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Bridging the gaps: from particle-resolved to multi-tubular reactor simulation

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Packed bed reactors have been widely used in the chemical and process industry for several decades. They can be difficult to design and operate due to their size and complexity. Therefore, there is still room for improvement of the performance of these reactors and rigorous simulations can help here to achieve desired goals with lower upfront investment. Recent advances in modeling particle-resolved packed beds allows a detailed inside in the flow, species and temperature distribution in the beds and therefore also into the conversion of the surface reactions. Based on these simulations, model parameters for 1D-models can be estimated. These 1-D models can then be used to calculate performance of multi-tubular reactors, either by running a computationally expensive simulation with resolved tubes or by coupling the CFD simulation to an advanced process modeling tool like gPROMS. In the latter case, the flow non-uniformity as well as certain flow properties like coolant velocity and temperature is taken from the CFD simulations, while on the process modeling side heat transfer and reactions in the packed bed are calculated based on the simplified 1D models. This modeling approach is fully 2-way coupled and highly efficient in terms of accuracy and especially runtime and it can be embedded into a flow sheet simulation.

In this contribution, we will present the whole simulation process and how the different steps intertwine with each other starting from the detailed particle resolved simulation all the way down to the flow sheet simulation. The benefit of this approach will be demonstrated based on several examples.

1. Introduction

Packed bed reactors are one of the working horses in the chemical and process industry. They are used amongst others for highly exothermic or endothermic catalytic reactions like (partial-)oxidation, dry reforming or stream reforming of Hydrocarbons. It is typically carried out in multi-tubular reactors to ensure safe thermal behavior. Multi-tubular reactors typically consist of several thousand tubes, each of a length of several meters and a diameter of a few centimeters, see Figure 1. Each tube is filled with catalytic particles and it has to be ensured to avoid maldistribution, that each tube exhibits the same pressure drop. All these tubes are bundled in a vessel-like shell with a liquid flow around them to transfer heat. The catalytic particles themselves can be porous, so that not only the outer surface of the particles is used for the reaction. Reactants can then also diffuse into the particle and react on the inner surface. Mostly cylindrically shaped particles were used in the past, attributed to manufacturing limitation and efficiency.

Designing and troubleshooting such reactors is challenging. On the one hand measurements are difficult due to accessibility and due to the operating conditions of such reactors. Lab-scale experiments consider ideal behavior which cannot be guaranteed in the reactor and often is not the case. Therefore, numerical experiments in a Digital Twin of the reactor or process offers new ways to better design multi-tubular reactors. Simulation methods are designed for a specific aspect or scale and no single method is capable to handle the whole multi—tubular reactor. In this contribution we present a workflow to bridge the various time and length scales occurring

in a multi-tubular reactor to allow simulations of the whole multi-tubular reactors in a reasonable time enabling new ways to design them in a better way.

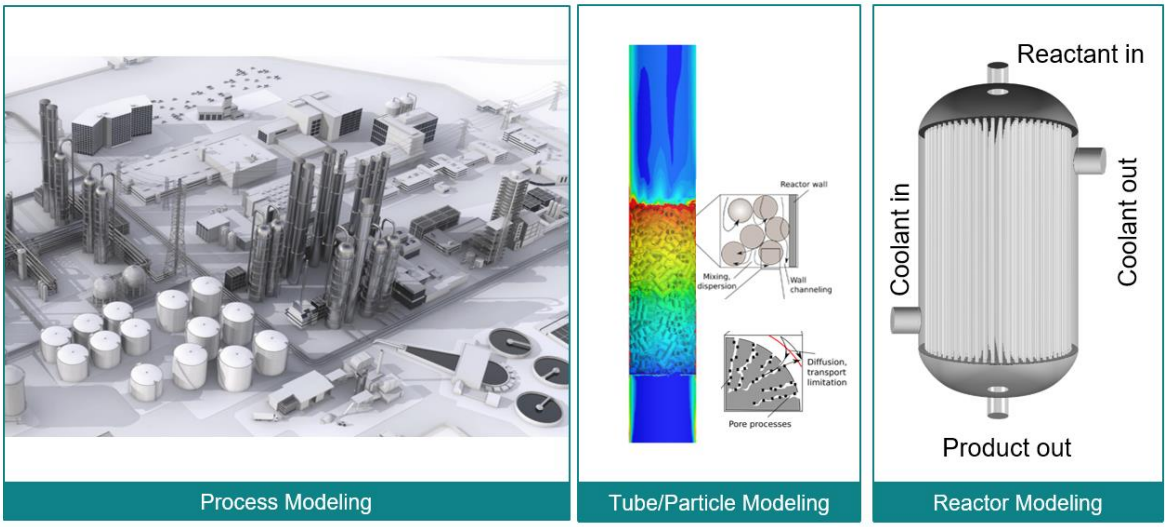

Figure 1: Schematic overview of multi-tubular reactor embedded in a larger process.

