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Influence of the macropore structure on heat and mass transfer inside catalytic washcoats

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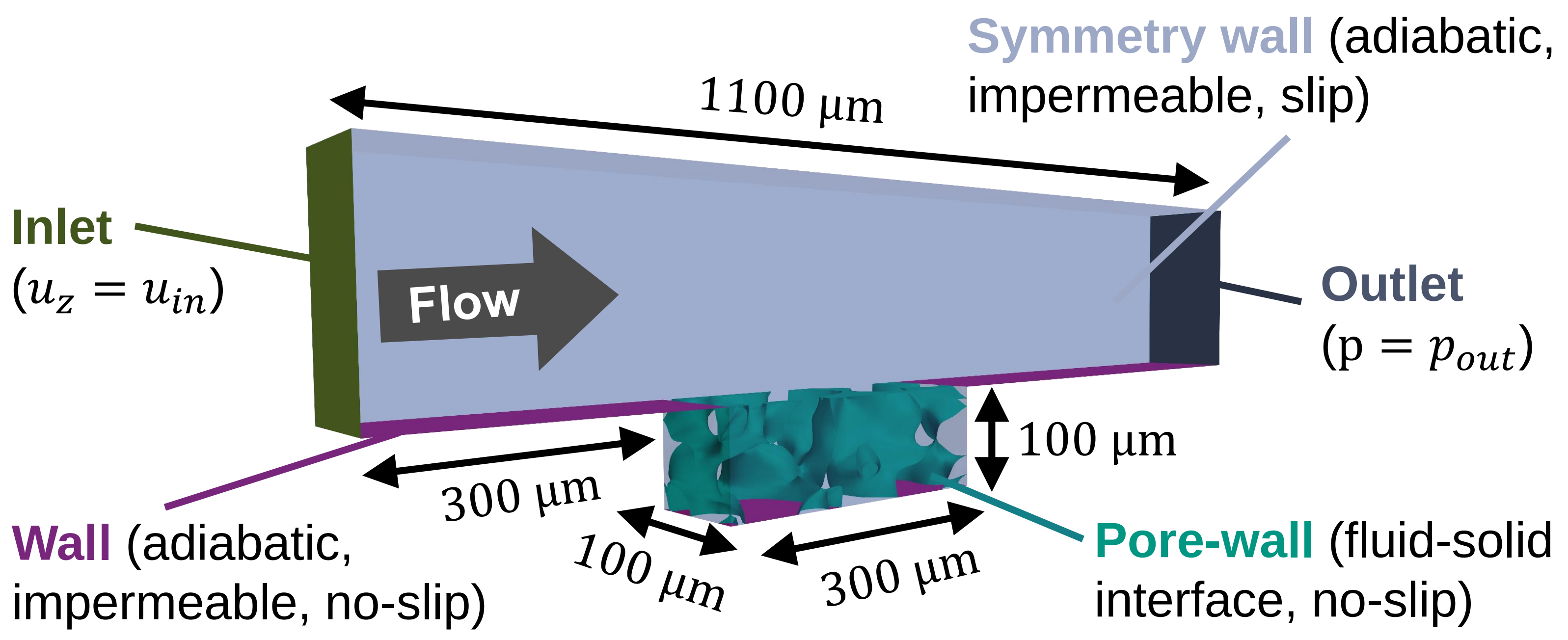
Motivation

The performance of catalytic coated monolith reactors, technically used in the reduction of pollutants and greenhouse gases, is strongly limited by heat and mass transport within the porous washcoat. Spatially resolved multiscale modelling improves the understanding of the processes inside the reactor and allows to optimize the catalyst design. In this contribution an approach to model the fluid flow inside an overflowed macroscopic pore structure coupled with transport and reaction within the mesoporous catalyst is presented. The simulations are used to study the impact of the macroscopic pore structure on transport phenomena and catalytic reaction.

