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Microtomography-based CFD modeling of a fixed-bed reactor with an open-cell foam monolith and experimental verification by reactor profile measurements

Ying Dong[a], Oliver Korup[a], Julian Gerdts[a], Beatriz Roldán Cuenya[b], Raimund Horn[a,*]

[a] Institute of Chemical Reaction Engineering, Hamburg University of Technology, Eißendorfer Str. 38, D-21073 Hamburg, Germany
[b] Department of Interface Science, Fritz-Haber-Institute of the Max-Planck Society, D-14195 Berlin, Germany

HIGHLIGHTS

- Microtomography-based CFD simulations of a catalytic foam monolith are presented.
- Sub-millimeter resolved species and temperature profiles were measured.
- Interplay between transport and chemistry was studied in detail.
- A critical comparison between simulation and measurement data was made.

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ABSTRACT

CFD simulations of catalytic reactors provide detailed insight into the chemical and physical processes within these devices such as 2D or 3D concentration-, flow- and temperature fields or even 3D pictures of the coverages of the various species on the surface of the catalyst. The validation of CFD models of catalytic reactors is hampered by the lack of experimental data against which the simulation results can be compared. The present work addresses this problem by presenting a critical comparison between CFD simulation results and sub-millimeter resolved species and temperature profiles measured through a reactor employing a catalytic foam monolith. CO oxidation on Pt nanoparticles with narrow size distribution supported on an α-Al_2O_3 foam was chosen as simple catalytic system with well known microkinetics. To keep uncertainties in the CFD geometry as small as possible, the structure of the foam catalyst was resolved by X-ray microtomography. Simulation parameters and boundary conditions were determined as accurately as possible. The CFD model includes flow, thermal conduction in the struts of the foam, conjugated heat transfer and heat radiation. Catalytic chemistry is incorporated by means of a microkinetic reaction model taken from literature. The comparison of CFD simulation results with high resolution spatial temperature and concentration data allows a critical assessment of strengths and weaknesses of both, the model and the experiment serving as basis for a knowledge-based design of reactors employing catalytic foams or similar random geometries.

1. Introduction

Foam monoliths made from mechanically stable, chemically inert and temperature resistant materials like refractory oxides, ceramics, silicon carbide or metals have attracted much interest as catalyst support in catalytic fixed-bed reactors. Due to their high porosity, foam monoliths generate low pressure drop even at high gas flow rates. High convection in the interconnected macro-pores enhances mass transfer, heat transfer and radial mixing [1–4]. If the foam is made of materials of high thermal conductivity, heat transport in the struts of the irregular cell network increases which can be beneficial for exothermic reactions suffering from selectivity loss or hot-spot formation [5]. The intrinsic low specific surface area of foam supports can be increased by a washcoat carrying finely dispersed catalyst particles [6].

Strong modeling efforts have been made to understand the physical and chemical processes occurring inside fixed-bed reactors with foam monoliths [7,8]. Much attention has been paid to hydrodynamics, heat transport inside the pore network [9–11] and heat transfer to the

* Corresponding author.
E-mail address: horn@tuhh.de (R. Horn).

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Nomenclature

Abbreviations

μ-CT	computed microtomography
BHS	back heat shield
FHS	front heat shield
PPI	pore per inch

Greek letters

β	temperature exponent [–]
δ_{ij}	Kronecker delta [–]
Γ	surface site density [mol/m^2]
κ	bulk viscosity [Pa·s]
λ	thermal conductivity [W/m/K]
μ	dynamic viscosity [Pa·s]
ρ	fluid density [kg/m^3]
σ	coordinate number [–]
τ_{ij}	stress tensor
θ	surface coverage [–]

