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Transport and reaction in catalytic wall-flow filters

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Abstract

Diesel particulate filters composed of so-called wall-flow monoliths are well established devices for diesel particulate abatement. Recent improvements in production technology allow implementation of full-featured catalyst functionality in the filter walls.

From a reactor engineering point of view such wall-flow reactors with wall-integrated catalyst show fundamental differences compared to conventional flow-through monoliths. The complex interactions of convection, diffusion and reaction in the wall-flow monolith are studied by means of numerical simulation. A two-dimensional model for the flow in one pair of inlet/outlet channels with a generic first order reaction in the catalytic filter wall is developed. Concentration profiles in the reactor and a conventional flow-through catalyst are compared.

It is found that in the range of moderate reactor conversion concentration gradients along the inlet channel of the filter are small. Thus the reactor can be described by an approximate one-dimensional model, taking into account only the radial flux through the filter wall and assuming a constant inlet concentration in axial direction along the inlet channel.

Light-off curves are computed for the wall-flow and for the conventional flow-through monolith. Significantly better conversion is found for the wall-flow configuration. This can be explained by mass transfer limitation in the conventional flow-through monolith.

Keywords: Wall-flow monolith; Diesel particulate filter; Fluid dynamic; Simulation; Modeling

1. Introduction

Diesel particulate filters are well established tools for the reduction of particulate emissions from diesel exhaust [1,2,3a]. Today, diesel particle filters usually consist of ceramic monoliths with alternately blocked inlet and outlet channels. Thus the exhaust gas is forced through the wall and the soot is collected on the surface of the inlet channels. Monoliths designed like this are therefore referred to as ‘wall-flow monoliths’.

The major technological challenge for the application of particulate filters in passenger cars is the controlled regeneration of the filters by periodic oxidation of the soot at increased temperatures. The soot ignition temperature for the uncatalyzed oxidation of diesel soot is above 600 °C [3b]. This temperature can be lowered by catalytic activation of the soot. For this purpose the catalyst can either be introduced directly into the soot utilizing a fuel additive or otherwise a ceramic filter with a catalytic coating can be used. The later design is generally referred to as a ‘catalyzed particulate filter’.

Catalyzed particulate filters well known for many years are currently employed in a number of series production vehicles [4]. Early work on catalytically activated particulate filters focused entirely on the coating’s ability to support filter regeneration. Recent development aimed at the expansion of the catalytic filter functionality. Due to latest improvements in the production process, most of the conventional catalyst functionalities now can also be implemented in the wall of a particulate filter.

From the reaction engineering point of view, the wall-flow filter and the conventional flow-through catalyst are fundamentally different systems. Intuitively, the differences are obvious: In the conventional flow-through monolith convective mass transport through the reactive medium is

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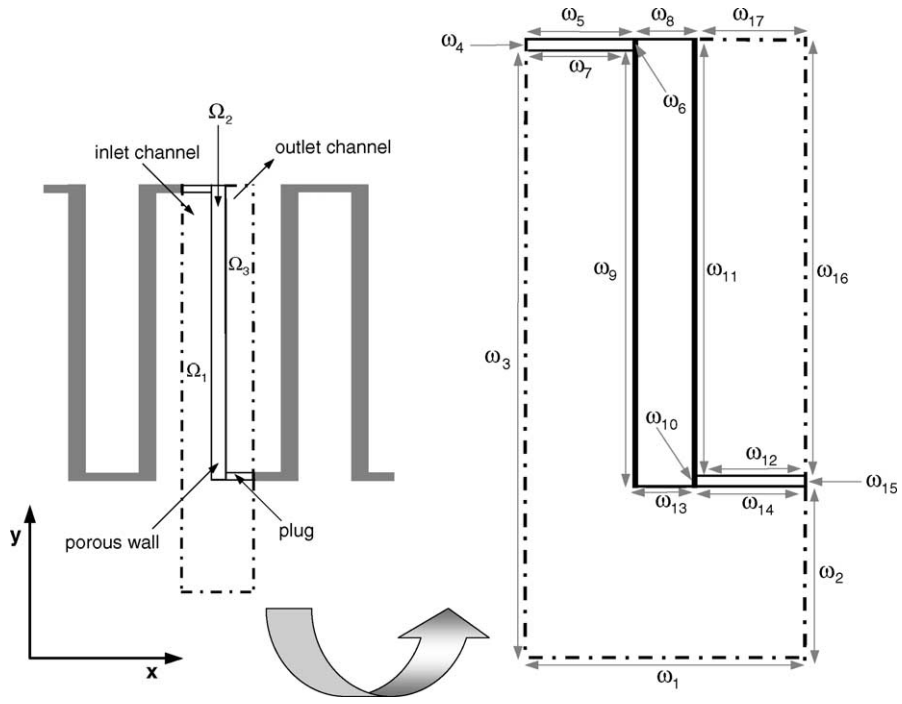

