Model-based analysis of SCR-coated filters performance aspects

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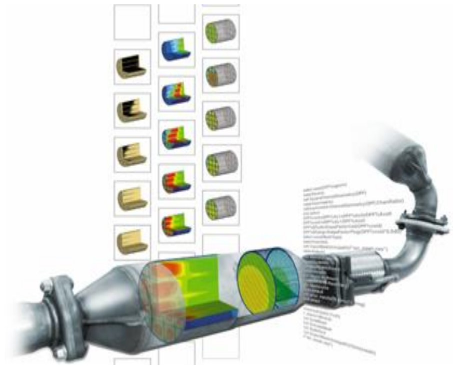
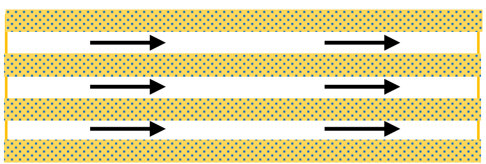


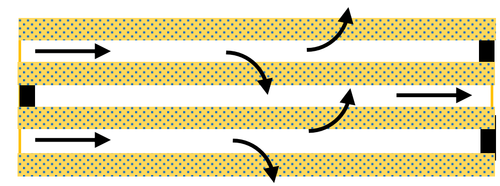
SCR + DPF ≠ SCRonF

SCR



Interaction of SCR with soot and ash

SCRonF



NOx conversion is affected by accumulated soot

SCR reactions

$NH_3 + NO + 1/4O_2 \longrightarrow N_2 + 3/2H_2O$

$2NH_3 + NO + \mathbf{NO_2} \longrightarrow 2N_2 + 3H_2O$