Methodology

Domain and simulation setup

The pore structure (PS) is synthetically generated with a defined porosity using the Python package PoreSpy [1]. The resulting TIFF stack is converted into a surface mesh with the open-source software Blender and inplemented into the following geometry:



Symmetry wall (adiabatic, impermeable, slip)

Inlet ($u_z = u_{in}$)

Outlet (p = p_{out})

Wall (adiabatic, impermeable, no-slip)

Pore-wall (fluid-solid interface, no-slip)

The entire domain is discretized using polyhedral volume cells to solve the governing equations with the finite volume method.

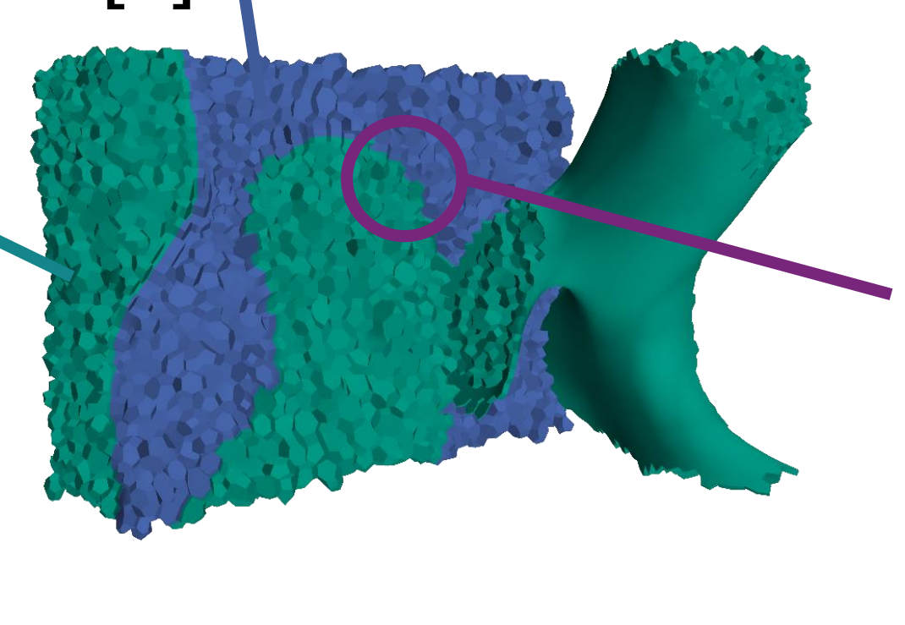
Pore-resolved CFD coupled with catalytic reaction

Fluid: Navier-Stokes equations for mass, momentum, energy and species conservation. [2]

Catalyst: Porous catalyst as pseudo continuum with effective transport parameters for heat and species transport:

$$\rho_{\mathrm{cat}} \sum_{j=1}^{N_{\mathrm{react}}} \Delta_{\mathrm{r}} H_j^{\circ} \dot{r}_j = \nabla \cdot \dot{q}_{\mathrm{eff}}$$

$$Mw_i \rho_{\mathrm{cat}} \sum_{j=1}^{N_{\mathrm{react}}} \nu_{i,j} \dot{r}_j = \nabla \cdot \dot{j}_{\mathrm{eff},i}$$