Latin letters

$\dot{s}_i$	molar net production rate of species i [mol/m^2·s]
$\vec{j}_i$	diffusion mass flux [kg/m^2·s]
A_{cat}/A_{geo}	ratio of catalytically active area to geometric area [–]
c_i	species concentration [mol/m^3 or mol/m^2]
c_p	specific heat capacity [J/kg·K]
$D_{i,m}$	diffusion coefficient of species i in the mixture [m^2/s]
$D_{k,i}$	binary diffusion coefficient [m^2/s]
h	specific enthalpy [J/kg]
K_s	number of elementary surface reactions [–]
M_i	molar mass of species i [kg/mol]
N_g	number of gas phase species [–]
N_s	number of adsorbed species [–]
p	pressure [Pa]
q'_{rad}	radiant flux [W/m^2]
R	ideal gas constant [J/K·mol]
R_i^{het}	net rate of production of species i due to heterogeneous reactions [kg/m^2·s]
u	velocity [m/s]
X_i	molar fraction of species i [–]
Y_i	mass fraction of species i [–]
x_i	coordinate in i direction [m]

reactor wall [12,13]. Heat and mass transport correlations have been derived from simulation data, which can be applied for reactor design and development [14,15,6,16–18].

In general, there are two approaches to model the foam structure. The unit cell approach [19] idealizes a foam monolith by periodic repetition of a representative unit cell in 3D space (e.g. Kelvin cells) [20]. The other approach is to simulate flow and transport using the foam geometry reconstructed from X-ray computed microtomography (μ-CT) scans [9,10]. μ-CT is more costly, but it captures the irregular and randomized foam structure in high fidelity. μ-CT based CFD simulations deliver results closer to reality because the foam geometry in the model is the very same as that in the reactor.

In order to gain confidence in μ-CT based CFD simulations, experimental validation is required. High resolution temperature and species concentration profiles measured through the centerline of foam catalysts offer currently the highest data point density for model validation [21,22]. Spatial reactor measurements have also been used for validating CFD simulations of fixed-bed reactors with random packings of spherical [23] and non-spherical particles [24]. In those studies, the stochastic packing geometry is generated by DEM simulations. It represents the packing in the reactor within statistical bounds but it is not an exact copy of it. If the packing geometry is reconstructed from μ-CT scans, as demonstrated in the present work for a foam monolith, the uncertainty in the bed structure is close to zero. Deviations between model and experiment are reduced to deficits in the kinetic model of the catalytic reaction, shortcomings in the description of momentum, mass and heat transport, inaccurate boundary conditions or erroneous experimental data. In the current work, we present, according to our knowledge for the first time, a comparison between temperature and concentration profiles measured through a foam catalyst under reaction conditions and μ-CT based CFD simulations of these data. We strive to keep uncertainties in the model as small as possible by choosing CO oxidation on Pt as a test reaction with well-known microkinetics, by using Pt nanoparticles with a narrow size distribution on a plain 45 ppi

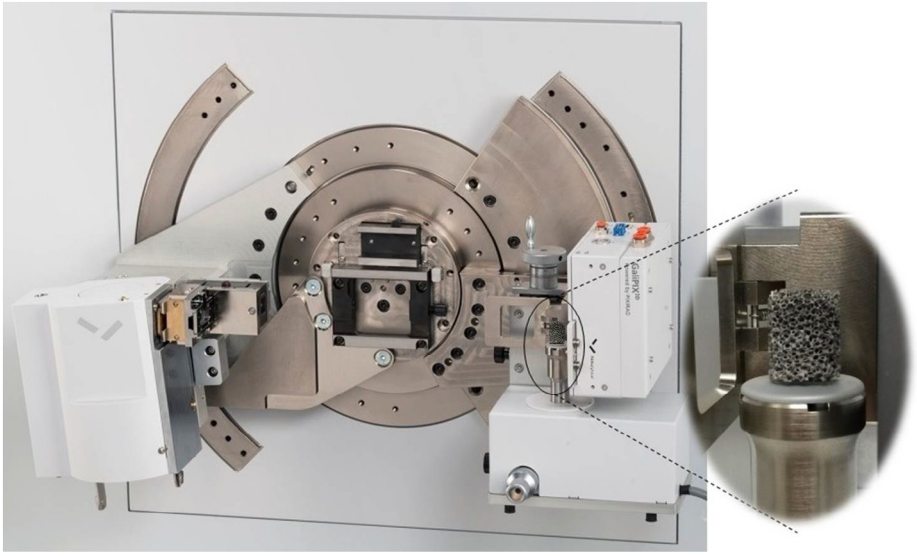

Fig. 1. Photography of the PANalytical Empyrean X-ray diffractometer operated in tomography mode: Molybdenum X-ray tube (left), rotatable sample stage with catalyst foam (zoomed inset) and GaliPIX^{3D} detector (right).