$NH_3 + 3/4\mathbf{NO_2} \longrightarrow 7/8N_2 + 3/2H_2O$

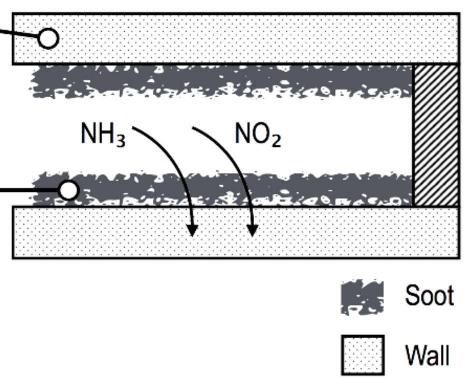
Soot reactions

$C + O_2 \longrightarrow CO_2$

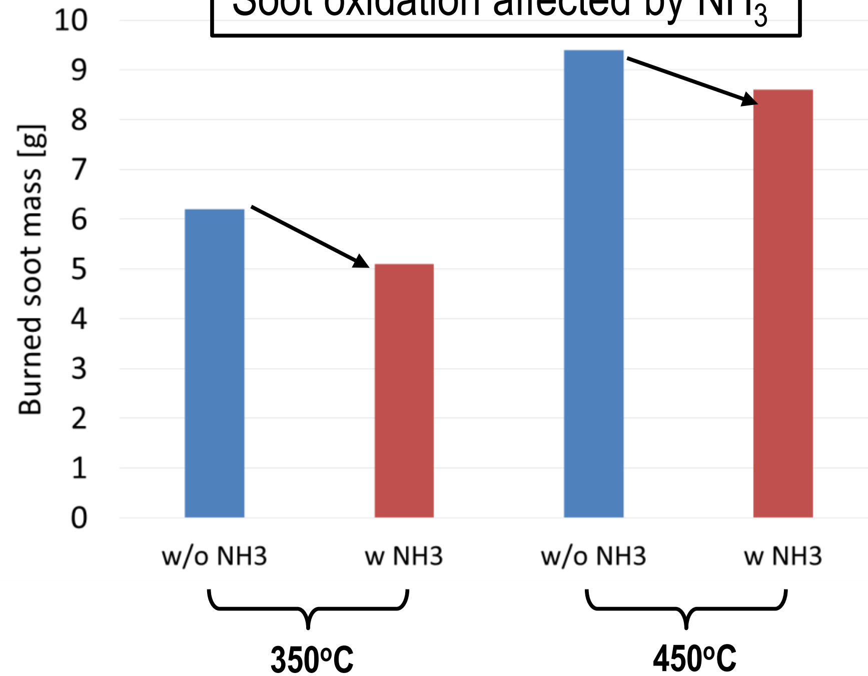
$C + 1/2O_2 \longrightarrow CO$

$C + 2\mathbf{NO_2} \longrightarrow CO_2 + 2NO$

$C + \mathbf{NO_2} \longrightarrow CO + NO$



Soot oxidation affected by NH_3

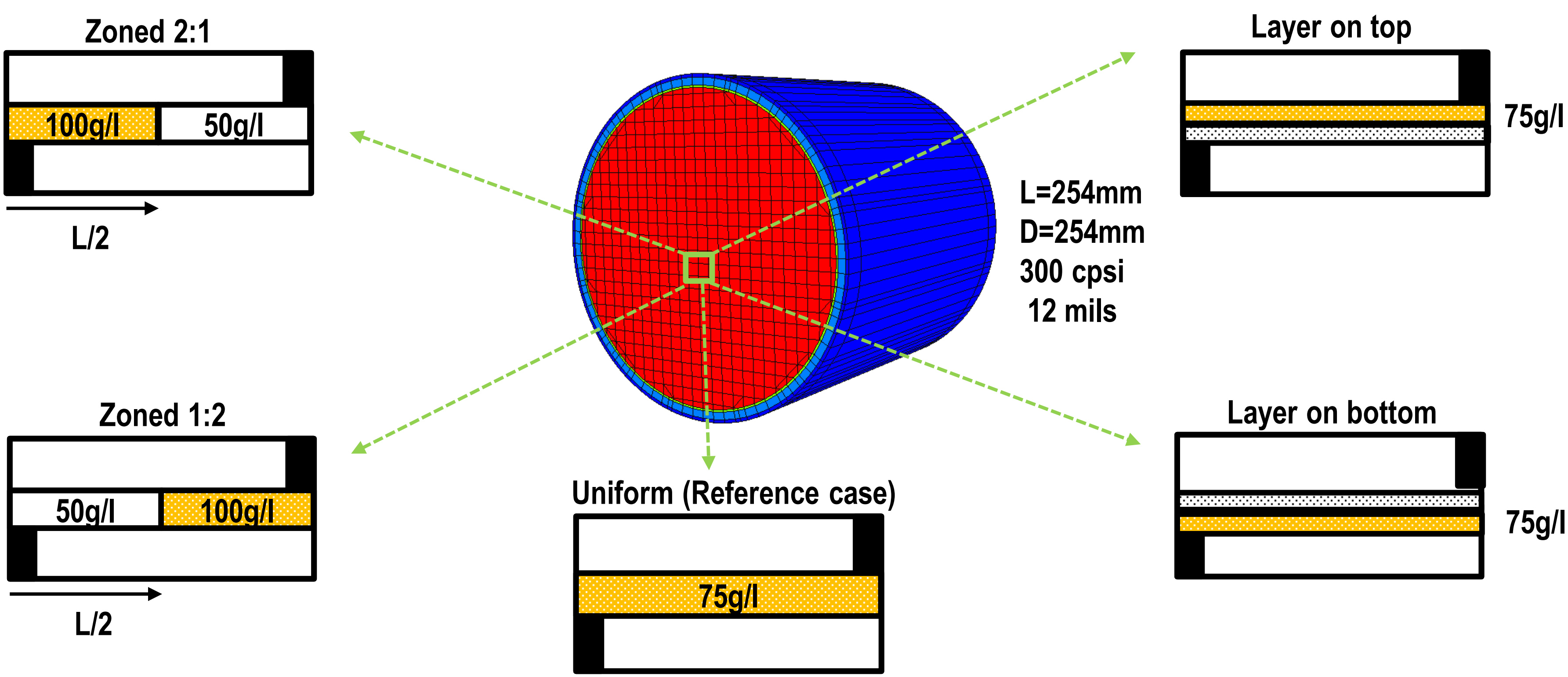


Impact of catalyst zoning and layering?