2. Simulation on Particle Level

Particle-resolved 3D CFD (computational fluid dynamics) simulation have been conducted at least for the last 20 years starting with a publication on heat transfer by Nijemeisland and Dixon (2000). But a break-through of this method was reached around 2010 where the numerical methods as well as the computational resources have developed in such a way that realistic simulations of a few hundred up to a few thousand particles could be conducted.
The goal of these simulations is to understand in detail the (local) flow and the pressure drop but also to evaluate the chemical reactions, the species and temperature distribution to e.g. avoid hot spots or to improve conversion and selectivity.

Eppinger et al. (2011) proposed a fully virtual and automated workflow for spherical particles by combining the Discrete Element Method (DEM) with CFD. DEM is used to create a realistic bed structure by pouring particles into the tube. The particles settle due to gravity and the final bed structure is created when all particles are in rest. The next step is to turn the final bed structure into a CAD description so that a CFD mesh can be created. One issue for the mesh generation is the particle point contact: This leads typically either to highly skewed cells which makes convergence of the CFD simulation difficult or requires refinement of the cells which increases the computation time severely. Eppinger proposed a new methodology where the particle surface is locally modified is such a way that a very small gaps is created which can be meshed with only a few cells with reasonable quality. This approach shows advantages regarding accuracy and automation compared to other methods. And it is extensively validated in terms of local and global void fraction, pressure drop, flow as well as temperature and reactant and product profiles (Wehinger et al. (2016)). Recently a generalized workflow which is also suitable for non-spherical particle shapes was published and validated by Eppinger and Wehinger (2020). This is even more important if recent trends are considered: optimizing particle shape for a certain application or process to reduce pressure drop while maintaining or improving product yield. It is attracting more and more interest due to new manufacturing possibilities with additive manufacturing to create more complicated particle shapes making it economically feasible.

2.1 Benefits of particle resolved simulation

Apart from detailed information in time and space from within the packed bed, this approach can also be used to investigate and optimize different geometric parameters like the particle shape or the tube configuration or optimize operating conditions. Wehinger et al. (2015) investigated different particles shapes and how they affect

conversion and yield of dry reforming of methane. A detailed mechanism for the dry reforming proposed by McGuire et al. (2011) was used. All relevant parameters like available active catalytic surface area, residence time and inlet composition were kept constant for all shapes. The investigated Raschig-rings unexpectedly showed a significantly reduced conversion and yield in the order of approx. 30%.
Jurtz et al. (2020) investigated the effect of internal heat fins on the pressure drop and heat transfer. This is a relatively new technique and fundamental research is required. In their investigation they found an excellent agreement between experimental and simulation results which is the basis to do numerical optimization studies. In order to do this efficiently the whole process from creating and modifying the heat fins, filling the tube, meshing and solving has to be automated. Within the used Software package Simcenter STAR-CCM+ by Siemens this is easily doable by using the built-in optimization capabilities and workflow automation.
One drawback of the 3D particle-resolved simulations is the computational cost which limits the number of particles to a few thousands which can be simulated today in a reasonable time. And with a few thousand particles only a small portion of a single tube can be represented. Nevertheless, even with such a relative short bed length a lot of knowledge and benefit can be extracted out of such simulation. In a recent publication Jurtz et al. (2019) showed how such detailed simulation can be used to extract model parameters to be used in simplified models.
A commonly used model to simulated flow through packed beds uses an axial dispersion coefficient D_{ax}.

$$\frac{\partial c}{\partial t} = -u\frac{\partial c}{\partial z} + D_{\mathrm{ax}}\frac{\partial^2 c}{\partial z^2} + \dot{R} \tag{1}$$

The axial dispersion coefficient includes all deviations from ideal flow. Typically for simple shapes and large tube-to-particle ratios N correlations are available. But for small values of N, which are typical for multi-tubular packed bed reactors, or for packings with newly developed or complex shapes, such correlations are not available and axial dispersion coefficients have to be determined experimentally. This is often costly and time-consuming, so that numerical experiments are an efficient alternative. Based on a packing with a suitable length, the tube is split into a number of compartments along the axis and the residence time distributions are determined with the help of a Lagrangian Particle (LMP) tracking simulation. These distributions are then averaged and fitted to a curve by Levenspiel (1999), so that D_{ax} could be determined. Finally, the resulting dispersion coefficient were validated against experimental data and showed an excellent agreement (Jurtz (2021)).

