SIMULATION OF NO\textsubscript{x} AND SOOT ABATEMENT VIA SCRoF WITH Cu-CHA AND Fe-ZSM5 CATALYSTS

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Abstract

The present work tackles the modeling of SCRoF technology using recently developed kinetic models of Cu-Chabazite and Fe-ZSM-5 catalysts. We use a 1-D model of flow, diffusion and reaction through the filter cake and filter wall depth to assess the performance of SCRoF technology under different conditions. The ability of the SCRoF configuration to meet the conventional SCR performances, as well as to ensure proper soot oxidation activity, were evaluated at different operating conditions of inlet NO/NO\textsubscript{x} ratios and NH\textsubscript{3} content in the feed gas. Combinations of Cu-Cha and Fe-ZSM5 catalysts were also devised in order to optimize the synergy between NO\textsubscript{x} reduction and soot passive regeneration.

Keywords

Selective Catalytic Reduction on Filter (SCRoF); Soot Oxidation; NO\textsubscript{x} Reduction; Numerical Modeling.

Introduction

Advanced diesel combustion engines have higher net efficiencies as well as lower NO\textsubscript{x} and particulate matter (PM) emissions than current ones, together with lower exhaust gas temperatures. Furthermore, space limitations in light-duty diesel engine vehicles force car manufacturers to combine different functionalities into single converters.

A solution is the embodiment of the NO\textsubscript{x} selective catalytic reduction (SCR) functionality in the already present diesel particulate filter (DPF), in the so-called SCR-on-Filter (SCRoF), and is achieved by depositing the SCR catalyst in the inner porosities of the filter walls, or on the top of them (Fig. 1). NO\textsubscript{x} are reduced through the addition of NH\textsubscript{3}, whereas soot accumulates on the filter walls, as in a conventional DPF, and is regenerated either by passive regeneration (through the effect of NO\textsubscript{2}-assisted soot oxidation) or active regeneration (through the increase of the upstream gas temperature in the Diesel Oxidation Catalyst –DOC– up to around 600° C to promote soot oxidation). The SCRoF can profit from the concomitant SCR reaction for NO\textsubscript{x} abatement and the passive soot regeneration occurring in the filter: when gas temperatures and NO\textsubscript{2} levels are adequate, the latter exploits NO\textsubscript{2}-mediated soot oxidation, which reduces the NO\textsubscript{2} content in the gas flow by converting it to NO; on the other hand, the SCR reaction completes the NO+NO\textsubscript{2} reduction to N\textsubscript{2}. The understanding of the interplay between these reactions, through the overall system modeling, is the objective of this work.

Figure 1. Sketch of the computational domain.

Methodology

The modeled system refers to the SCRoF wall separating two neighboring channels, which entails a homogeneous concentration of gaseous species in the inlet
channel of the filter, and a rather flat profile of through-wall velocity along the axial direction. These assumptions apply in the presence of a soot cake, which flattens the velocity profile along the channel axis due to its low permeability, and during a filter passive regeneration, which proceeds with equal kinetics along the channel given the absence of strong temperature gradients.

Under these conditions, the relevant gradients occur all along the depth of the filter wall (Fig. 1), which becomes the coordinate of interest for our purpose. The model entails a 1D discretization of the soot cake and filter wall depth, where continuity and momentum conservation are imposed, as well as the transport of enthalpy (with a common gas and solid temperature, which was verified given the high heat exchange coefficient inside the porous media), gas species (NO, NO$_2$, NH$_3$, N$_2$O, NH$_4$NO$_3$, H$_2$O, O$_2$, N$_2$) and PM (in the gas phase and as deposited solid, both inside the wall and on the top of it as a cake layer). The physical parameters of the modeled SCRoF are gathered in Table 1. The chemical parameters for the computation of the SCR kinetics were retrieved from Metkar (2013), and are referred to Cu-Chabazite and Fe-ZSM5 SCR catalysts; the O$_2$ and NO$_2$-mediated soot oxidation kinetics are from Zouaoui (2014).