- 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

Only standard SCR is considered with alpha =1

Same SCR catalyst amount



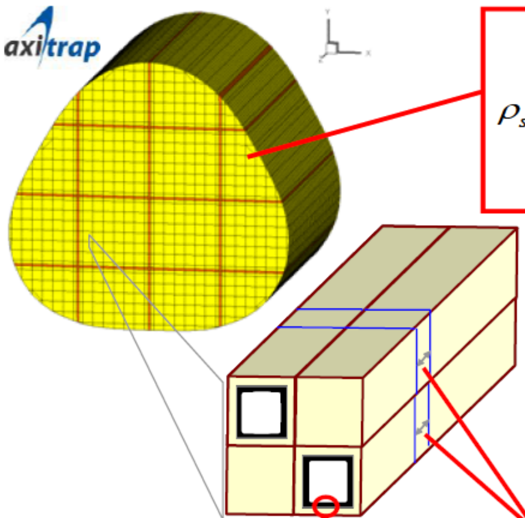
SCR reaction model

Cu-Zeolite reaction scheme

Type	Reaction
NH_3 adsorption	$NH_3 + S1 \rightarrow S1NH_3$ $NH_3 + S2 \rightarrow S2NH_3$
NH_3 desorption	$S1NH_3 \rightarrow NH_3 + S1$ $S2NH_3 \rightarrow NH_3 + S2$
Standard SCR	$4NH_3 + 4NO + O_2 \rightarrow 4N_2 + 6H_2O$
Fast SCR	$2NH_3 + NO + NO_2 \rightarrow 2N_2 + 3H_2O$
NO_2 SCR	$8NH_3 + 6NO_2 \rightarrow 7N_2 + 12H_2O$ $NH_3 + NO_2 \rightarrow ½\, N_2 + ½\, N_2O + 3/2\, H_2O$
NO to NO_2 oxidation	$2NO + O_2 \rightarrow 2NO_2$
NO_2 dissociation	$2NO_2 \rightarrow 2NO + O_2$
NH_3 oxidation towards N_2	$4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O$
NH_3 oxidation towards NO	4NH3 + 5O2 $\rightarrow$ 4NO + 6H2O
NH_3 oxidation towards N_2O	$NH_3 + O_2 \rightarrow ½\, N_2O + 3/2\, H_2O$

Reference: G. Koltsakis et al., Model-based optimization of advanced SCR substrates, International Journal of Automotive Engineering, Vol. 6 No. 2 (2015)

Wall-flow reactor model



Filter scale: 3-d solid energy balance

$$\rho_s \cdot C_{p,s}\frac{\partial T_s}{\partial t} = k_{s,x}\frac{\partial^2 T_s}{\partial x^2} + k_{s,y}\frac{\partial^2 T_s}{\partial y^2} + k_{s,z}\frac{\partial^2 T_s}{\partial z^2} + S$$

$$S = H_{conv} + H_{wall} + H_{react} + H_{rad}$$

Channel scale: gas balances
Mass/momentum/energy/species

$$\frac{\partial}{\partial z}\left(d_i^{\,2}\rho_i v_i\right) = (-1)^i 4d\rho_w v_w$$

$$\frac{\partial p_i}{\partial z} + \frac{\partial}{\partial z}\left(\rho_i v_i^2\right) = -\alpha_1 \mu v_i / d_i^{\,2}$$

$$C_{p,g}\rho_1 v_1\big|_z \frac{\partial T_1}{\partial z} = h_1 \frac{4}{d_1}(T_s - T_1)$$

$$C_{p,g}\rho_2 v_2\big|_z \frac{\partial T_2}{\partial z} = (h_2 + C_{p,g}\rho_w v_w)\frac{4}{d}(T_s - T_2)$$

$$\frac{\partial}{\partial z}\left(v_1 y_{1,j}\right) = -\frac{1}{df_{-w}^{\;2}} v_w y_{1,j} + \frac{1}{df_{-w}} k_{1,j}\left(y_{1s,j} - y_{1,j}\right)$$

$$\frac{\partial}{\partial z}\left(v_2 y_{2,j}\right) = \frac{1}{df_{w_s}^{\;2}} v_w y_{2s,j} + \frac{1}{df_{w_s}} k_{2,j}\left(y_{2s,j} - y_{2,j}\right)$$

