Model-based analysis of SCR-coated filters performance aspects

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From SCR to SCR\textsubscript{onF}

**SCR**

**SCR\textsubscript{onF}**

Interaction of SCR with soot and ash

SCR + DPF \neq SCR\textsubscript{onF}

NO\textsubscript{x} conversion is affected by accumulated soot

<table>
<thead>
<tr>
<th>SCR reactions</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{NH}_3 + \text{NO} + \frac{1}{4}\text{O}_2 \rightarrow \text{N}_2 + \frac{3}{2}\text{H}_2\text{O} )</td>
</tr>
<tr>
<td>( 2\text{NH}_3 + \text{NO} + \text{NO}_2 \rightarrow 2\text{N}_2 + 3\text{H}_2\text{O} )</td>
</tr>
<tr>
<td>( \text{NH}_3 + \frac{3}{4}\text{NO}_2 \rightarrow \frac{7}{8}\text{N}_2 + \frac{3}{2}\text{H}_2\text{O} )</td>
</tr>
</tbody>
</table>

<table>
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<tr>
<th>Soot reactions</th>
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<tbody>
<tr>
<td>( \text{C} + \text{O}_2 \rightarrow \text{CO}_2 )</td>
</tr>
<tr>
<td>( \text{C} + \frac{1}{2}\text{O}_2 \rightarrow \text{CO} )</td>
</tr>
<tr>
<td>( \text{C} + 2\text{NO}_2 \rightarrow \text{CO}_2 + 2\text{NO} )</td>
</tr>
<tr>
<td>( \text{C} + \text{NO}_2 \rightarrow \text{CO} + \text{NO} )</td>
</tr>
</tbody>
</table>

Soot oxidation affected by \( \text{NH}_3 \)

\[ \text{Burned soot mass [g]} \]

\[ \text{w/o NH}_3 \quad \text{w NH}_3 \quad \text{w/o NH}_3 \quad \text{w NH}_3 \]

\[ \text{350°C} \quad \text{450°C} \]
Target: study the impact of coating variations on:
- NOx conversion at clean state
- Soot and ash impact on NOx conversion
- NH$_3$-SCR impact on soot oxidation rate

Impact of catalyst zoning and layering?

Only standard SCR is considered with alpha = 1
Same SCR catalyst amount
**Cu-Zeolite reaction scheme**

<table>
<thead>
<tr>
<th>Type</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₃ adsorption</td>
<td>( \text{NH}_3 + S1 \rightarrow \text{S1NH}_3 )</td>
</tr>
<tr>
<td></td>
<td>( \text{NH}_3 + S2 \rightarrow \text{S2NH}_3 )</td>
</tr>
<tr>
<td>NH₃ desorption</td>
<td>( \text{S1NH}_3 \rightarrow \text{NH}_3 + S1 )</td>
</tr>
<tr>
<td></td>
<td>( \text{S2NH}_3 \rightarrow \text{NH}_3 + S2 )</td>
</tr>
<tr>
<td>Standard SCR</td>
<td>( 4\text{NH}_3 + 4\text{NO} + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O} )</td>
</tr>
<tr>
<td>Fast SCR</td>
<td>( 2\text{NH}_3 + \text{NO} + \text{NO}_2 \rightarrow 2\text{N}_2 + 3\text{H}_2\text{O} )</td>
</tr>
<tr>
<td>NO₂ SCR</td>
<td>( 8\text{NH}_3 + 6\text{NO}_2 \rightarrow 7\text{N}_2 + 12\text{H}_2\text{O} )</td>
</tr>
<tr>
<td></td>
<td>( \text{NH}_3 + \text{NO}_2 \rightarrow \frac{1}{2} \text{N}_2 + \frac{1}{2} \text{N}_2\text{O} + 3/2 \text{H}_2\text{O} )</td>
</tr>
<tr>
<td>NO to NO₂ oxidation</td>
<td>( 2\text{NO} + \text{O}_2 \rightarrow 2\text{NO}_2 )</td>
</tr>
<tr>
<td>NO₂ dissociation</td>
<td>( 2\text{NO}_2 \rightarrow 2\text{NO} + \text{O}_2 )</td>
</tr>
<tr>
<td>NH₃ oxidation towards N₂</td>
<td>( 4\text{NH}_3 + 3\text{O}_2 \rightarrow 2\text{N}_2 + 6\text{H}_2\text{O} )</td>
</tr>
<tr>
<td>NH₃ oxidation towards NO</td>
<td>( 4\text{NH}_3 + 5\text{O}_2 \rightarrow 4\text{NO} + 6\text{H}_2\text{O} )</td>
</tr>
<tr>
<td>NH₃ oxidation towards N₂O</td>
<td>( \text{NH}_3 + \text{O}_2 \rightarrow \frac{1}{2} \text{N}_2\text{O} + 3/2 \text{H}_2\text{O} )</td>
</tr>
</tbody>
</table>