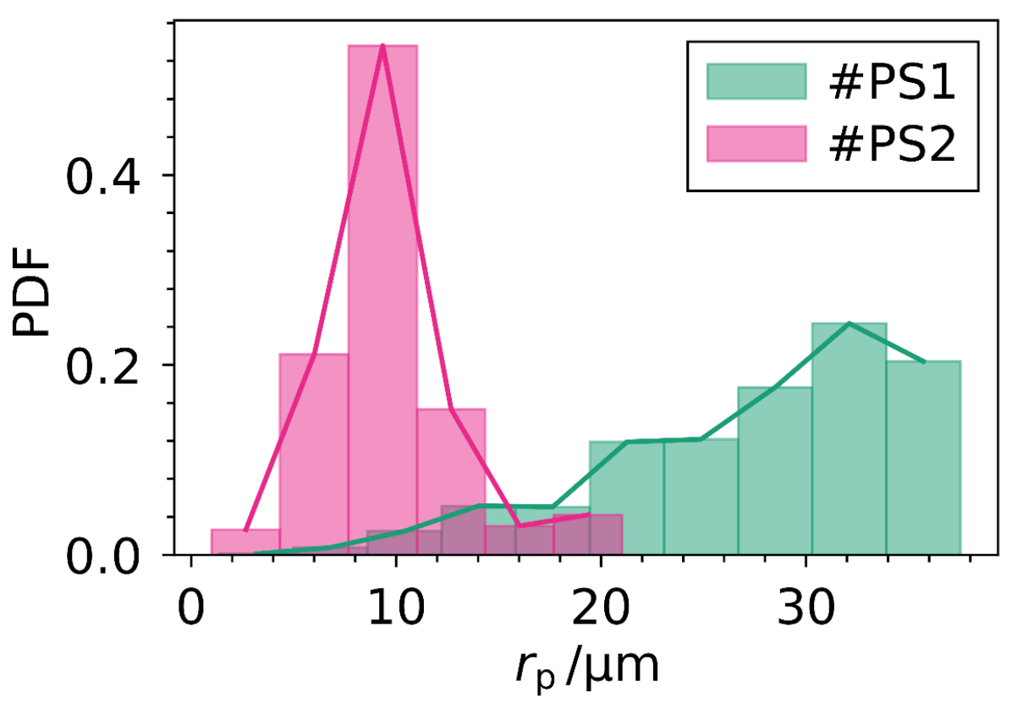
Coupling of fluid and catalyst domain via conjugated heat **(CHT)** and mass transfer **(CMT)** models [3].

- **Fickian** approach for diffusive fluxes $\dot{j}$.
- Considering **Knudsen diffusion** within the mesoporous catalyst phase by the **Wilke-Bosanquet** approximation.

As a model reaction, the oxidation of CO to CO_2 over a platinum catalyst is implemented using the reaction rate according to Shishu et al. [4].