Wall/soot scale balances
Momentum/soot/ species

$$\frac{dp}{dx} = \frac{\mu v(x)}{k_p}$$

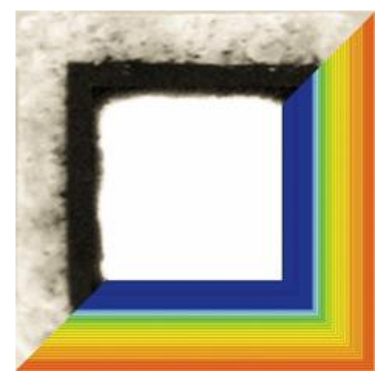
$$\frac{d\hat{m}_p}{dt} = -\hat{m}_p \sum R'_k + s_F \rho_w v_w \mu_p$$

$$v_w \frac{\partial y_j}{\partial x} - D_j \frac{\partial}{\partial x}\left(f_x \frac{\partial y_j}{\partial x}\right) = \frac{f_x}{c_m}\sum_k c_{j,k} R_k$$

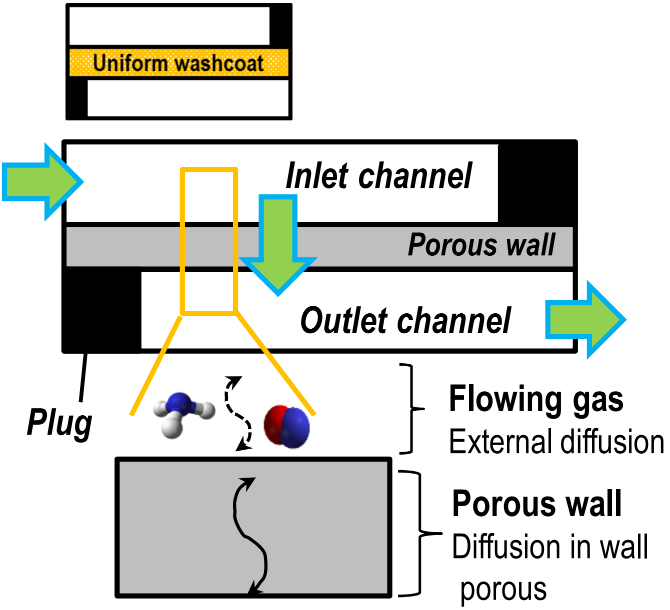
Key features

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

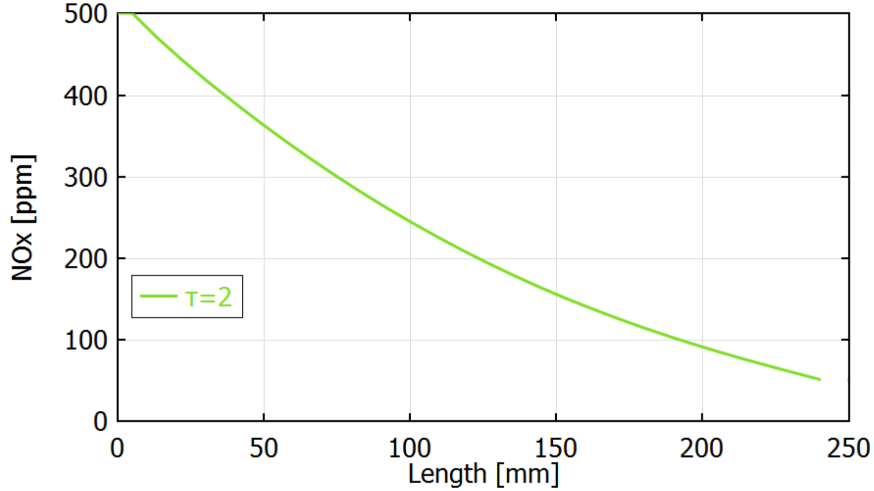
$$v_w \frac{\partial y_j}{\partial w} - D_{w,j} \frac{\partial}{\partial w}\left(f_w \frac{\partial y_j}{\partial w} \right) = \frac{f_w}{c_m} \sum_k c_{j,k} R_k$$

