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A simulation study on the conversion efficiency of catalytically active particulate filters

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HIGHLIGHTS

- Conversion efficiency of catalyzed particulate filters was studied.
- Conversion studied as function of the governing dimensionless parameters.
- Conversion efficiency of catalysed filter is very close to ideal plug flow reactor.
- Systematic comparison of filter reactor and conventional open monolith.

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ABSTRACT

A catalytically active particulate filter with a first order catalytic reaction taking place inside the filter walls is investigated by numerical simulation. The conversion efficiency for different channel geometries and operating conditions is systematically studied as a function of the governing dimensionless parameters. It is found that the conversion efficiency of a catalytically coated wall flow filter is very close to that of an ideal plug flow reactor over the full range of realistic operating conditions. Only in a range of intermediate residence times, the filter reactor shows some diffusion limitation which leads to conversion efficiencies slightly below that of the plug flow reactor. In all cases, these deviations from ideal conversion behaviour are below 15%.

If the filter and the open monolith are compared at identical operating conditions and channel geometries, for fast reactions, the filter reactor shows higher conversion efficiency than the open monolith, since in this case the open monolith becomes strongly mass transfer limited.

However, there can be a small range of conditions where the monolith is slightly more efficient than the filter. The reason for this effect is that due to the thinner washcoat layer the onset of mass transfer limitation in the coated monolith is shifted to higher reaction rates, compared to the filter reactor.

1. Introduction

Today, Diesel Particulate Filters (DPFs) are standard emission control devices for diesel engines (Johnson, 2014). While the main purpose of the filter is to remove particulate matter from the exhaust, recently there is a strong interest to integrate additional catalytic functionality into the walls of the filter. The main motivation for the development of such multifunctional filters is to reduce the total number of devices in the exhaust system, resulting in a reduced packaging complexity and a lower overall system cost. Furthermore, an integrated catalyst/filter device shows an improved heat up behaviour due to a reduced thermal capacity of the exhaust system (Lee et al., 2008; He et al., 2009). In recent years, a variety of different catalyst functionalities has been implemented in the Diesel Particulate Filter, including the Diesel Oxidation Catalyst (DOC) (Russell and Epling, 2011; Dardiotis et al., 2008) and the Selective Catalytic Reduction (SCR) of NO_x by ammonia (Ballinger et al., 2009; Tan et al., 2011; Rappe, 2014). Future legislation will also require particulate filters for certain gasoline vehicles. This has led to the development of the so called Gasoline Particulate Filter (GPF) that implements a three-way-catalyst in the walls of a particulate filter (Saito et al., 2011; Richter et al., 2012; Spiess et al., 2013; Chan et al., 2014)

The most common type of particulate filters used in automotive applications nowadays is the ceramic filter that is derived from an open flow-through monolith by plugging alternating channels at the inlet and the outlet so that the gas is forced to flow through the wall. In a catalysed filter, a catalytically active material is

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deposited inside the porous wall. From a reactor engineering point of view, there are fundamental differences between a catalysed filter and the conventional open monolith coated with a thin catalytic layer. While the open flow-through monolith relies on diffusion for the transport of reactants and products between the gas phase and the catalytically active washcoat, in the wall-flow filter the reactants are forced through the catalytically active wall by convection. Due to its specific heat- and mass transport characteristics, the filter reactor has also been proposed for other industrial applications besides exhaust catalysis. Potential examples are selective oxidation reactions (Votsmeier et al., 2009) and the water gas shift reaction (Palma et al., 2014; Palma et al., 2015). Further commercial development of such applications requires a much better understanding of the complex interplay of convective and diffusive transport with chemical reaction in the catalysed filter.

There are a number of simulation studies that analyse the steady state conversion behaviour of the catalysed filter and compare its performance to the conventional flow through monolith. Knoth et al. (2005) compared catalysed filters and open monoliths with identical channel geometry and demonstrated that for fast reactions the catalysed filter shows higher conversion than the open monolith. This result could be explained by the absence of diffusion limitations in the filter configuration. A more detailed analysis of the complex interplay of diffusion, convection and reaction in the wall flow reactor has been provided by Votsmeier et al. (2007). It was shown that for the wall-flow filter operation two limiting cases can be distinguished:

- At low reaction rates and low flow velocities, mass transfer in radial direction is entirely dominated by diffusion. This means that radial concentration gradients between the inlet/outlet channel and the wall are negligible. The main concentration gradient is oriented in axial direction, just as in the case of the flow-through monolith.
- If reaction rates and flow velocities are high, radial transport is dominated by convection. In this case axial gradients along the channels become negligible and the main gradient is oriented in radial direction through the filter wall.