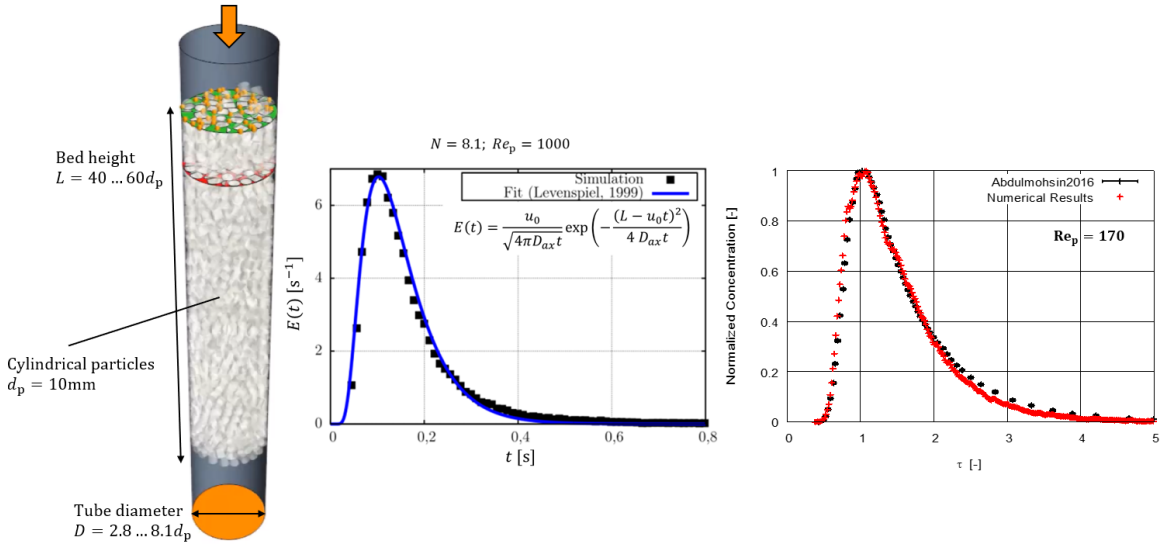

Figure 2: Determining axial dispersion coefficient from detailed CFD-DEM simulations to be used in simplified models.

3. Simulation on Reactor Level

To obtain accurate results and to make use of the predictive capabilities of simulations on reactor level, the local variations need to be considered. Specifically, the local velocity of the coolant, the temperature of the coolant and as a result the local heat transfer coefficient on the tube walls which varies along the tube and but depends also on the relative position of the tubes in the reactor. And this obviously also affects all processes in the tube, namely the heat transfer and the catalytic surface reactions. Simulating several thousand tubes and the coolant flow around them is still computationally too expensive even with simplified models (Eq(1)) as discussed in the previous section.

Therefore, a coupled simulation between a 3D CFD simulation and an advanced process modelling tool is proposed. The overall idea is to split the large variation in space and time and solve different aspect with different tools effectively. A schematic overview is depicted in Figure 3. The 3D CFD simulation analyzes the shell side by simulating the coolant flow and coolant temperature distribution. In this analysis, the tubes are not resolved but represented by a porous media approach, which acts as a resistance affecting flow pattern. On the other hand, the catalytic processes in the tubes is analysed efficiently by gPROMS by PSE, an advanced process modelling environment by solving a 1D model along the tube axis. There are no limitations regarding the complexity of the processes which can be simulated, including heat transfer, gas phase reactions, surface reactions, as well as intraparticle processes & radiation effects.