Wall-flow reactor model

**Key features**

Transport-reaction coupling enabling modeling of soot/SCR interactions.

Intra-layer dimension for filtration and reaction modeling.
Internal diffusion effect on NOx conversion

Advection-Diffusion-Reaction equation

\[ v_w \frac{\partial y_j}{\partial w} - D_{w,j} \frac{\partial}{\partial w} \left( f_w \frac{\partial y_j}{\partial w} \right) = \frac{f_w}{c_m} \sum_k c_{j,k} R_k \]

\[ \frac{1}{D_{w,j}} = \tau \frac{1}{D_{mol,j}} + \frac{1}{D_{knud,j}} \epsilon_{pore} \]

NOx = 500 ppm, NOy/NOx = 0, NH3/NOx = 1, T = 250°C, GHSV = 50,000 h⁻¹
Soot loading = 0 g/l
Internal diffusion effect on NOx conversion

The axial NOx concentration profile is affected by wall diffusion.

Advection-Diffusion-Reaction equation

$$v_w \frac{\partial y_j}{\partial w} - D_{w,j} \frac{\partial}{\partial w} \left( f_w \frac{\partial y_j}{\partial w} \right) = \frac{f_w}{c_m} \sum_k c_{j,k} R_k$$

Wall tortuosity

$$\frac{1}{D_{w,j}} = \tau \left( \frac{1}{D_{mol,j}} + \frac{1}{D_{knud,j}} \right)$$

NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}
Soot loading = 0 g/l
Internal diffusion effect on NOx conversion

Advection-Diffusion-Reaction equation

$$v_w \frac{\partial y_j}{\partial w} - D_{w,j} \frac{\partial}{\partial w} \left( f_w \frac{\partial y_j}{\partial w} \right) = f_w \frac{c_w}{c_m} \sum_k c_{j,k} R_k$$

Wall tortuosity

$$\frac{1}{D_{w,j}} = \frac{\tau}{\varepsilon_{pore}} \left( \frac{1}{D_{mol,j}} + \frac{1}{D_{knud,j}} \right)$$

- Increased diffusion resistance decreases NOx conversion

NOx=500ppm, NO_3/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

Soot loading = 0 g/l

Graph showing NOx conversion percentage with increasing diffusion resistance.

Outlet channel

Graph showing NOx concentration with increasing length.
Effect of catalyst 'layering'

NOx=500ppm, NO2/NOx=0, NH3/NOx=1, T=250°C, GHSV=50,000 h⁻¹
Soot loading = 0 g/l

The inert part of the wall acts as a ‘diffusion-barrier’
NOx conversion decreases
Effect of catalyst ‘zoning’

**250°C**

\[ \text{NOx}=500\text{ppm, NO}_2/\text{NOx}=0, \text{NH}_3/\text{NOx}=1, T=250^\circ\text{C}, \text{GHSV}=50,000 \ \text{h}^{-1} \]

- **inlet channel**
  - Uniform
  - Zoned 2:1
  - Zoned 1:2

- **outlet channel**

**Higher loading = lower permeability = less wall flow**
Soot has a **negative effect** on standard SCR acting as diffusion barrier.
Ash effect on NOx conversion

