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A one-dimensional modeling approach for dual-layer monolithic catalysts

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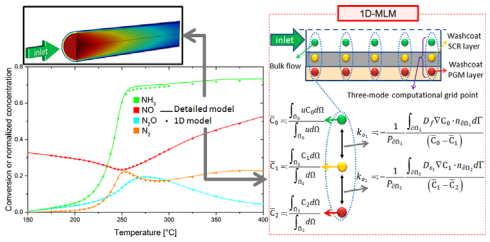
HIGHLIGHTS

- A 1D model for monolithic catalysts with dual-layer washcoat (DLWC) is derived.
- Analytical expressions for mass transfer coefficients for circular channels in DLWC.
- Expressions of internal Sherwood numbers based on Thiele moduli in DLWC.
- Model is applicable for control and optimization of automotive catalysts.
- Numerical simulation of dual-layer SCR/PGM catalysts.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper presents a model extension to dual-layer monolithic catalysts from a one-dimensional (1D) (Balakotaiah, 2008; Joshi et al., 2009) mathematical model used for single-layer monolithic catalysts. The novel transient non-isothermal model computes average concentrations inside the bulk flow and in each layer of washcoat at every computational grid point along the monolith channel. The model has the capability that physical (e.g. diffusivity, pore radius, and thickness) and chemical (reaction kinetics) properties of the two layers of the washcoat can be different from each other. The relatively low computational cost offers real-time simulations of multi-layer monolithic catalysts. Analytically derived expressions for transverse mass transfer coefficients between the bulk flow and washcoat layers are presented for single-layer and dual-layer catalysts with circular cross-section, which can be easily extended for multi-layer catalysts. In addition, three expressions are presented for calculation of three internal Sherwood numbers, which are used in the dual-layer model, based on the values of Thiele moduli of the washcoat layers. These expressions are especially useful for calculation of the mass transfer coefficients for dual-layer catalysts with non-circular cross-sections. Application limitations of the 1D multi-layer model with respect to physical and chemical operating conditions are discussed. The derived model is applied for an automotive aftertreatment catalyst and the results of this model are compared with the results of a detailed 2D model (selective catalytic reduction/platinum group metals dual-layer catalysts).

1. Introduction

Monolithic honeycombs are widely used in environmental catalysis. Their low pressure drop has made them a common catalytic converter in gas turbines (Forzatti and Groppi, 1999;

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Carroni et al., 2002; Groppi et al., 1999) and automotive vehicles (Kašpar et al., 2003; Geus and van Giezen, 1999; Guettel and Turek, 2009; Koltsakis et al., 1998) to control the emission level of outlet gases. Hence, wide ranges of studies have been carried out in order to increase the efficiency and/or to reduce the overall production cost of the monolithic catalysts (Guettel and Turek, 2009; Koltsakis et al., 1998; Ji et al., 2008; Kočí et al., 2004; Kröcher et al., 2006; Clayton et al., 2008). The models for numerical simulation of monolithic catalyst vary from simplified 0D and 1D to 2D and 3D CFD models. 3D and 2D models are expected to provide more accurate results, while 1D and 0D models benefit from lower computational costs. Raja et al. (2000) investigated three models (Navier-Stokes, boundary-layer, and plug flow) for steady-state behavior simulation of a single monolithic channel. They concluded that among the three models, the Navier-Stokes model provides valid results for a wide range of settings but it is computationally expensive, while the plug-flow model has lowest computational cost, however, with a limited validity of the results. Mladenov et al. (2010) studied the effect of using effectiveness factor and reaction–diffusion approaches for consideration of diffusion limitation within washcoat on the accuracy of the results of three models for channel performance simulation (1D plug-flow, 2D boundary-layer and Navier-Stokes, and 3D Navier-Stokes) assuming isothermal and steady-state conditions. It was demonstrated that the 3D Navier-Stokes model equipped with the detailed washcoat model presents better solution for a broader range of parameters but it suffers from an enormous computational cost which is in order of several days. On the contrary, the plug-flow model which contains no washcoat model has a CPU time of order of few seconds, however, the results may be reliable only for a small range of parameters. Deutschmann et al. (2001) simulated a single rectangular channel with a 3D Navier-Stokes model. In their approach, additional one-dimensional reaction–diffusion equations are applied to consider reactions in the washcoat. Hettel et al. (2013) utilized a CFD modeling approach to investigate the accuracy of the data obtained by suction probe technique in catalytic monoliths. They used a 3D model of several channels of the monolith, with a capillary inside one of them, to calculate the flow field in the channels. It was concluded that the validity of the measured data is highly dependent on the position of the probe in the channel. Braun et al. (2002) used a combination of a 2D model (for flow simulation inside several representative channels) and a transient 3D model (for monolith structure temperature field computation). However, they mentioned the requirement of speed-up for their approach. Maestri et al. (2008) developed a 2D model for simulation of an annular channel with emphasis on the importance of homogeneous reactions at high temperatures, and also, the axial diffusivity. They assumed a fully developed laminar flow inside the channel. Ramanathan et al. (2004) investigated the role of channel cross-section geometry on Sherwood and Nusselt numbers, and also ignition inside the channels. Stutz and Poulikakos (2008) examined the washcoat thickness effect on syngas production by two axisymmetric 2D models. In contrast to the other one, one model is equipped with a washcoat model. They stressed importance of consideration of species diffusion inside the washcoat for accurate simulations. Holmgren and Andersson (1998) investigated the mass transfer in rounded square channel. They compared the experimental data with the results of a 3D CFD model, and concluded that disagreement between the experimental data and the simulation results is mainly because of laminar assumption of the turbulent flow at the channel inlet which has a remarkable effect at higher flow rates. Canu and Vecchi (2002) compared results of 2D and 3D CFD models and concluded that for simulation of rectangular channels using 2D axisymmetric model provides different ignition behavior comparing to the 3D model. Washcoat geometry effect on mass transfer was studied by Hayes et al. (2004) through 2D and 1D modeling approaches. They mentioned that 3D CFD modeling of a channel with complex kinetic mechanisms is a computationally prohibitive method.