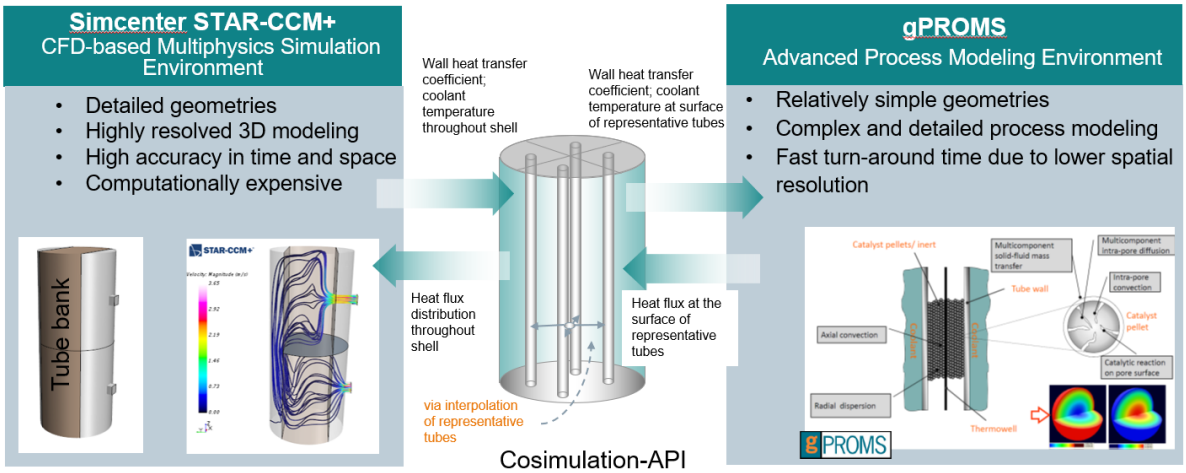

Figure 3: Schematic overview of the coupling workflow for simulation on reactor level.

Solving these 1D equations for all tubes is not very efficient. Therefore, an intermediate layer between the CFD-side and the process modelling side is introduced. This intermediate layer connects both simulation via a limited number of representative tubes, typically some tens up to 100, where the relevant variables are interpolated. This results in the following simulation workflow for a catalytic packed bed:

1. Solve cold coolant flow on the CFD side
2. Extract local coolant velocities and coolant temperature and pass this to the intermediate layer via the Co-simulation-API
3. The intermediate layer determines based on the local coolant velocity and temperature the local heat transfer coefficient for each representative tube
4. Process model calculates the reactions along each representative tube and is using the local heat transfer as boundary condition. Finally, the heat of reaction is passed back to the intermedia layer.
5. Based on the representative tubes, the intermediate layer calculates a volumetric heat source terms to be used by the CFD side.
6. The volumetric heat source term is applied and the whole procedure is repeated until the whole simulation is converged.

With this approach a multi-tubular packed bed reactor can be simulated in a few hours enabling rigorous design optimization to improve reactor performance. Typical design decisions comprise the reactor diameter and

height, tube diameter and height, baffled design, if any, tube bundle arrangement, coolant mass flow rate and temperature, process flow rate, composition and temperature in the tubes as well as catalyst properties. Additionally, also certain constraints have to be considered, like reactor dimension constraints, safety and environment protection aspects, quality and capacity of the product etc.

Figure 4 shows the base line design with large differences between the temperature profiles for tubes at different positions in the reactor and it is clearly not an optimal design regarding product yield and does not minimize the risk of hot-spot formation and subsequent catalyst burn-out in certain areas of the tube bundle.

Using the proposed methodology, it is possible to study the effect of aspects of geometry (e.g. baffle spacing, baffle pitch, tube pitch, and the detailed geometry of the 3D tube bank) that affect the shell-side coolant flow and heat transfer coefficients. This is done iteratively adjusting the dimensions within the CFD model and simulating the reactor with the altered geometry until more uniform performance is achieved. It is also possible to investigate the effect of tube-side "configuration" aspects such as the catalyst layer height and catalyst loading. The results of optimizing the reactor shell geometry in shown in Figure 4. By adjusting aspects of the internal geometry, it has been possible to achieve virtually uniform radial temperature profiles. As a result, the reactants in all tubes are subject to the same or very similar external conditions at any cross section of the reactor, with little discrepancy of performance arising from the radial position of the tube within the tube bundle. This means that the reactions occurring within the tubes, and hence the conversion, are very similar for all the tubes across the bundle.

