Modelling heterogeneous catalysis in open-cell porous foam structures with particle based simulation methods

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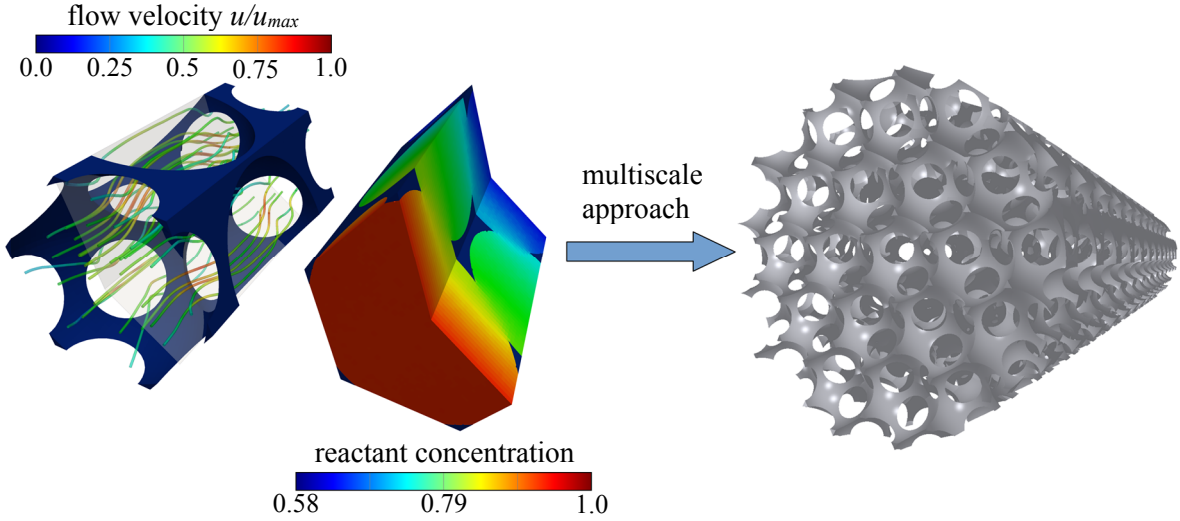
ABSTRACT

Heterogeneous catalysis in metallic or ceramic foam structures represents a very promising alternative to catalysis in packed beds or monoliths. Due to high porosity, specific surface and tortuosity these structures provide excellent mass transport properties at moderate pressure drops [1].

Simulating catalysis in foam structures requires to merge reaction kinetics into gas dynamics within complex geometries. Particle based simulation methods are eminently suitable for this, allowing us to decouple the simulation grid and the boundary representation using constructive solid geometry. In order to completely eliminate the influence of the simulation grid, we propose an essentially grid-free variant of stochastic rotation dynamics, a popular numerical method for the modelling of fluids on mesoscopic scale [2, 3]. Instead of Cartesian grid cells, we use spherical coarse-grained interaction volumes, which we randomly distribute over the domain [4]. Employing this method together with constructive solid geometry, we investigate heterogeneous catalysis in open-cell foam structures, modelled by inverse sphere packings [5].



As prototype reaction we have chosen the low temperature water gas shift following the Langmuir-Hinshelwood reaction mechanism [6]. The foam structure serves as substrate and is assumed to be coated with $CuO/ZnO/Al_2O_3$ washcoat. The effective reaction rate in the washcoat layer is computed using precomputed look-up tables for the effectiveness factor [7]. Among other parameters, the effective reaction rate depends on the partial surface pressures of the reactants, which can be computed from the collision fluxes on the surface. Hence, the relevant quantities are evaluated exactly at the reactive boundary. Concerning particle based methods, pressure boundary conditions often suffer from instabilities, if not implemented carefully [8]. Therefore, we connect inlet and outlet via an extended periodic boundary condition allowing for discontinuities in the concentration field, while the density, temperature and the velocity fields are strictly periodic. In order to drive the flow, an external acceleration is applied.

First, we investigate the flow through single basis foam cells for a wide range of parameters. Second, we will employ a multiscale approach based on the gathered data and investigate complicated porous structures on macroscopic scale such as an entire catalytic converter. Further, we will apply this approach to find optimum foam structures on macroscopic scale, which for example minimize flow resistance while maximizing the reaction rate.

REFERENCES

[1] F. Lucci, A. Della Torre, G. Montenegro and P. D. Eggenschwiler, "On the catalytic performance of open cell structures versus honeycombs", *Chem. Eng. J.*, **264**, 514-521 (2015).

[2] A. Malevanets and R. Kapral, "Mesoscopic model for solvent dynamics", *J. Chem. Phys.*, **110**, 8605-8613 (1999).

[3] G. Gompper, T. Ihle, D. M. Kroll and R. G. Winkler, "Multi-Particle Collision Dynamics: A Particle-Based Mesoscale Simulation Approach to the Hydrodynamics of Complex Fluids", in *Advanced Computer Simulation Approaches for Soft Matter Sciences III*, Springer Berlin Heidelberg, (2009).

[4] S. Mühlbauer, S. Strobl, K.-W. Lee, T. Pöschel, "Stochastic rotation dynamics with isotropic interaction", (manuscript in preparation).

[5] O. Smorygo, V. Mikutski, A. Marukovich, A. Ilyushchanka, V. Sadykov and A. Smirnova, "An inverted spherical model of an open-cell foam structure", *Acta Mater.*, **59**, 2669-2678 (2011).

[6] J. L. Ayastuy, M. A. Gutiérrez-Ortiz, J. A. González-Marcos, A. Aranzabal and J. R. González-Velasco, "Kinetics of the Low-Temperature WGS Reaction over a $CuO/ZnO/Al_2O_3$ Catalyst", *Ind. Eng. Chem. Res.*, **44**, 41-50 (2005).

[7] G. W. Roberts and C. N. Satterfield, "Effectiveness Factor for Porous Catalysts. Langmuir-Hinshelwood Kinetic Expressions for Bimolecular Surface Reactions", *Ind. Eng. Chem. Fundamen.*, **5**, 317-325 (1966).

[8] M. Wang and Z. Li, "Simulations for gas flows in microgeometries using the direct simulation Monte Carlo method", *Int. J. Heat Fluid Flow*, **25**, 975-985 (2004).