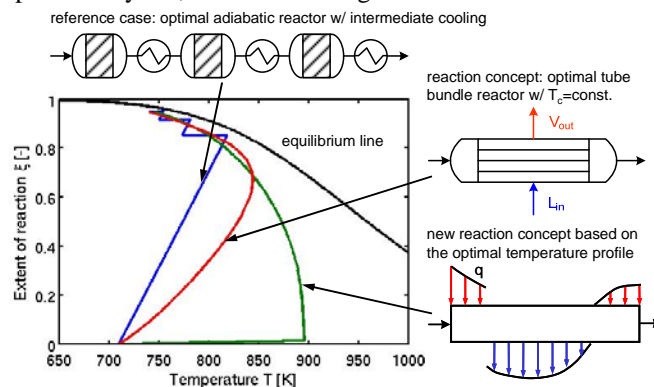


Fig. 1: State space trajectories for different reaction concepts

On the first level, different integrated reactor concepts are compared. The blue line represents an optimal path for an adiabatic reaction concept with intermediate cooling (no integration of reaction and heat removal). The red line constitutes the optimal path for a system with constant, but optimal coolant temperature and heat exchange area. Finally, the green line corresponds to a system where the heat flux is controlled optimally. These calculations yield the potential of an ideal temperature control in comparison to the optimized technical reference case (adiabatic reaction with intermediate cooling).

On the second level, the possibilities for controlling the heat flux are systematically evaluated. The heat flux can be manipulated by adjusting the local values of the heat transfer area (e.g. by changing the geometry), the heat transfer coefficient (e.g. by changing the flow regime) or the temperature difference between cooling media and fluid (i.e. changing the driving force). Based on these results, the best control variable profiles are approximated by a new reactor design giving rise to a technical optimal reactor. Technical approximations of different control variable profiles are possible and will be presented in our contribution.

References

- (1) Glasser, D.; Hildebrandt, D.G. A Geometric Approach to Steady Flow Reactors - The Attainable Region and Optimization in Concentration Space. *Ind. Eng. Chem. Res.* **1987**, 26(9), 1803.
- (2) Lakshmanan, A.; Biegler, L.T. Synthesis of Optimal Chemical Reactor Networks. *Ind. Eng. Chem. Res.* **1996**, 35(4), 1344.
- (3) Kokossis, A.C.; Floudas, C.A. Optimization of Complex Reactor Networks. 2. Nonisothermal Operation. *Chem. Eng. Sci.* **1994**, 49(7), 1037.
- (4) Freund, H.; Sundmacher, K. Towards a Methodology for the Systematic Analysis and Design of Efficient Chemical Processes Part 1. From Unit Operations to Elementary Process Functions. *Chem. Eng. Process.* **2008**, 47(12), 2051.

(5) Logsdon, J.S.; Biegler, L.T. Accurate Solution of Differential Algebraic Optimization Problems. *Ind. Eng. Chem. Res.* **1989**, 28(11), 1628.