**Table 1. System major parameters**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>Through-wall velocity</td>
<td>1.7 cm/s</td>
</tr>
<tr>
<td>Wall/soot cake thicknesses</td>
<td>367/70 μm</td>
</tr>
<tr>
<td>Wall porosity (uncoated/coated)</td>
<td>0.68/0.53</td>
</tr>
<tr>
<td>Catalyst site density (Cu-Cha/Fe-ZSM5)</td>
<td>380/190 mol/m$^3$</td>
</tr>
</tbody>
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**Results and Discussion**

The results shown in Fig. 2 are referred to the SCRoF de-NOx activity with Cu-Cha catalyst, in the absence and presence of soot. Firstly, in (a) one can see that, at the investigated operating conditions and in the absence of soot, the highest removal of NO$_x$ can be achieved around 300°C. Although the inlet is equimolar in NO and NO$_2$, their reduction is not equivalent, given the presence of the standard and NO$_2$ SCR reactions (the former faster than the latter), in parallel to the fast SCR. The increase in NO$_2$ reduction at higher temperatures is due to NO$_2$ to NO conversion. Plot (b) compares the SCR activity in the absence and presence of soot (the latter truly replicating a SCRoF). One can see that the presence of soot negligibly affects the NO$_x$ conversion efficiency and the N$_2$ yield (calculated as outlet N$_2$ over NO+NO$_2$+NH$_3$ at the inlet, therefore possibly being higher than NO$_2$ conversion), given the slow dynamics of passive regeneration. This conclusion is true for 1:1 NO and NO$_2$ concentrations in the feed, coupled to an NH$_3$ to NO$_x$ ratio of 1, but could lead to different conclusions i.e. for NO$_2$ to NO$_x$ ratios of 0.35 or 0.65 (not shown here for the sake of brevity). If one focuses on soot passive oxidation, one can see that (over a time of 3600 s) the reduction in cake thickness in the absence of any SCR activity (as in a conventional DPF) significantly differs in its presence (in the SCRoF), leading to a nearly half consumption of soot over this timeframe. The NO$_2$-mediated soot passive regeneration is largely reduced by the lower NO$_2$ in the gas mixture crossing the cake, and this is due to back-diffusion phenomena which are relevant (axial Pe over the wall+cake thickness is around 1). The inclusion also of the kinetics of Fe-ZSM5 catalysts (Metkar, 2013), not reported here, was performed to expand the operating windows of the SCRoF, to achieve greater passive regeneration at higher temperatures, still being active for SCR (otherwise declining for Cu-Cha).

Finally, the combination of Cu-Cha and Fe-ZSM5 was also investigated, reaching an optimal activity both at low temperature and high NO content (for which Cu-Cha excels while Fe-ZSM5 offers poor performances) and high temperature (at which Fe-ZSM5 is more active), still limiting the loss of soot passive oxidation activity.

![Figure 2](image)

**Figure 2.** (a) NO, NO$_2$ and total NO$_x$ conversion; (b) NO$_x$ total conversion, N$_2$ yield and soot conversion; Inlet: O$_2$ 0.05%, NH$_3$ 500 ppm, NO 250 ppm, NO$_2$ 250 ppm.

**Conclusions**

The simulation of a SCRoF was performed, in order to disclose the best conditions for the simultaneous NO$_x$ and soot abatement in a single converter, with Cu-Cha and Fe-ZSM5 catalysts. Hybrid catalytic formulations were also simulated to exploit their different operating ranges.

**References**


Zouaoui, N., Labaki, M., Jeguirim, M., (2014). Diesel soot oxidation by NO$_2$, O$_2$ and H$_2$O under engine exhaust conditions: kinetics data related to the reaction mechanism *C.R. Chimie, 17*, 672-680.