Towards more economical flow simulations in structured catalysts

Volume-averaged modeling and simulation of structured catalysts: assumptions on the effective diffusivity in steam-methane reforming

***J. A. Medina Méndez, H. Schmidt**, Lehrstuhl Numerische Strömungs- und Gasdynamik, Brandenburgische Technische Universität Cottbus-Senftenberg*

In Short

- The project focuses on the fluid-dynamic study of a structured catalytic reactor for steam-methane reforming (SMR).
- Several 3-D numerical simulations of a honeycomb-like monolith will be performed. The numerical simulations assume different simplified forms for the effective diffusivity tensor, e.g., concerning the nature of the (mass) diffusion of chemical species and permeability. Sensitivity to the flow inlet temperature and Reynolds number is also evaluated.
- Detailed knowledge on the tensor can help formulate homogeneous models for the flow within the catalytic reactor; these are significantly more economical than pore-scale Direct Numerical Simulations (DNS) of the Navier-Stokes equations.

With almost 47% share as of 2021, SMR is the dominant process used in global H_2 production. SMR can achieve overall efficiency values in the range of $80 - 85\%$. These efficiency values consider the use of highly optimized catalyst materials and/or catalytic surfaces, as well as typical operating conditions, i.e., generally assumed adiabatic reactors and low flow rates. Fluid flows within structured catalytic reactors can be analyzed in the context of porous media flows and volume-averaging theory (VAT), see [1]. For reaction-diffusion dominated systems, which largely correspond to the laminar flows representing the focus of this study, VAT defines some closure strategies for the so-called effective diffusivity tensors. These are required for proper modeling of unresolved micro-scale dynamics in macro-scale simulations, see [1]. Pursuing one universal formulation or closed form for such tensors is akin to the challenge of finding closed forms of the subgrid-scale (SGS) Reynolds stress tensor in large eddy simulations (LES). This is because VAT essentially considers the application of spatial filters of specific type on the flow field variables. In the field of catalysis, large efforts are dedicated to the understanding of chemical reaction pathways and mechanisms. Several models for reaction kinetics in SMR (reaction mechanisms) can be found in the literature, e.g., see [2]. In contrast, detailed CFD or experimental published work related to fluid-dynamic issues is more limited, see a relevant example in [3]. Nevertheless, multiscale modeling and simulation are very relevant for the optimization of catalyst design and manufacturing. In principle, it may be possible to avoid the need to have extremely efficient kinetics or catalyst materials, if other factors such as the macroscopic flow can be optimized, e.g., optimal heat transfer distribution within the catalyst reactor, local optimal mass transfer due to flow stabilization or destabilization, among other strategies. Modest efforts in specialized CFD research on monolith catalyst reactors may have been stunted due to the necessity of large computational infrastructures required to perform 3-D numerical simulations, or in some cases even pore-scale direct numerical simulations (DNS). More often than not, 3-D simulations are the only ones which can offer accurate and relevant insights on the nature of the closure tensors required for the volume-averaged dynamics. Therefore, it is of interest to explore different modeling strategies for effective diffusivity tensors, which can be formulated from 3-D simulation data. Such strategies should allow a more efficient use of computational resources when simulating structured catalysts. The PI shows in [4] an introduction and some preliminary ideas regarding the formulation of quasi-1-D and 3-D homogeneous models for CO_2 methanation (CO_2-MT) structured catalysts. In a more recent work, more details are discussed for the formulation of both 1-D and 3-D homogeneous models, comparing SMR and CO_2-MT processes, see [5]. In the 3-D model, a comparison is shown between different assumptions on the nature of the representative elementary volume (REV) for a volume-averaged reactor analysis. The assumptions consider an impermeable reactor material ('impermeable' model), a porous reactor material ('porous' model), and a homogenized reactor ('homogeneous' model), i.e., a canonical reacting pipe-flow.

