EFFICIENT SOLUTION METHODS FOR INTRAPARTICLE DIFFUSION SUITABLE FOR REACTOR OPTIMIZATION

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Abstract

The Multi-Level-Reactor-Design (MLRD) is a novel, model-based optimization method that enables the development of innovative and tailored chemical reactor concepts in a three level approach. Within the scope of this method it is necessary to allow for consideration of mass transport effects inside the catalyst pellets as this directly affects the optimal catalyst and reactor design. However, to include mass transfer inside the pellet into the optimization framework it is mandatory to apply efficient solution methods in order to keep computational times reasonable. In this regard, the Adomian Decomposition Method (ADM) is investigated as an efficient solution method for the pellet balance equations. With the explicit consideration of the pellet mass transfer the optimal design of catalyst pellets can be addressed within the MLRD optimization framework as well.

Keywords

Dusty-Gas-Model (DGM), Adomian Decomposition Method (ADM), Rigorous Optimization, Reactor Design, Catalyst Design.

Introduction

The design of chemical reactors, which are the core of chemical processes, largely affects the performance of the overall process. Therefore, optimization of chemical reactors offers large potential to enhance the energy and raw material efficiency of today's chemical industry. The Multi-Level-Reactor-Design (MLRD) is a novel, model-based optimization method that enables the development of such innovative and tailored chemical reactor concepts. It is based on the concept of elementary process functions and follows a three-level approach (Freund et al., 2010).

Multi-Level-Reactor-Design (MLRD)

The reaction system of interest is modeled rigorously (i.e. based on balances) in an arbitrary shaped fluid element. The fluid element has a volume in which chemical reactions take place (inner fluxes) and an outer surface across which energy and mass may enter or leave (outer fluxes). These fluxes lead to a change of state within the volume. An obvious and important difference between inner and outer fluxes is that outer fluxes may be directly manipulated while inner (reaction) fluxes are always explicitly expressed by reaction kinetics. Within the MLRD method the fluid element is now tracked over time in state space while optimizing its inner state in order to fulfil a certain objective. Obviously, the optimal state in the fluid element changes with time due to chemical reaction. In order to reach the optimal state as close as possible at any time, the outer fluxes are adjusted accordingly. This results in a dynamic optimization problem, which can be solved in a simultaneous approach by using an appropriate discretization scheme and by solving the resulting large scale non linear programming (NLP) problem using appropriate solvers (e.g. IPOPT (Wächter and Biegler, 2006)).

On level one of the MLRD method, outer fluxes may be adjusted freely and the fluid element model is only constrained by reaction kinetics and system inherent bounds (e.g. maximum catalyst temperature). As result, the full potential of the reaction system is revealed independent from known apparatuses. On level two, kinetic approaches (e.g. Fourier's law for heat transport) are introduced also for the outer fluxes. The fluxes can now only be adjusted indirectly via appropriate control variables. The results are profiles over time for these control variables. On level three, the profiles from level two are approximated with a specifically designed reactor setup. As result, the optimal design parameters of the chosen setup are obtained.

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So far, this methodology has been successfully applied to numerous reaction systems, including the oxidation of sulfur dioxide (Freund et al., 2010) and the synthesis of ethylene oxide (Peschel et al., 2012).

MLRD and internal mass transport

The above-mentioned reactions are heterogeneously catalyzed, but were modeled as pseudo-homogeneous systems on all levels. Therefore, the effects of diffusive mass transport in the catalyst pellet were not considered. This neglect of diffusive mass transport is even necessary on level one of the methodology in order to reveal the full potential of the reaction system, provided that intrinsic reaction kinetics are used.

On level two, however, the effects of diffusive mass transport cannot be neglected, in particular if the reaction under investigation is prone to be strongly influenced by internal mass transport. In that case, the reaction flux depends on both, reaction kinetics and mass transport kinetics. Therefore, also mass transport has to be considered in the fluid element model. Rigorous description of internal mass transport greatly increases model complexity and computational effort, since this entails the necessity to additionally solve the pellet balances at any time for the corresponding fluid element state.