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

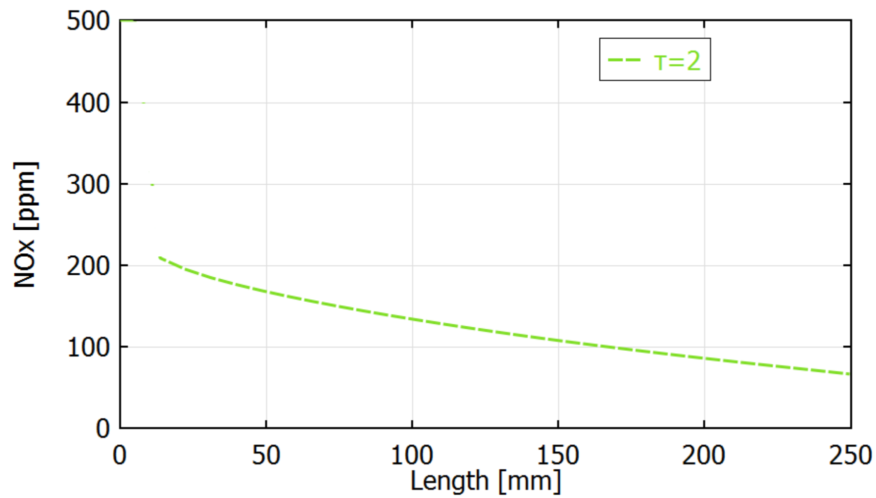
NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

Soot loading = 0 g/l

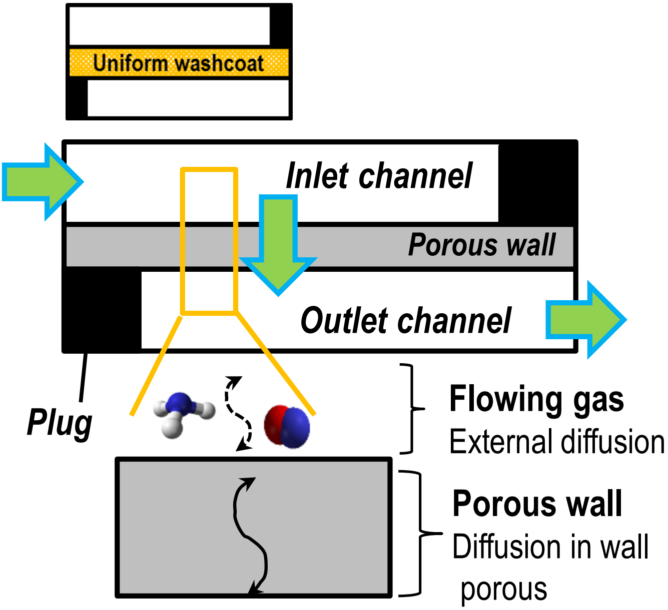
inlet channel



outlet channel



Internal diffusion effect on NOx conversion



NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

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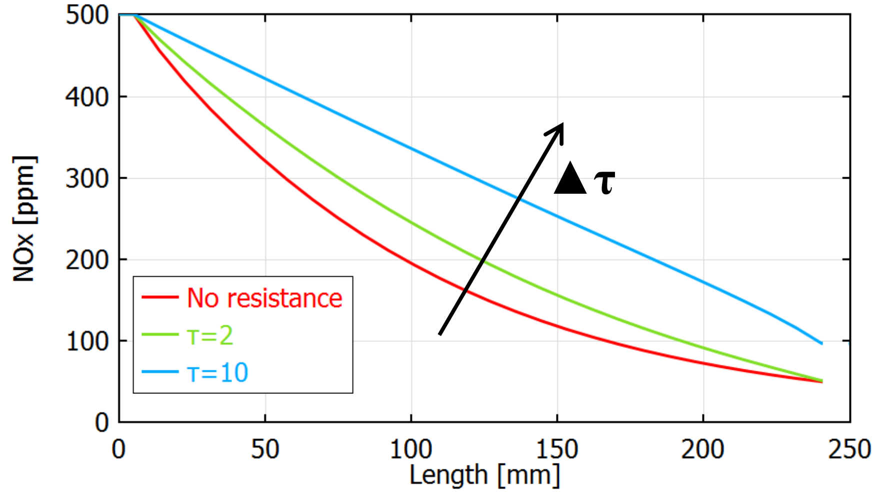
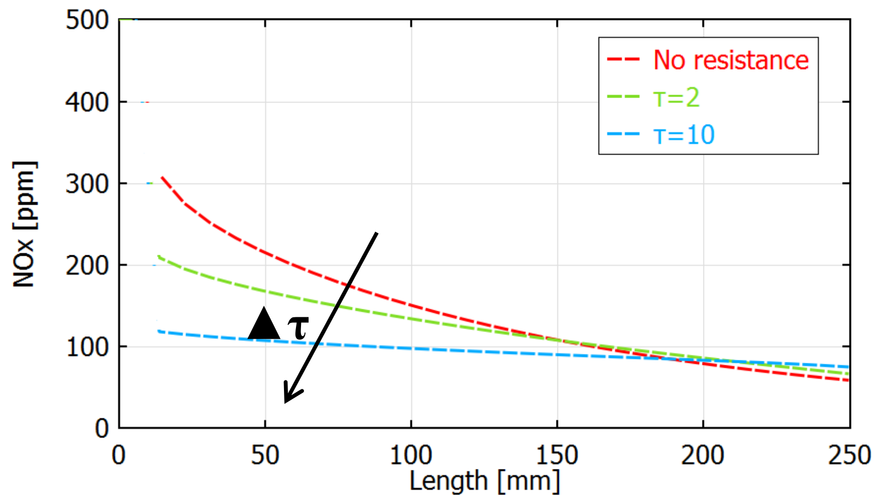


