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Model-based optimization of catalyst zoning on SCR-coated particulate filters

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HIGHLIGHTS

- Zone-coated filters with SCR function are numerically benchmarked.
- Wall-flow selectivity resulting from catalyst zoning governs filter performance.
- Distributing the coating near the rear of the filter shows improvement potential.

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ABSTRACT

The integration of selective catalytic reduction (SCR) and diesel particulate filter (DPF) into a single device (so called SCRF® or SDPF) can decrease the packaging volume and costs in modern diesel exhaust aftertreatment systems. While SDPF has already found its way into commercial application in passenger cars, for heavy-duty and non-road applications additional aspects need to be considered. Since these systems typically rely on NO_2-assisted regeneration for the removal of soot, the interactions between the SCR and DPF functionalities become important and influence the SDPF performance. Within this frame, advanced coating techniques are investigated in order to limit the detrimental effect of these cross-interactions and achieve optimal system design and control. This study focuses on the evaluation of non-uniform wall impregnation in the axial direction, referred to as catalyst zoning. A validated SDPF mathematical model, able to quantitatively predict the SCR/DPF interactions, is used. The performance of the zone-coated filters is evaluated under both steady-state and transient conditions while the observed phenomena and underlying mechanisms are analytically discussed.

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1. Introduction

The enforcement of stricter emission standards on a global level has enabled the development of efficient aftertreatment technologies for the removal of NO_X and particulate matter from diesel exhaust. These two functions are typically performed by separate devices (Tan et al., 2011). Selective catalytic reduction (SCR) catalysts are established as the most promising and fuel-efficient solution for the abatement of NO_X emissions from heavy-duty diesel engines (Naseri et al., 2011) while "wall-flow" diesel particulate filters (DPF) have been used for more than 15 years to control PM emissions from all diesel engines.

Although both SCR and DPF devices have proven their good efficiency, there is great interest in combining them into a single aftertreatment component, with the motivation being cost- and performance-driven (Tronconi and Nova, 2014; Tang et al., 2013; He et al., 2009; Lee et al., 2008). One major objective is to reduce the packaging volume and the associated costs by decreasing the number of aftertreatment components which has been growing since the introduction of Euro 4/IV and EPA 2007 emission standards. Secondly, considering the efforts for higher thermal efficiency of future engines, a lower average exhaust temperature is expected which can negatively impact cold-start emissions. For this reason, it is necessary to move the aftertreatment components (such as the SCR) closer to the engine in order to retain their performance levels. Based on the above, catalyst manufacturers focused on the development of combined DPF + SCR systems (referred to as SDPF or with the commercial name SCRF®) in which the porous walls of the DPF are impregnated with SCR catalytic material.

Apart from the technological challenge of merging the de-NO_X and PM control functionalities, the interactions between the two is a crucial aspect which needs to be understood in order to enable

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proper component design and optimal system performance. Cross-interactions between the SCR and soot chemistries can result in an SDPF performance which is lower than that of the individual devices, leading to a compromise in the expected gains from an SCR + DPF integration. Indeed, ammonia presence can decrease the passive regeneration rate as a result of the competitive consumption of NO_2 between SCR and soot reactions (Colombo et al., 2011; Schrade et al., 2012; Watling et al., 2012). In addition, the impact on the mass-transfer characteristics by the soot/ash presence can negatively affect de-NO_X performance (Colombo et al., 2011; Cavataio et al., 2009). In an attempt to minimize the impact of these interactions and simultaneously maximize the associated benefits, advanced coating techniques could be employed. Opposed to the conventional uniform wall impregnation, new techniques can selectively concentrate the washcoat in the axial (zoning) or the wall (layering) direction. Such methods, well established for other applications such as the TWC or LNT, can be employed in order to isolate the SCR and soot chemistries from each other. In this respect, simulation tools can effectively support the development process by evaluating new coating designs and at the same time reducing the required time and costs (Tsinoglou et al., 2007; Koltsakis et al., 2010).

The present work is dedicated to the evaluation of axial catalyst zoning and its effect on the SDPF performance. Three different exemplary coating configurations were selected, as schematically illustrated in Fig. 1. Washcoat is always assumed to be located entirely inside the wall, uniformly in the radial direction, with the investigated coating variations being limited to the axial direction. To ensure direct comparability, all configurations are characterized by the same washcoat amount of 60 g/l (expressed per liter of total filter volume). The uniformly-coated SDPF serves as the reference case against which the two zoned configurations, namely ‘Zoned 2:1’ and ‘Zoned 1:2’, are benchmarked. In the first case more washcoat is placed at the front while in the second case more is placed at the rear. The higher washcoat zone has a loading of 80 g/l and the lower one of 40 g/l. Since both zones have the same length, global washcoat loading is always 60 g/l. The impact of this local washcoat loading variation on SCR reactivity is directly taken into account in the model as it is associated with a linear variation of the ammonia storage capacity and of the respective reaction rates (see Table 1).

All the above-described coating configurations will be evaluated by steady-state simulations in terms of pressure drop, passive regeneration in presence of NH_3 as well as NO_X conversion efficiency at both clean and soot loaded state. In a final step, transient simulations with consecutive driving cycles will be performed in order to evaluate the possible zoning advantages under “real-world” driving conditions.

