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Multiscale modelling strategy for structured catalytic reactors

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ABSTRACT

Structured reactors, and especially arranged reactors, are multiscale systems, in which the design on the small scale essentially determines the macroscopic reactor behaviour. In the present contribution, we propose a modelling strategy for the design and optimisation of an arranged flat-bed reforming reactor. It consists of three model levels, each describing the structure of the reactor on a different scale and detail level. The elements of this hierarchy are consistent and compatible, which means that results obtained with one model can be transferred to the other models. The possible applications of this model hierarchy range from estimation of pressure drop coefficients up to the integration of the structured reactor into a process plant.

1. Introduction

Structured reactors play a key role in the design of intensified multiphase processes in chemical engineering (Pangarkar et al., 2008). In contrast to classical fixed beds, which are random packings of catalytically active particles, structured reactors can be purposefully designed on all scales from the catalyst level up to the reactor level (Kreutzer et al., 2006). For, say, spherical non-porous catalyst particles, the structure of a fixed bed has only one design parameter, which is the size of the particles. It determines the size of the available catalyst surface and thereby the reaction rate and catalyst efficiency; small particles are preferred here. The particle size also determines the interparticle length, which has a strong impact on the total pressure drop of the reactor; large particles are preferred in order to keep the pressure loss low. Selecting the particle size therefore is a compromise between maximising the catalyst efficiency and minimising the pressure loss. Application of porous or non-spherical particles adds some degrees of freedom to the design, but it does not allow to design mass transport and reaction independently.

In a structured packing, the design of the structure introduces many new degrees of freedom for reactor design, allowing to decouple the hydrodynamic state (pressure drop), the reaction rate (catalyst efficiency) and the transport of heat and mass (reaction control) to a wide extent (Pangarkar et al., 2008). Thus, in a structured reactor these processes can be adjusted in order to control the temperature and concentration profiles along the flow path, thereby improving selectivity and conversion, and avoiding local temperature peaks.

According to Cybulski and Moulijn, (2006), structured reactors can be categorised into monolithic reactors, membrane reactors and arranged reactors (see Fig. 1). Numerous contributions about the modelling and simulation of monolithic reactors have been published, including modelling of specific details and simulations of complete reactors (e.g. Chen et al., 2008; Tischer and Deutschmann, 2002). Also, a wide range of contributions on modelling of membrane reactors is available (e.g. Marcano and Tsotsis, 2002). With regard to arranged reactors, many contributions treat the modelling of gas–liquid systems in structured packings (see Pangarkar et al., 2008), which are widely applied, for example in reactive distillation. However, only a few publications treat arranged reactors for heterogeneously catalysed gas-phase reactions. von Scala et al. (1999) investigated flow paths in Katapak-M packings and estimated pressure drops from CFD simulations of the structure for single phase flow. Their model-based results correspond very well with experimental data. Petre et al. (2003) proposed a computational method for a priori estimation of the pressure drop in a structured packing by analysis of dissipation rates in recurrent structure elements. Calis et al. (2001) focussed on regular fixed bed channels with very low tube-to-particle-diameter ratio. They applied CFD simulations to predict pressure drop correlations, which showed reasonable agreement with experimental results. However, none of these contributions treats mass and heat transfer in arranged, parallel passage reactors, which is the focus of this work.

Like all structured reactors, arranged reactors are multiscale systems; the geometrical design of the structure determines the

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processes of transport and reaction on this scale and has a strong impact on the behaviour of the complete reactor on a larger scale. Thus, the ability to describe the processes on a small scale and transfer this knowledge to the reactor scale is a prerequisite for a purposeful reactor design. While the simulation of the processes on the scale of the repeating structures can be done with established methods such as computational fluid dynamics (CFD), the same level of detail cannot be applied to the description of the whole reactor due to high computational costs. Instead, reduced reactor models are required, which reflect the essential behaviour on the small scale, but which are computationally cheap and thus applicable to the simulation of the complete reactor.

In this contribution, we propose a multiscale hierarchy of three model levels for the design and analysis of structured reactors (Fig. 2). Each model describes the reactor or a part of it on a different level of detail and is tailored for specific tasks:

- The *detailed model* describes one or several repeating sections of the structure in all geometric details. It is not only applicable for the analysis and design of the structure, but it also allows to estimate heat and mass transport parameters needed to transfer the essential characteristics of the structure to the reduced models (see below).
- The *zone model* neglects exact geometrical details. Instead, it considers discrete volumes in which the reaction takes place. It is useful, e.g. for designing the reactor-wide flow pattern.
- The *phase model* has spatially homogenised reactive and non-reactive phases, coupled by interphase mass and heat transfer. It is suitable to describe the spatially distributed reactor behaviour and its interaction with surrounding devices in a computationally efficient way. It can be applied for optimisation of the reactor and its integration into complex devices.