Advection-Diffusion-Reaction equation

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

Wall tortuosity

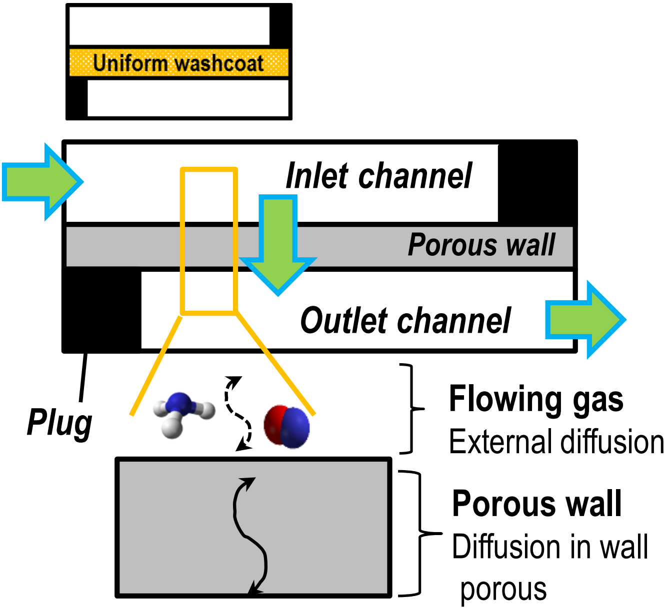
$$\frac{1}{D_{w,j}} = \frac{\tau}{\varepsilon_{pore}} \left(\frac{1}{D_{mol,j}} + \frac{1}{D_{knud,j}} \right)$$



– The axial NOx concentration profile is affected by wall diffusion.

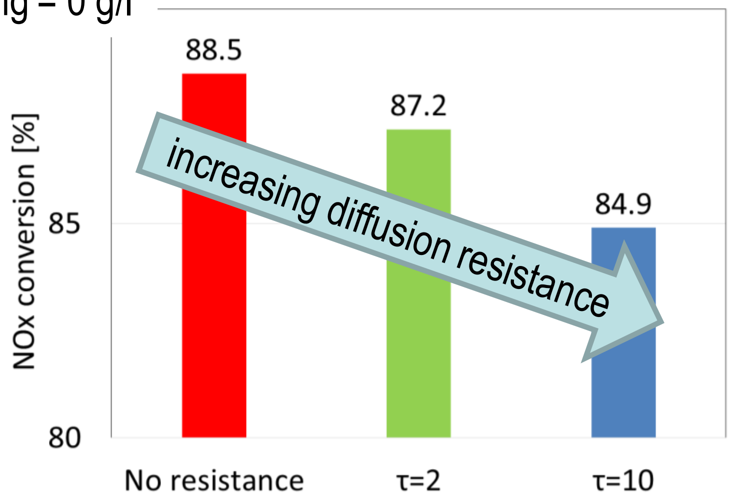
Internal diffusion effect on NOx conversion