2. Mathematical model

2.1. Overview

The present model-based study was performed using the commercial axisuite® simulation platform. A thoroughly calibrated and validated SDPF model was employed, analytically presented in a previous publication (Tronconi et al., 2015). The respective 5.66 × 10 in. SCR-coated filter was Cu-zeolite based. Intrinsic SCR reaction kinetics were extracted from experiments over powdered catalysts while soot reaction kinetics were obtained based on engine-bench tests. Assuming a simple superimposition, the two chemistries were then combined and transferred into a physico-chemical model of the flow, transport and reaction processes in wall-flow monolith channels. The detrimental effect of NH_3-SCR reactions on the passive regeneration rate as well as the negative impact of soot on NO_X conversion efficiency were experimentally observed and successfully predicted by the model. The latter was possible due to the proper consideration of intra-layer species diffusion through the soot layer and substrate wall. Further details on the above modeling methodology and the respective simulation results are provided elsewhere (Tronconi et al., 2015).

Table 1
SCR reaction scheme.

Type	Reaction
NH_3 storage/ release	$NH_3 \leftrightarrow NH_3^*$ $r_{ads} = k_{ads} C_{\mathrm{NH3}} (1 - \theta_{\mathrm{NH3}} - \theta_{nit})$ $r_{des} = exp\left[k_{des}^0 - E_{des}\left(\frac{1000}{T}(1 - \alpha\theta_{\mathrm{NH3}}) - \frac{1000}{473}\right)\right]\theta_{\mathrm{NH3}}$
NH_3 oxidation to N_2	$NH_3^* + 3/4O_2 \rightarrow 1/2N_2 + 3/2H_2O$ $r_{ox} = exp\left(k_{ox}^0 - E_{ox}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)\theta_{\mathrm{NH3}}(1 + \gamma \cdot C_{\mathrm{NO}})$
NH_3 oxidation to NO	$NH_3^* + 5/4O_2 \rightarrow NO + 3/2H_2O$ $r_{oxb} = exp\left(k_{oxb}^0 - E_{oxb}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)\theta_{\mathrm{NH3}}$
NO oxidation	$NO + 1/2O_2 \leftrightarrow NO_2$ $r_{\mathrm{NO}_{ox}} = exp\left(k_{\mathrm{NO}_{ox}}^0 - E_{\mathrm{NO}_{ox}}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)\frac{(C_{\mathrm{NO}}\sqrt{P_{\mathrm{O2}}} - (C_{\mathrm{NO2}}/K_{\mathrm{NO2}}^{eq}))}{1 + K_{\mathrm{NO2}} C_{\mathrm{NO2}}}$
Standard-SCR	$NH_3^* + NO + 1/4O_2 \rightarrow N_2 + 3/2H_2O$ $r_{\mathrm{NO}} = exp\left(k_{std}^0 - E_{\mathrm{NO}}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)C_{\mathrm{NO}}\theta_{\mathrm{NH3}}(1 - \sigma_{\mathrm{NH3}})$
Fast-SCR	$NH_3^* + 1/2NO + 1/2NO_2 \rightarrow N_2 + 3/2H_2O$ $r_{Fast} = exp\left(k_{Fast}^0 - E_{Fast}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)C_{\mathrm{NO}} C_{\mathrm{NO2}} \theta_{\mathrm{NH3}}$
NO_2-SCR	$NH_3^* + 3/4NO_2 \rightarrow 7/8N_2 + 3/2H_2O$ $r_{\mathrm{NO2}} = exp\left(k_{\mathrm{NO2}}^0 - E_{\mathrm{NO2}}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)C_{\mathrm{NO2}}\theta_{\mathrm{NH3}}$
N_2O formation	$NH_3^* + NO_2 \rightarrow 1/2N_2O + 1/2N_2 + 3/2H_2O$ $r_{\mathrm{N2O}} = exp\left(k_{\mathrm{N2O}}^0 - E_{\mathrm{N2O}}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)C_{\mathrm{NO2}}\theta_{\mathrm{NH3}}$
Ammonium nitrate formation	$NH_3^* + NO_2 \rightarrow 1/2NH_4NO_3^* + 1/2N_2 + 1/2H_2O$ $r_{Amm} = \frac{exp\left(k_{Amm}^0 - E_{Amm}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)\theta_{\mathrm{NH3}} C_{\mathrm{NO2}}^2}{1 + K_{Amm}\theta_{nit}}$
Ammonium nitrate sublimation	$NH_4NO_3^* \rightarrow (NH_3) + (HNO_3) \rightarrow NH_4NO_3^*(s)$ $r_{Nit} = exp\left(k_{Nit}^0 - E_{Nit}\left(\frac{1000}{T} - \frac{1000}{473}\right)\right)\theta_{Nit}$

2.2. Reaction scheme

Regarding SCR reactivity, a well-accepted kinetic mechanism, describing the SCR chemistry over Cu-zeolites, was used (Tronconi et al., 2015; Colombo et al., 2010). The respective chemical reactions and reaction rate expressions are listed in Table 1.

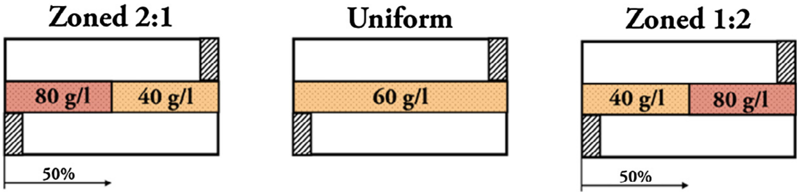

Fig. 1. SDPF coating configurations considered in the model-based analysis.

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