Results

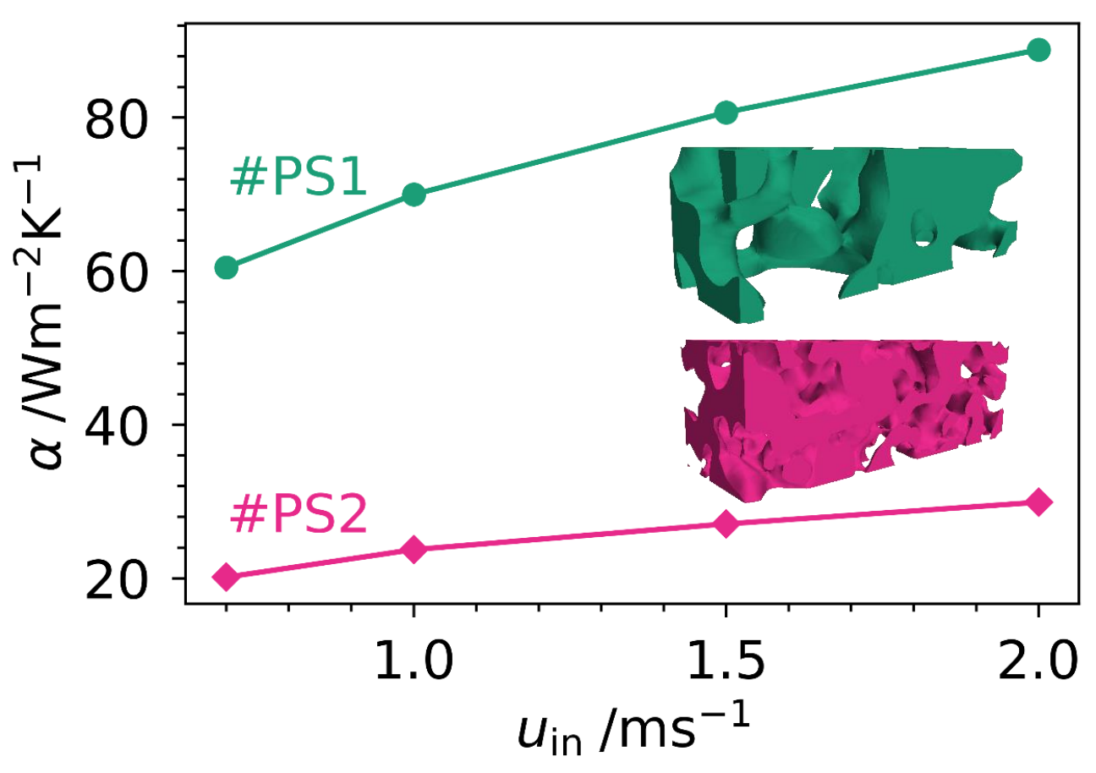
Pore-structure



	ε	A/mm^2	τ_x	τ_y	τ_z
#PS1	0.5	0.10	1.73	1.37	1.62
#PS2	0.5	0.22	1.71	1.66	1.59

Tortuosity $\boldsymbol{\tau}$ calculated with the method presented by Cooper et al. [5]

Heat transfer between pore-wall and fluid



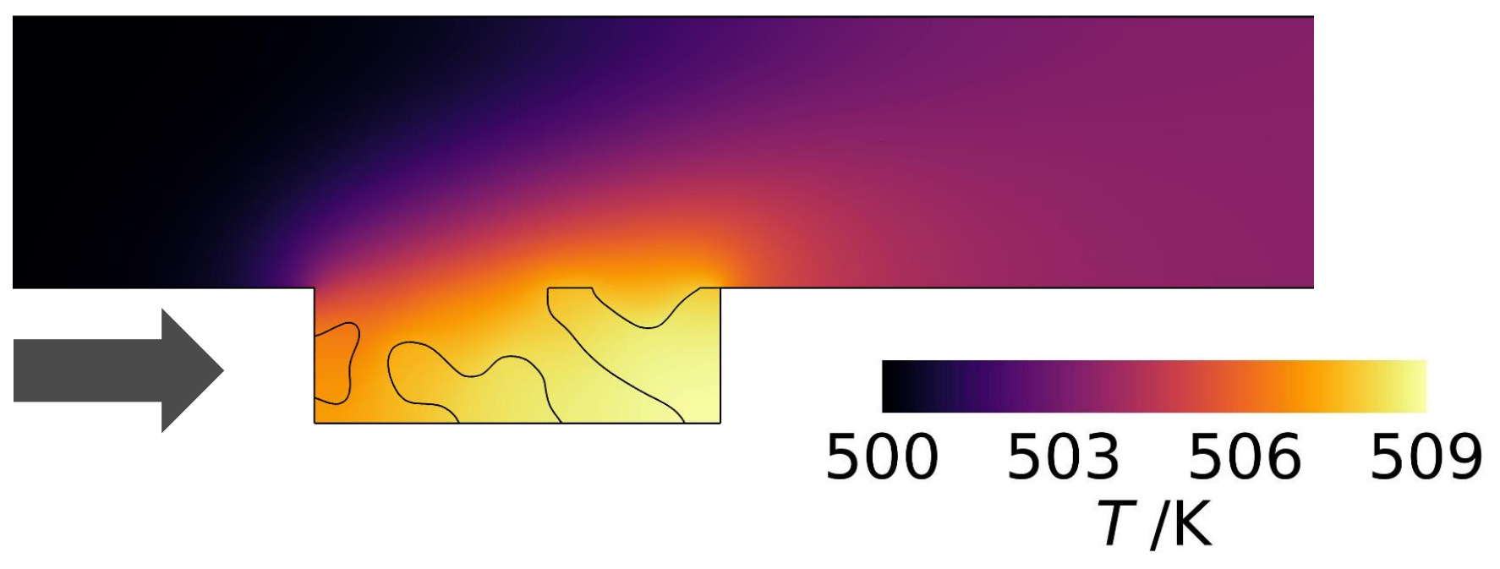
- Pore structure with smaller pores and higher surface area (#PS2) results in a **decreased heat transfer coefficient α** due to **less convective transport**
- Optimal pore structure is a trade-off between surface area and convective transport