The 'porous' model formulated in [5] and shown in Figure 1 is the one for consideration in this project. The inlet flow is fully laminar, and is characterized by a Reynolds number $\mathrm{Re} \approx 0.35$ based on the reactor diameter and the inlet flow velocity (and kinematic viscosity). The near-wall resolution at the reactor inlet and outlet is estimated as $\Delta r_0^+ \approx 0.01$, considering a viscous length-scale calculated with a steady

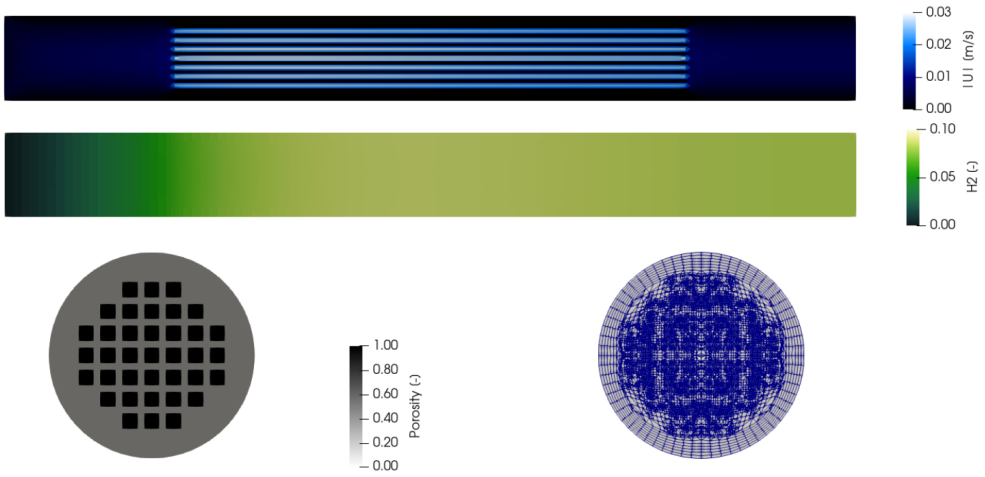

Figure 1: *Flow snapshot of the monolith reactor ('porous' model in [5]): Longitudinal section showing the magnitude of the velocity field within the reactor (top), longitudinal section showing the H_2 mass fraction field (middle), cross-section at reactor half-length showing the porosity field (bottom left), cross-sectional view of the numerical mesh at reactor half-length (bottom right).*

laminar pipe-flow solution. The flow is assumed isothermal. The nature of the effective diffusivity tensors used in [5] is of diagonal character, i.e., assuming that the effective diffusivity tensor consists only of principal components. Another assumption is that the (mass) diffusivity of chemical species is determined considering an assumption of unity Lewis numbers. As part of this project, some of these assumptions will be relaxed, and the corresponding changes in global quantities of interest (conversion efficiency, pressure drop, and similar) will be assessed. Specifically, project objectives aim to quantify the change in global quantities of interest due to changes in the assumed nature of the chemical species effective diffusivity (tensor), changes in the assumed form of the permeability (tensor), changes in the inlet flow Reynolds number, and changes in the inlet flow temperature. To that extent, several numerical simulations will be carried out with the open source CFD library OpenFOAM. The use of HPC architecture will allow a feasible project time frame, otherwise unrealistic. Extended implications of this study in the context of engineering applications imply, naturally, the optimization of catalytic reactors with the purpose of their eventual large-scale deployment and operation.

WWW

https://www.b-tu.de/en/fg-stroemungsmodellierung/divisions/flow-modelling-for-energy-and-process-engineering

More Information

[1] S. Whitaker, *The Method of Volume Averaging*, vol. 13 of *Theory and Applications of Transport in Porous Media*, Springer Science & Business Media, B.V. (1999).

[2] B. A. Haberman, J. B. Young, *Int. J. Heat Mass Transfer* **47**(17), 3617-3629 (2004). doi: 10.1016/j.ijheatmasstransfer.2004.04.010

[3] V. Palma, M. Miccio, A. Ricca, E. Meloni, P. Ciambelli *Fuel* **138**, 80-90 (2014). doi: 10.1016/j.fuel.2014.06.043

[4] J. A. Medina Méndez, M. González-Castaño, F. M. Baena-Moreno, H. Arellano-García, *J. Phys.: Conf. Ser.* **2367**(1), 012015 (2022). doi: 10.1088/1742-6596/2367/1/012015

[5] J. A. Medina Méndez, B. Dorneanu, H. Schmidt, H. Arellano-García, *J. Phys.: Conf. Ser.*, Accepted for publication (2024).

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