Exothermia



NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

Soot loading = 0 g/l



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

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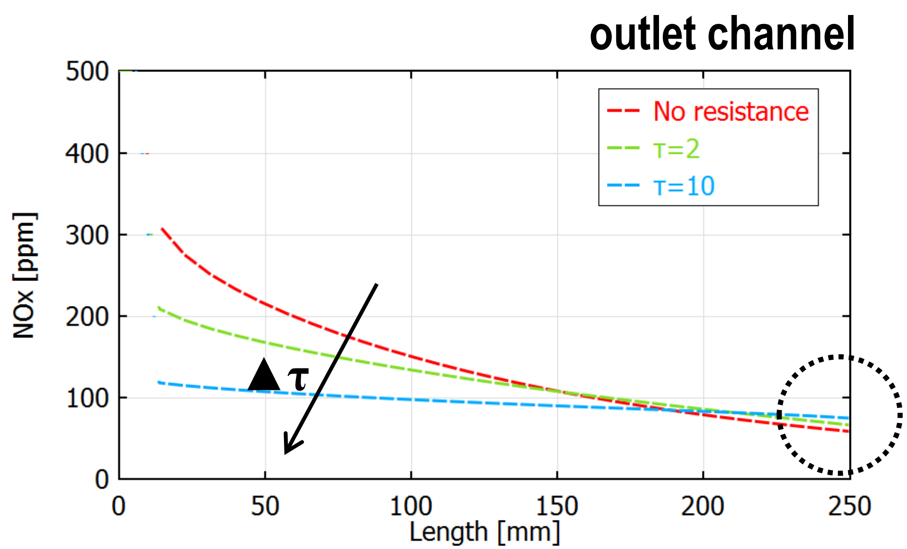
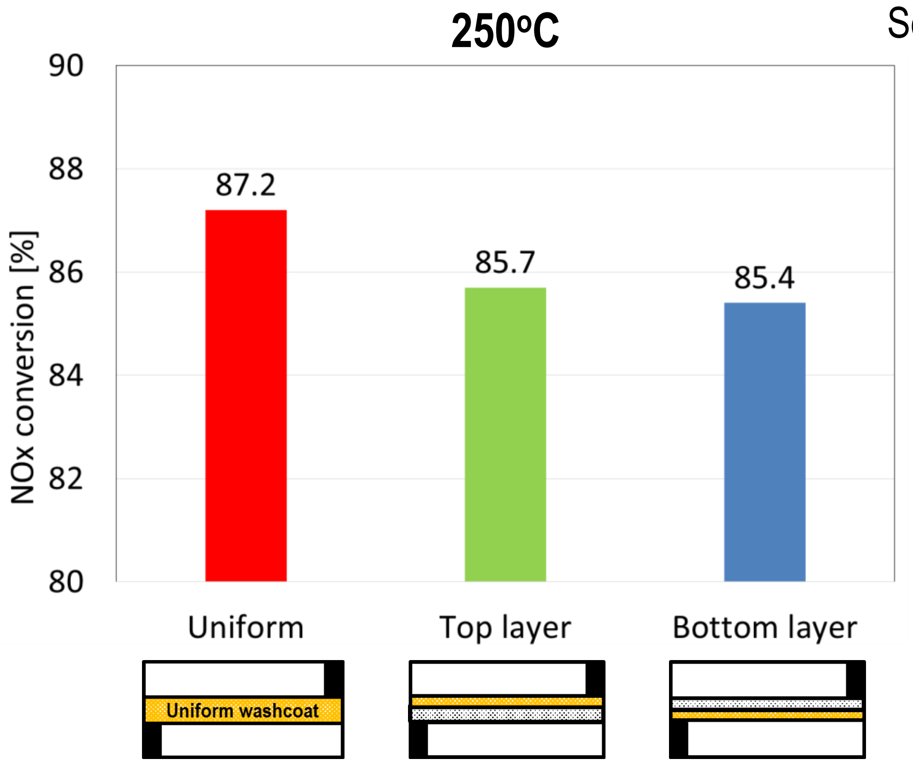