As starting point, we decoupled pellet and fluid element model and investigated a possibility to solve the pellet balance equations with regard to reduce computational effort with the Adomian Decomposition Method (ADM). This method provides a series solution for a single differential equation or a set of differential equations (Adomian, 1994). It thereby resigns the computation of discretized methods and leaves the solution of the differential equation as simple evaluation of the series function. The quality of the series solution strongly depends on the number of series terms. The series terms were derived using a computer algebra system (MATLAB® MuPAD).

To evaluate the ADM as option for solving the pellet balance equations one simple textbook example and one industrial relevant example were chosen. The simple system is an arbitrary first order reaction with Fickian mass transport (Eq. 1).

$$N_i = -D_i \frac{dc_i}{dr} \tag{1}$$

For this system, an analytical solution exists which serves as reference for the series solution. As technical system the synthesis of methanol from syngas was chosen. The reaction kinetic model was adapted from Graaf et al., who reported significant mass transport effects (Graaf et al., 1990) for this system. In this model, internal mass transport is described by the Dusty-Gas-Model (DGM).

$$\frac{dx_i}{dr} = -\frac{RT}{p}\left(\sum_{i=1,\,j\neq i}\left(\frac{x_j N_i - x_i N_j}{D_{ij}^{eff}}\right) + \frac{N_i}{D_{K,i}^{eff}}\right) - \frac{x_i}{p}\left(1 + \frac{B_0 p}{D_{K,i}^{eff}\mu}\right)\frac{dp}{dr} \tag{2}$$

For this case numerical results serve as reference.

Results and Conclusion

In accordance with a recent study (Rach et al., 2014), we found that 8-10 series terms are necessary to achieve an appropriate solution quality. For the simple case, the series solution with an infinitely high number of terms converges to the analytical solution (Fig. 1). Furthermore, we found that the quality of the series solution strongly depends on the curvature of the solution. If curvature is small (e.g. as for ϕ =1.5) three series terms are already sufficient to perfectly depict the exact solution. Nonetheless, the feasibility to derive 8-10 series terms depends on model complexity, e.g. with regards to the number and complexity of the model equations.

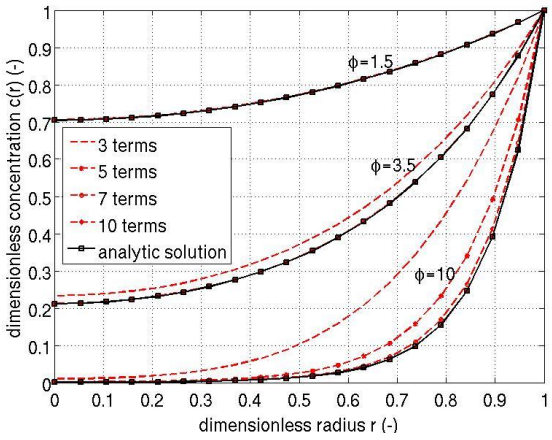

Figure 1: Dimensionless concentration profiles in the catalyst pellet for different Thiele modules. Exact solution: solid lines with squares. Series solution with different number of terms: dashed lines.

In our ongoing work we further investigate the applicability of the ADM for reactor optimization regarding computing time requirements. First results show that by using the ADM the computing time can be decreased by a factor of up to seven. Another aspect under investigation is the optimization of the catalyst pellet itself. An optimization model with coupled pellet and fluid element model will be developed which allows for the derivation of reactor and/or catalyst design parameters that account for mass transport effects in the catalyst pellet.

References

Adomian, G. (1994). *Kluwer Academic Publishers*. ISBN 0-7923-2644-X.

Freund, H., Peschel, A., Sundmacher, K. (2010). *Chem.-Ing.-Tech.*, *83*, 420.

Graaf, G. H., Scholtens, H., Stamhuis, E. J., Beenackers, A. A. C. M. (1990). *Chem. Eng. Sci.*, *45*, 773.

Peschel, A. Jörke, A., Sundmacher, K., Freund, H. (2012). *Chem. Eng. J.*, *207-208*, 656.

Rach, R., Duan, J.-S., Wazwaz, A.-M. (2013). *J. Math. Chem.*, *52*, 255.

Wächter, A., Biegler, L. T. (2006). *Math. Program.*, *106*, 25.