NOx=500ppm, NO2/NOx=0, NH3/NOx=1, T=250°C, GHSV=50,000 h⁻¹

<table>
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<tr>
<th>Ash property</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>Packing density</td>
<td>300 kg/m³</td>
</tr>
<tr>
<td>Permeability</td>
<td>5e-14 m²</td>
</tr>
<tr>
<td>Pore diameter</td>
<td>100 nm</td>
</tr>
<tr>
<td>Pore volume fraction</td>
<td>90 %</td>
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</tbody>
</table>
Ash effect on NOx conversion

**Ash property** | **Value**
---|---
Packing density | 300 kg/m$^3$
Permeability | 5e-14 m$^2$
Pore diameter | 100 nm
Pore volume fraction | 90 %

NOx=500ppm, NO$_2$/NO$_x$=0, NH$_3$/NO$_x$=1, T=250°C, GHSV=50,000 h$^{-1}$
Ash effect on NOx conversion
Impact of catalyst zoning

NOx=500 ppm, \( \frac{\text{NO}_2}{\text{NOx}} = 0 \), \( \frac{\text{NH}_3}{\text{NOx}} = 1 \), T=250°C, GHSV=50,000 h\(^{-1}\)

**Graph:**

- **Uniform:**
  - Clean: 87.2 ± 0.2%
  - Layer ash 40 g/l: 79.9 ± 0.9%

- **Zoned 2:1:**
  - Clean: 84.2 ± 0.2%
  - Layer ash 40 g/l: 70.9 ± 0.9%

- **Zoned 1:2:**
  - Clean: 86.5 ± 0.2%
  - Layer ash 40 g/l: 82.1 ± 0.9%
  - Plug ash 40 g/l: 83.1 ± 0.9%

**Legend:**
- Clean
- Layer ash 40 g/l
- Plug ash 40 g/l

**Note:**
+2.3% increase in NOx conversion with zoned 1:2 configuration.
Effect of NH$_3$/SCR on soot oxidation

NO$_x$=600ppm, NO$_2$/NO$_x$=0.3, T=350°C, GHSV=90,000 h$^{-1}$

Average burn rate

23 g/h
SCR effect on soot oxidation @ 350°C

NOx=600ppm, NO₂/NOx=0.3, T=350°C, GHSV=90,000 h⁻¹

-48% decrease in soot concentration

Flow direction:
- Soot concentration with and without NH₃
- Reaction rate comparison: C+O₂ vs. C+NO₂

Soot concentration over time:
- w/o NH₃: 4 g/l to 2 g/l in 6000 s
- w NH₃, α=1: 3 g/l to 1.5 g/l in 6000 s

Graphs showing:
- Soot concentration decrease
- Reaction rate comparison

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SCR effect on soot oxidation with ash @ 350°C

NOx=600ppm, NO₂/NOx=0.3, T=350°C, GHSV=90,000 h⁻¹

Flow

Soot [g/l]

- w/o NH₃
- w NH₃, α=1
- w NH₃, α=1, ash layer 10g/l

<table>
<thead>
<tr>
<th>Flow rate [g/h]</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>12</td>
<td>-48%</td>
</tr>
<tr>
<td>19</td>
<td>+58%</td>
</tr>
<tr>
<td>23</td>
<td></td>
</tr>
</tbody>
</table>

Reactor temperatures:
- NOx=600ppm, NO₂/NOx=0.3, T=350°C, GHSV=90,000 h⁻¹

Reaction rate [mol/s]

- w/o NH₃
- w NH₃
- ash layer 10g/l

NO₂ [ppm]
SCR and ash effect on soot balance point

\[ T = 350^\circ C, \text{ Flow}=1000\text{kg/h}, \frac{\text{NO}_2}{\text{NO}_x}=0.3, \frac{\text{NO}_x}{\text{soot}}=35 \text{ kg/kg} \]
Conclusions

- Internal wall diffusion processes affect NOx conversion.
- Soot and ash layers act as diffusion barriers, reducing catalyst efficiency.
- Catalyst zoning shows interesting behavior in case of accumulated plug-ash.
- NO$_2$-driven soot oxidation at low temperatures is subject to competition with NO$_2$-SCR reactivity.
- Diffusion barriers (e.g. ash layer) have a positive impact on passive soot oxidation under SCR-reacting conditions.
Thank you very much for your attention!