Alongside the single-layer catalysts, the technology of dual-layer catalysts has been developed. One of the main improvement focuses for this type of catalysts is to reduce NO_x emission of the lean exhaust in automotive exhaust aftertreatment systems (Pérez-Ramírez et al., 2000; Marnellos et al., 2004). One main idea is to combine a selective catalytic reduction (SCR) layer with a lean NO_x trap (LNT) layer to make a dual-layer catalyst. The SCR layer is deposited on top of the LNT layer (and therefore, in contact with the bulk flow inside the channel). With this configuration, captured NO_x in the LNT layer is reduced to NH_3 during the fuel-rich phase. This NH_3 is stored in the SCR layer and reduces NO_x during the fuel-lean phase. In this manner, instead of a urea injection system, the SCR utilizes the produced NH_3 by the adjacent LNT layer. In addition, since NO_x reduction takes place in both SCR and LNT layers, the dual-layer architecture needs lower volume of the LNT layer compared to single-layer LNT technology, which decreases the amount of platinum group metals (PGM) used within the washcoat. Several modeling studies on dual-layer catalysts are presented in literature with the main focus on 2D and 1D+1D models. Colombo et al. (2012) employed a 1D+1D modeling approach to investigate a dual-layer SCR/PGM catalyst. Because of the computational cost, they just used a reaction–diffusion model for the SCR layer and the assumption that reactions take place on the surface for the PGM layer. In another study (Colombo et al., 2013), the superiority concerning NO_x conversion of the design of the SCR layer on top of the PGM layer has been confirmed by the numerical simulation in comparison with designs in which the PGM layer is on top of the SCR layer and in the mixed layers. Although, under the assumed operating conditions, the results show higher NH_3 yield for the configuration in which SCR layer is on top of PGM layer. Scheuer et al. (2012) developed an axisymmetric 2D model including two washcoat layers, a SCR layer on top of a Pt/Al_2O_3 layer. They utilized a spline mapping method to compute rates and source terms in reaction–diffusion equations for the washcoat layers. They reported that the used mapping approach could speed up the simulation by a factor of 60. In another study (Scheuer et al., 2012), a dual-layer ammonia oxidation catalyst was modeled with 1D+1D and 2D modeling approaches, with main attention to lower the cost of numerical simulations. It was concluded that the results of both approaches are in good agreement, and moreover, the mapping approach of the solution could decrease computational cost by several orders of magnitude. Shakya et al. (2014) utilized a 1D+1D modeling approach to investigate the effect of loading and operating temperature on the performance of a dual-layer SCR/LNT catalyst. They also compared dual-layer architecture with dual-brick design, and concluded that in a wide range of studied operating conditions dual-layer architecture yields higher NO_x conversion.

Developing mathematical models with lower computational expense and with adequate accuracy for performance simulation of monolithic catalysts has been always of interest. The computationally low-cost and real-time models are especially useful for control and optimization of catalytic reactors. Several models have been proposed in the literature for this purpose and some of them, i.e. the effectiveness factor approach, are widely used for computationally low-cost simulations. However, accuracy and application limitation are considered as main challenges for the real-time simulators. Recently, Bissett (2015) has presented a new low-cost modeling method which considers the diffusion limitation in washcoats. In his mathematical approach, by considering the asymptotic solution and applying the boundary conditions, the