Fig. 1. Computational domains and boundaries for the 2D model (up scaled graph at the right).

impossible. Usually, the reaction is mass transfer limited by diffusive transport of the reactants into and out of the washcoat. Furthermore the reaction coordinate is oriented along the channels (*y*-direction, Fig. 1). In contrast the reactants are forced through the catalytically active wall in the wall-flow filter case. Thus, the mass transport into the reactive medium is a combination of convective and diffusive transport. Depending on the relative importance of convection and diffusion the reaction coordinate is located either along the channels (*y*-direction) or perpendicular to the channels (*x*-direction, Fig. 1).

This work focuses on numerical simulations of concentration distributions for a generic first order reaction in a clean wall-flow monolith. Thereby a fundamental understanding of the complex interaction of convection, diffusion and reaction in the wall-flow monolith reactor should be obtained. By means of the simulations the behavior of the wall-flow reactor is compared with the conventional flow-through monolith reactor.

2. Two-dimensional model

The two-dimensional model of the wall-flow monolith consists of one inlet channel (domain Ω_1), the porous wall (domain Ω_2) and one outlet channel (domain Ω_3) (Fig. 1). Due to model symmetry only one half of the inlet and outlet channels need to be considered. Dimensions of the modeled geometry are presented in Table 1.

The flow field is expressed by the Navier–Stokes Eq. (1) in the subdomains Ω_1 and Ω_3 combined with the continuity Eq. (2) [5]. The flow through the porous medium (Ω_2) is described by a combination of the continuity Eq. (2) and the Brinkman Eq. (3) [6], which is an extension of the Darcy equation. These Eqs. (1)–(3) are given below for the steady-state model:

Table 1
Dimensions of the simulated wall-flow filter, depicted in Fig. 1

Boundary	Length [mm]
ω_1	1.9
ω_2	20
ω_3	70
ω_4, ω_6, ω_{10}, ω_{15}	1
ω_5, ω_7, ω_{12}, ω_{14}, ω_{17}	0.77
ω_8, ω_{13}	0.36
ω_9, ω_{11}, ω_{16}	50

$$\rho \cdot (\mathbf{u} \cdot \nabla)\mathbf{u} = -\nabla p + \eta \cdot \Delta \mathbf{u} \quad (1)$$

$$\nabla \cdot (\rho \cdot \mathbf{u}) = 0 \quad (2)$$

$$-\eta \cdot \Delta \mathbf{u} + \kappa \mathbf{u} = -\nabla p. \quad (3)$$

The Nabla-operator ($\bigtriangledown$) represents the first order spatial derivatives, and the Laplace-operator (Δ) stands for the second order spatial derivatives, p denotes the excess pressure. Density is calculated by the ideal gas law. To calculate the dynamic viscosity (η), experimental data for air are used [7] and constant value of 0.55 μm^2 is assumed for the permeability (κ).

The following boundary conditions were used to solve the impulse balances (Table 2). Where **g** is the tangential vector