Under most operating conditions relevant in automotive exhaust catalysis, the filter is operated in an intermediate range where diffusion and convection both contribute to radial transport.

A model based analysis of the differences between the flow-through and wall-flow reactor concept has also been presented by Dardiotis et al. (2006). This study confirms the conclusion that under steady state operation the wall-flow filter is expected to show higher conversion in those situations where the conventional flow-through monolith becomes mass transfer limited (short residence times, high reaction rates).

There are also a number of simulation studies that focus on specific types of catalysed filters. In (Opitz et al., 2014) the cold start behaviour of gasoline particulate filters with a three way catalyst deposited in the filter wall was analysed and its performance was compared to a conventional flow through three way catalyst.

Park et al. Park and Rutland (2013) performed a simulation study of a Diesel Particulate Filter with an SCR catalyst incorporated into the wall. They compared the performance of the filter to the performance of a conventional open SCR catalyst and found that the wall flow filter shows a lower conversion than the open monolith. Karamitros and Koltsakis (Karamitros and Koltsakis, 2014) performed a similar comparison but covered a large range of flow velocities. At low flow velocities the conversion of the open monolith was higher than that of the filter, which is in line with the results of Park et al. Park and Rutland (2013). However, at high flow velocities the ranking was reversed and the wall flow filter showed a better performance. Redaelli et al. investigated the effect of the sample size on SDPF performance and found that very small samples (4–9 channels) showed a reduced conversion efficiency. The effect could be explained by increased diffusion limitation in the peripheral walls (Redaelli et al., 2012).

For the conventional open monolith reactor, the interplay of convection, diffusion and reaction has been studied over a long period of time (Aris, 1975). Considering a first order reaction, the reactor efficiency can be understood in terms of a few dimensionless numbers like the Thiele modulus and the Biot number. A similar analysis for the wall-flow filter reactor is still pending and will be addressed in the current paper. The objective is to first identify the dimensionless quantities that determine the conversion efficiency of the catalysed filter and then to systematically investigate the filter's conversion efficiency as a function of these dimensionless numbers. A further objective of this paper is to provide a systematic comparison of the filter reactor and the conventional open monolith with respect to their conversion efficiency.

2. Description of the numerical model

2.1. 1D+1D model of the wall-flow filter

The reactor model used in this work was previously published in (Opitz et al., 2014). It solves the governing equations for the conservation of momentum, energy and mass for one representative pair of inlet and outlet channels and the corresponding wall. One-dimensional velocity, temperature and concentration profiles are computed for the inlet and outlet channel with radial transport effects described by the corresponding transfer coefficients. For each axial location, the concentration profiles in the wall are resolved in radial direction whereas radial heat conduction is assumed to be fast so that one radially averaged temperature is computed for the wall. A schematic side and front view of the filter geometry implemented in the 1D+1D model is given in Fig. 1.

2.2. Momentum transport

The one dimensional governing equation for momentum transport of the exhaust gas in the inlet ($j=1$) and the outlet ($j=2$) channel is described by:

$$\rho_j \cdot \frac{d}{dz}\left(v_j^2\right) = -\frac{dp_j}{dz} - F_j \cdot \frac{\eta_j \cdot v_j}{d_{\text{channel}}^2} \quad (1)$$

Where v_j is the radially averaged gas velocity, p_j the average gas pressure and η_j the dynamic viscosity. F_j is the momentum transfer coefficient for fully developed laminar flow in a square tube ($F_j=28.454$) (Konstandopoulos and Johnson, 1989).

2.2.1. Pressure drop across the porous wall

The pressure drop across the porous wall, which couples the inlet and outlet channel, is calculated using the Darcy law Eq. (2) for porous media.

$$\Delta p = \frac{\eta \cdot u_{\text{wall}}}{k_{\text{wall}}} \cdot d_{\text{wall}} \quad (2)$$

Thereby k_{wall} is the wall permeability of the filter wall including the washcoat inside the pores. This combined wall permeability is determined by fitting the simulation model to pressure drop experiments. Therefore the filter's pressure drop was measured at