Catalytic reaction within the pore-structure

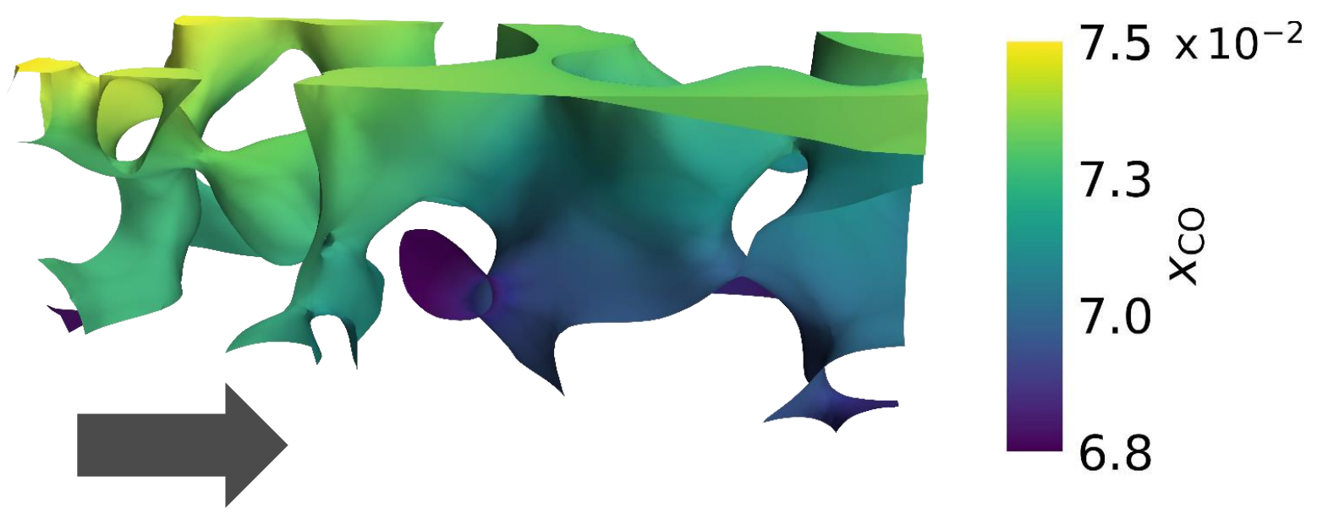
Pore-structure: #PS1; **Boundary conditions**: $T_{\mathrm{in}} = 500\ K$, $x_{\mathrm{in,CO}} = 0.075$, $x_{\mathrm{in,O_2}} = 0.039$, $x_{\mathrm{in,CO_2}} = 0$, $x_{\mathrm{in,Ar}} = 0.886$, $p_{\mathrm{out}} = 1$ atm.

Spatially resolved velocity, temperature and mole fraction $\left(\boldsymbol{u}_{\mathbf{in}} = \mathbf{1\ ms^{-1}}\right)$

