

Chemistry and Flow inside a Catalytic Foam Resolved by Tomography Based CFD Simulations and Reactor Profile Measurements.

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Highlights

- Tomography based CFD simulation of a catalytic foam
- Comparison of simulated and experimental profiles in the very same foam
- Systematic exploration of model deficiencies and experimental artifacts

1. Introduction

Catalytic foams have found widespread application in chemical reaction engineering [1]. In contrast to classical fixed beds consisting of randomly packed catalyst pellets, catalytic foams offer higher porosity, lower pressure drop and higher mechanical stability. In most cases, the catalytic active component is supported on a chemically inert foam support. Support materials range from alumina, titania and other ceramics over silicon carbide to metals.

The optimization of fixed-bed reactors containing catalytic foams relies on a sound model of chemistry and transport processes inside the foam. Spatially resolved measurements of species and temperature profiles through catalytic foams gained popularity in recent years and provided much insight into the interaction of chemistry and transport processes in these systems [2,3]. In most cases, experimental profiles were compared with numerical reactor simulations using pseudo-homogeneous or heterogeneous reactor models.

To explore the accuracy of the method of spatial profile measurements through catalytic foams and to identify random and systematic deviations the authors strive in the present work to model a catalytic reaction in a catalytic foam as accurately in terms of flow, heat transport, mass transport and chemistry as possible. To reach this goal the authors compare tomography based CFD simulations of the kinetically well understood oxidation of CO to CO₂ on a Pt coated α -Al₂O₃ foam with species and temperature profiles measured in the very same foam in a dedicated profile reactor. Tomography delivers the accurate geometry for the model including channel and sampling capillary. Discrepancies between model and experiment are systematically explored.

2. Methods

μ -CT measurements were conducted on an EMPYRAN X-Ray diffractometer (Panalytical) using a sample stage, a Mo X-Ray tube and the GaliPIX3D detector. Tomography object was a 45ppi α -Al₂O₃ foam coated with Pt nanoparticles (Fig. 1). The average particle size was 51nm determined by SEM. Before coating with Pt, a central channel was drilled through the foam fitting a 700 μ m sampling capillary with a 100 μ m side sampling orifice. After tomography, the same foam was mounted in a profile reactor described earlier [4]. Species and temperature profiles were measured for CO oxidation to CO₂. CFD simulations were carried out in Star CCM+ 11.04 using the reconstructed geometry of the foam, a polyhedral mesh of the fluid and the solid domain and two layers of prism mesh at each solid surface. Laminar flow (Re<10), conjugated heat transfer and surface to surface radiation was included in the model. A surface microkinetic model for CO oxidation on Pt was taken from Deutschmann et al. [5].

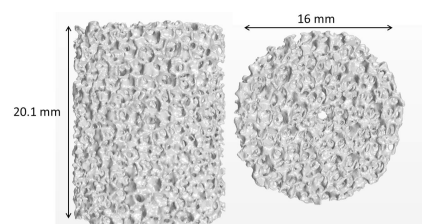


Fig. 1 Reconstruction of the foam geometry from tomographic measurements.

3. Results and discussion

Figure 2 shows simulation plots of pressure, velocity and temperature in a central cut of the foam in flow direction. The reactor wall and the wall of the sampling capillary were set adiabatic in this simulation.

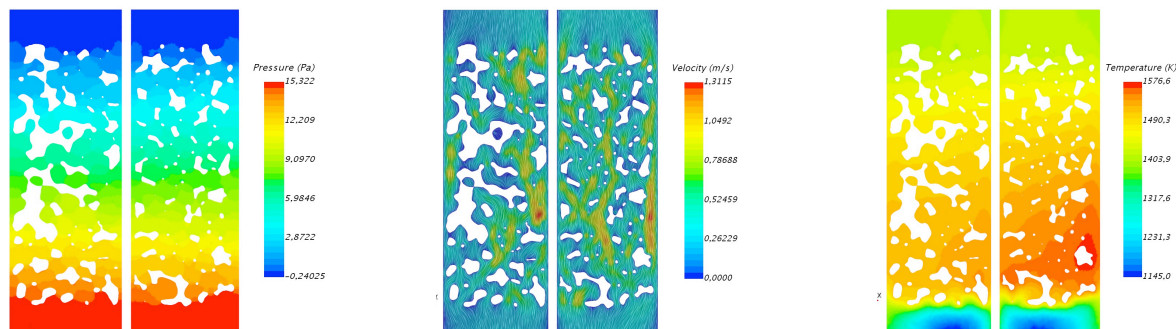


Fig. 2 Simulated pressure field (left), velocity field (middle) and temperature field (right) inside the foam.

The pressure drop through the foam is very small (~ 15 Pa) and there is basically no radial pressure gradient. The velocity field inside the foam is very inhomogeneous. Bypass flow occurs in between the struts. Flow velocities in front of and in the wake of the struts are low. Cross mixing is small. Temperature gradients are observed in flow direction and perpendicular to it. Fig. 3 compares experimental and simulated species and temperature profiles along a randomly selected scan line. Good agreement is found for CO, O₂, and CO₂. Simulation results along other scan lines (not shown) show that deviations are due to the random nature of the foam. Systematic deviations between simulation and experiment are found for the temperature profile. While the simulated profile peaks at about 2.5mm and declines slowly afterwards, the measured hotspot is right at the foam inlet (0mm) followed by a much steeper decline than simulated. The measured temperature maximum at 0mm must be an artifact. It is physically impossible because there is no other heat source than the reaction. The tip of the pyrometer fiber collecting heat radiation from the foam surface must look “ahead”. If it is nominally positioned at 0mm, the fiber collects radiation from about 2.5mm downstream. The steeper descent of the experimental temperature indicates that the reactor is not adiabatic. There must be heat losses through the reactor wall despite insulation. Further aspects such as influence of the pore structure on the profiles, mass and heat transport limitations; accessible surface sites, surface coverages, radial mixing and disturbance by the capillary will be discussed in the paper as well.

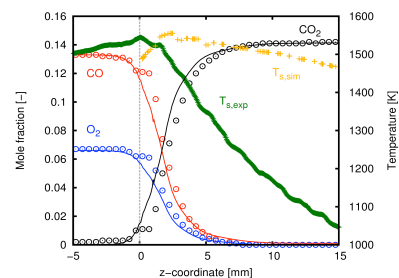


Fig. 3 Comparison of experimental and simulated species and temperature profiles.

4. Conclusions

Profile measurements through catalytic foams provide insight into chemistry and transport processes in these systems. A quantitative evaluation of the profiles requires consideration of random effects such as the pore structure and systematic influences such as temperature offsets and heat losses.

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Keywords

catalytic foam, CO oxidation, platinum, tomography based CFD, species and temperature profiles