Tomography based simulation of reactive flow at the micro-scale: Catalytic particulate filters and cauliflower-structured platinum wires

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Highlights

- 3D tomography-based pore scale simulation of particulate filter coated with catalyst
- Confinement of washcoat in the pores leads to diffusion limitations
- Tomography-based simulation of Pt-wire with cauliflower structure using surface kinetics
- Cauliflower structure significantly influences N_2O selectivity

1. Introduction

Due to the need for high contact area, most catalytic systems exhibit a µ-scale structure. The standard simulation approach is to not resolve this µ-scale structure which is then described in terms of volume averaged homogeneous model equations. Today, µ-scale geometries can be routinely obtained by x-ray tomography. In this contribution, the application of tomography-based µ-scale reactive flow simulation is demonstrated for two examples, the reactive flow in the pores of a catalytic particulate filter and the oxidation of NH_3 over a platinum wire with the typical cauliflower structure. It is demonstrated that for both cases, µ-scale transport effects have a significant effect on overall catalyst performance.

1. Simulation of reactive flow in the pores of a catalytic particulate filter

A wall segment of a catalyzed particulate filter was analyzed by X-Ray tomography with a spatial resolution of ~1.4 µm and each volume element was assigned as either pore, washcoat or substrate. The flowfield and the concentration profiles were then computed using the code PoreChem [2], assuming a first order reaction in the washcoat. The conversion in the filter wall was compared to the performance of homogeneous model with the same dimensions and catalyst content, see Figure 1b. It was found that the conversion in the pore network is lower than predicted by the homogeneous model, indicating the presence of some kind of in-pore transport limitation. Potential explanations are flow channeling through bigger pores leading to a broadening of the residence time distribution or diffusion limitations within the washcoat. It was found, that in our case the reduced conversion can nearly entirely be attributed to diffusion limitations in the wall integrated washcoat. If diffusion in the washcoat was described by a standard effectiveness factor model with the effective catalyst diameter fitted to the results of the full pore scale simulation, a very good agreement between the full simulation and the homogeneous model was found, see Fig 1b. The effective washcoat diameter obtained by this fit is surprisingly large, compared to the apparent size of the washcoat particles. This can be explained by the limited accessibility of the washcoat due to the confinement in the pore structure.

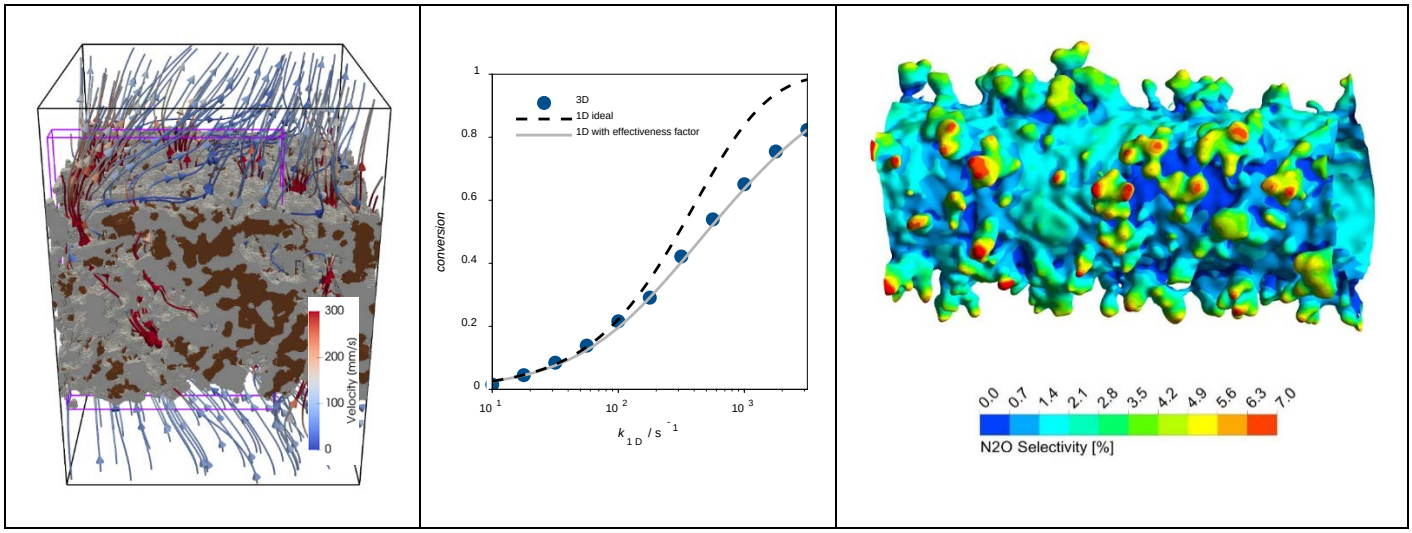

Figure 1. Left: Section of the 3D simulated domain of a GPF wall segment of cordierite (grey) with catalyst (brown). Lines show the flow field. Middle: conversion of the different models in dependence of the reaction rate. Right: Local N_2O selectivity over the cauliflower shaped platinum wire

3. NH_3 oxidation on a platinum wire with µ-scale cauliflower structure

Within a few hours of operation, Pt/Rh gauzes used for industrial NH_3 oxidation form a so-called cauliflower-structure. A typical wire geometry was obtained by µ-tomography and the reactive flow around the wire was computed using Fluent, describing the chemical reactions by a surface kinetic model [3]. It was observed that the µ-structure has a significant impact on the predicted N_2O selectivity and that the effect of the cauliflowers cannot be represented in a trivial way by a representative smooth wire. The local N_2O selectivity obtained over the cauliflower structure is shown in Figure 3c. It can be observed that the tips of the cauliflowers most exposed to the incoming flow show a significantly a significantly higher N_2O selectivity. This can be explained by increased local mass transfer coefficients and slightly higher temperatures at the exposed tips.

4. Conclusions

Two examples of tomography-based µ-scale simulation have been presented. In both cases, µ-scale transport has significant effect on the reactor performance, which would not have been captured by conventional homogeneous models. The results demonstrate the potential of tomography-based simulation for µ-scale catalyst design.

References

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Keywords
X-ray tomography, Catalyzed particulate filter, Reactive flow, NH_3 oxidation, Cauliflower structure