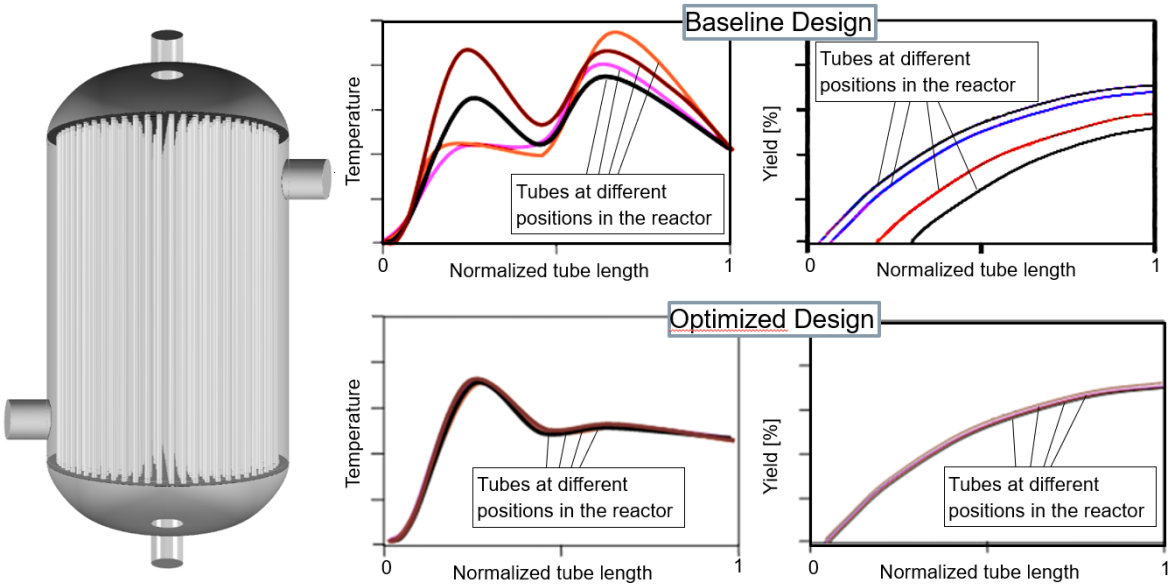

Figure 4: Baseline design and optimized design of a multi-tubular reactor

And finally, the results of the simulation can be integrated into a flow-sheet model of the whole process or plant. This can be done e.g. via Model Order Reduction. This is needed, because by default, a digital twin model as this one is usually still too slow for flow-sheeting simulations. Making use of ROM's, which run in real time and fixed time step, it is possible to simulate closed loop systems in real time for better control and operation of the existing plant

4. Summary

In this contribution, a workflow for a fully Virtual Twin of multi-tubular packed bed reactors was presented.

- On the particle level, a combination of DEM and CFD with some advanced meshing technology was used to get detailed information about all relevant fields (velocity, pressure, temperature, species …) and parameter (bed porosity, pressure drop, yield, selectivity…). This can include detailed catalytic surface chemistry. With that, rigorous optimization on particle level is possible, e.g. for particle shapes or operating conditions.
- On the tube level, a combination of DEM and CFD can be used for fundamental numerical studies to derive model parameter for simplified models and can replace costly and time-consuming real-world experiments.
- On the reactor level, a combined approach of CFD simulation and process modeling can simulate multi-tubular reactors in a reasonable amount of time. With that, rigorous design optimization on the reactor level is possible.

References

Abdulmohsin R. S., Al-Dahhan M. H. (2016), Axial Dispersion and Mixing Phenomena of the Gas Phase in a Packed Pebble-Bed Reactor, Annals of Nuclear Energy, 88, 100-111

Eppinger T., Seidler K., Kraume M. (2011), DEM-CFD simulations of fixed bed reactors with small tube to particle diameter ratios, Chemical Engineering Journal, 166, 324-331

Jurtz N., Waldherr P., Lucht D., Eppinger T., Kraume M. (2019), Determination of Effective Transport Parameters from Particle-Resolved CFD Simulations for a Simplified Fixed-Bed Reactor Modeling, AIChE Spring Meeting 2019, 65a

Jurtz N., Flaischlen S., Scherf S.C., Kraume M., Wehinger G.D. (2020), Enhancing the Thermal Performance of Slender Packed Beds through Internal Heat Fins, Processes, 8, https://doi.org/10.3390/pr8121528

Jurtz N., Schönherr T. D., Kraume, M., Numerical investigation of mechanical axial dispersion in slender fixed-beds. AIChE Journal. Wiley. 2021. [submitted]

McGuire N. E., Sullivan N.P., Deutschmann O., Zhu H., Kee R.J. (2011), Dry reforming of methane in a stagnation-flow reactor using Rh supported on strontium-substituted hexaaluminate, Applied Catalysis A: General, 394, 257 – 265

Nijemeisland M., Dixon, A.G. (2001), Comparison of CFD simulations to experiment for convective heat transfer in a gas-solid fixed bed, Chemical Engineering Journal, 82, 231-246

Wehinger G.D., Eppinger T., Kraume M. (2015), Evaluating Catalytic Fixed-Bed Reactors for Dry Reforming of Methane with Detailed CFD, Chemie Ingenieur Technik, 87, 734-745

Wehinger G.D., Kraume M., Berg V., Korup O., Mette K., Schlögl R., Behrens M., Horn R. (2016), Investigating dry reforming of methane with spatial reactor profiles and particle-resolved CFD simulations, AIChE Journal, 62, 4436-4452