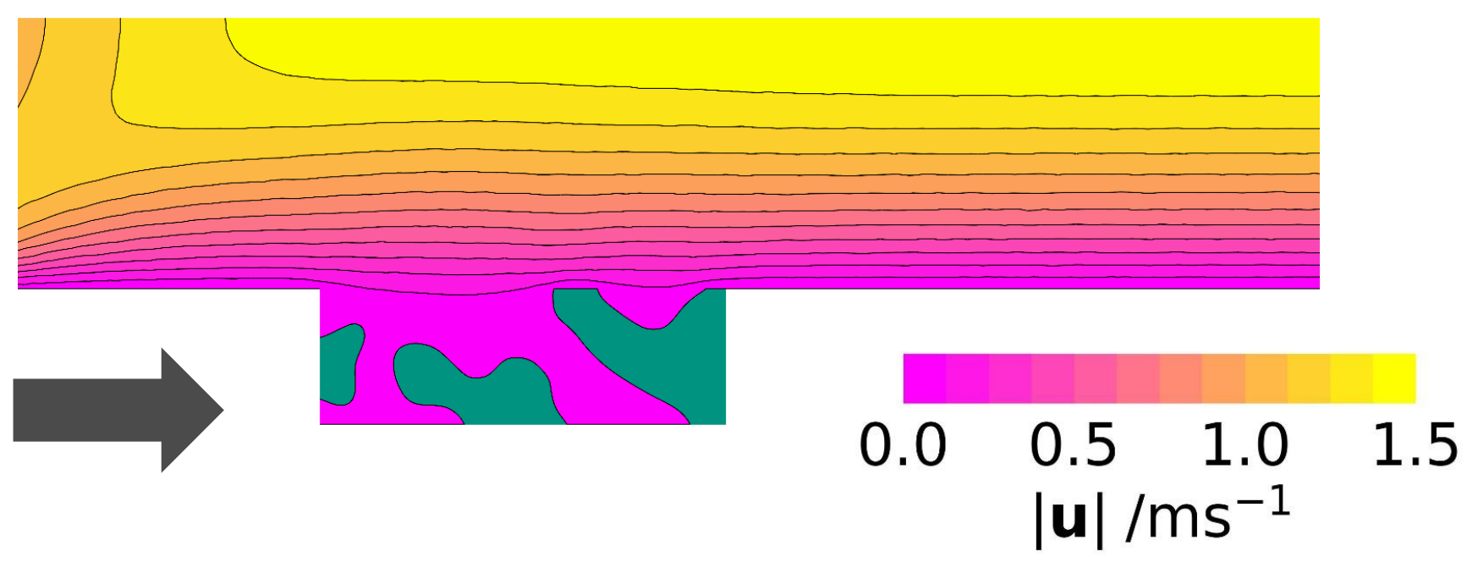
Temperature at a plane section:



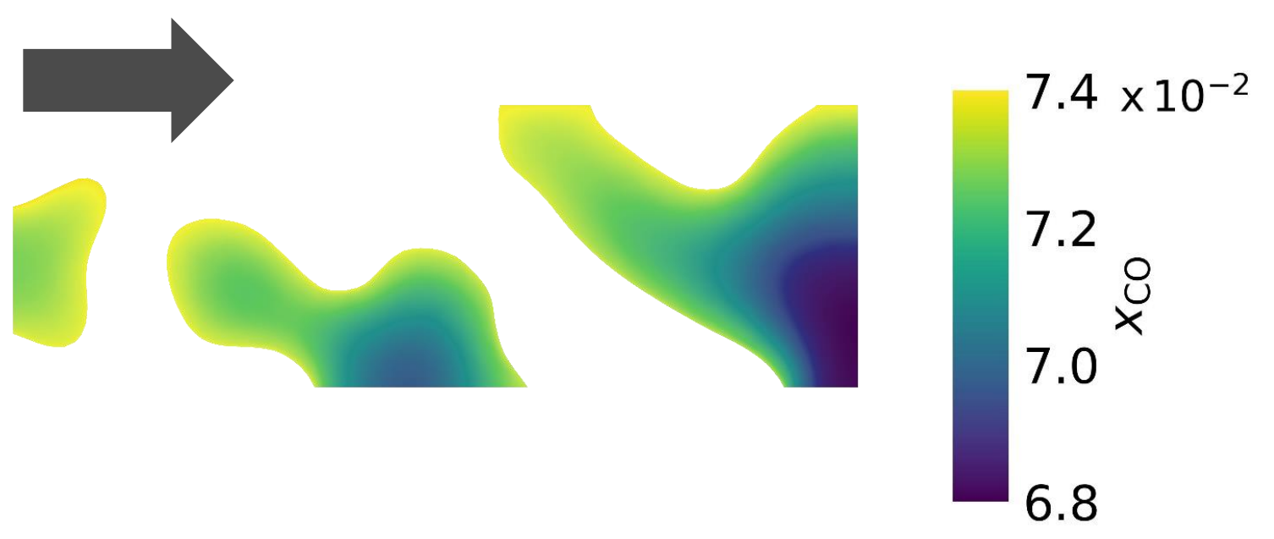
Mole fraction at the pore wall:



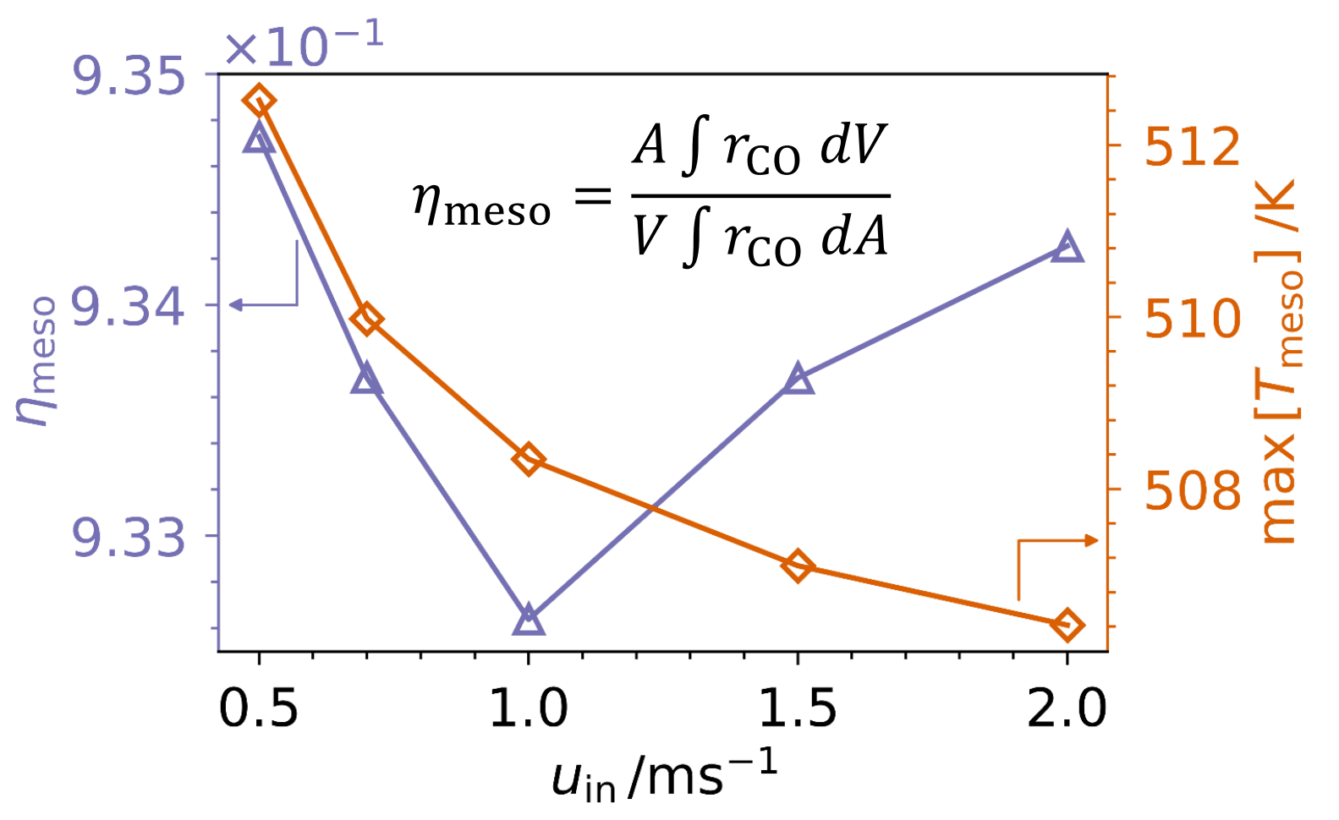
Velocity at a plane section:



Mole fraction within mesopores:



Catalyst efficiency and maximal temperature within the pore structure



- Maximal temperature in the mesoporous catalyst decreases with increasing inlet velocity.
- Inlet velocity has a small impact on the catalyst efficiency η_{meso}

Conclusion and outlook

This work demonstrates how pore-resolved simulations of catalytic washcoats can contribute to a better understanding of the interplay between transport and reaction. In the future, the non-reactive phase of the macroporous washcoat will be included and the more accurate Maxwell-Stefan diffusion approach will be implemented.

References

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[5] Cooper et al., *J. Power Sources,* 2014, 247, 1033-1039, DOI: 10.1016/j.jpowsour.2013.04.156.

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