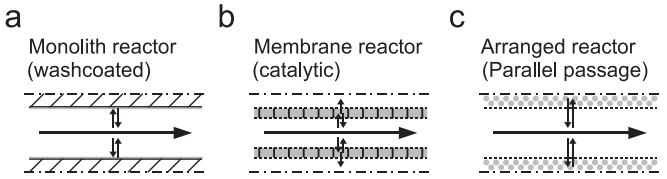

Fig. 1. Classes of structured reactors according to Cybulski and Moulijn (2006).

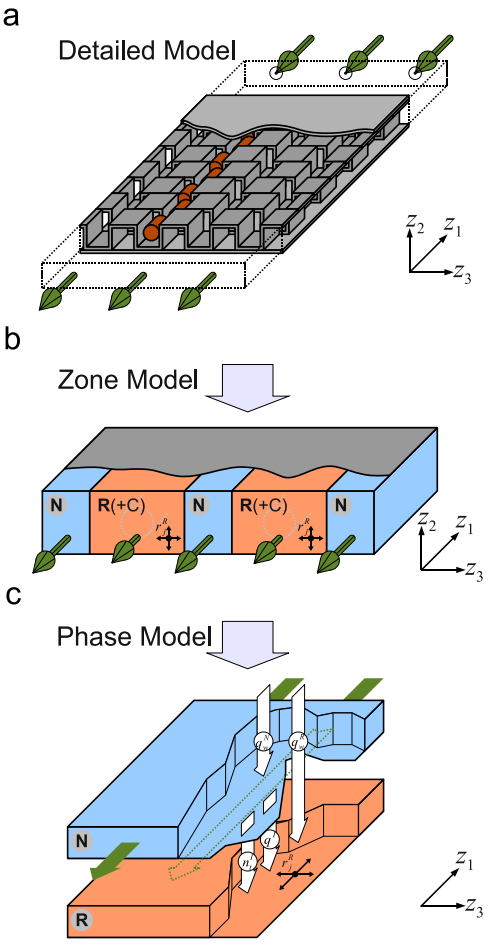

Fig. 2. Proposed model hierarchy for structured reactors. N, R: non-reactive and reactive zone or phase; C: catalyst phase, n_i, q: molar fluxes and heat flux, respectively. Drawings are not to scale.

None of these models is entirely new. The novelty of this contribution is the hierarchy of coherent and compatible models; results obtained with one of the models can be transferred to other elements of the model hierarchy. Furthermore, the application of the models covers a wide spectrum of tasks ranging from the design of the structure up to the heat integration and optimal control of the complete reactor. It is the latter aspect, in particular, which makes the proposed hierarchy a powerful and useful tool.

In this contribution, we derive and discuss the three models illustrated in Fig. 2, thereby focussing on a flat bed reforming reactor that is used as an indirect internal reforming unit in high temperature molten carbonate fuel cell (MCFC) systems (Bischoff and Huppmann, 2002; Pfafferodt et al., 2010). Its purpose is to reform the fuel gas (usually a methane/steam mixture) to a certain conversion before the gas mixture is fed to the fuel cells. They are inserted into the fuel cell stack at certain intervals, so they absorb heat from the fuel cells they are attached to. The elementary cell of this arranged reactor is shown in Fig. 3 left. It consists of two corrugated metal sheets, which are aligned in a staggered way, forming small chambers. In some of these, cylindrical catalyst particles are inserted.

Because the derivations of the zone and the phase models are motivated by the simulation results obtained from their preceding models, we discuss the models directly together with their results and some examples of typical applications. The detailed model and the zone model have already been published elsewhere in detail (Pfafferodt et al., 2008). For the sake of completeness, we repeat the most important aspects of these models here, so the reader can follow the argumentation, and refer to the mentioned publications for further details. Because the phase model has not been published elsewhere, we discuss its derivation in detail.

In Section 5, we also discuss how this hierarchical modelling approach can be transferred to other arranged, parallel passage reactors such as Katapak structures.

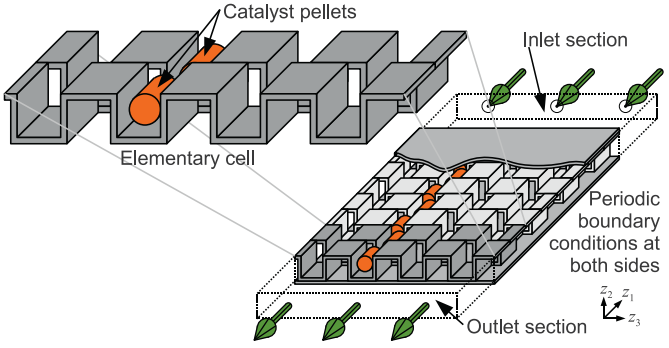

Fig. 3. Elementary cell and scheme of the detailed model of the reforming reactor (U.S.-Patent 6200696, not to scale).