– Increased diffusion resistance decreases NOx conversion

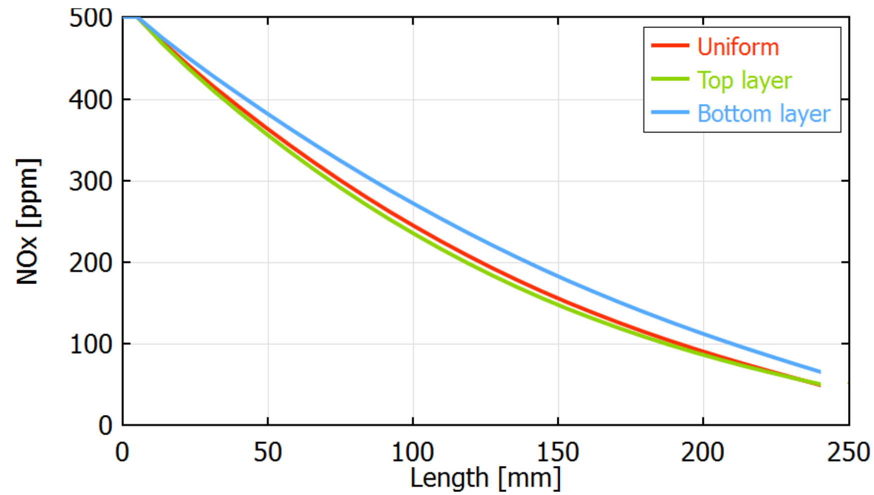
Effect of catalyst 'layering'

NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

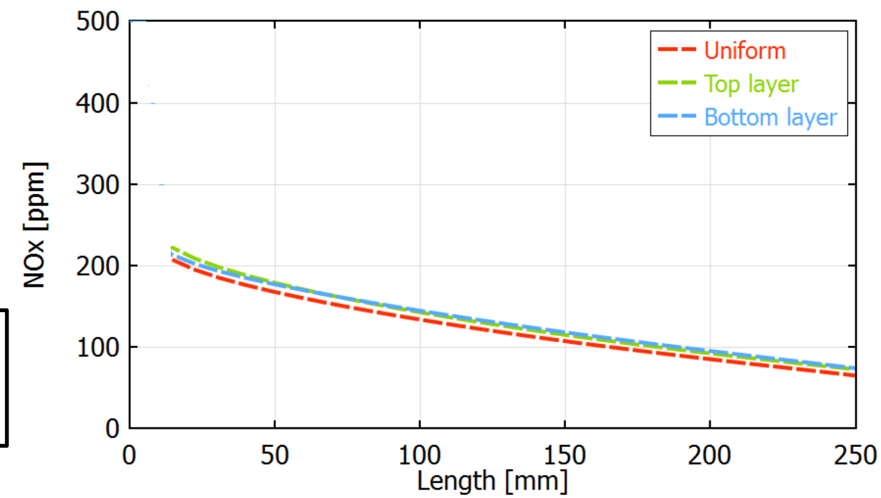
Soot loading = 0 g/l



inlet channel



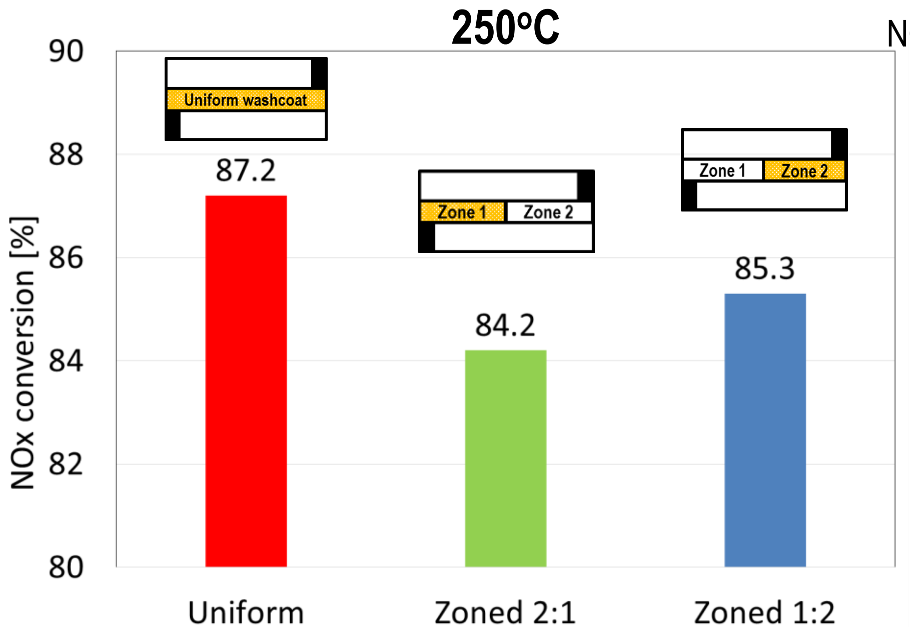
outlet channel

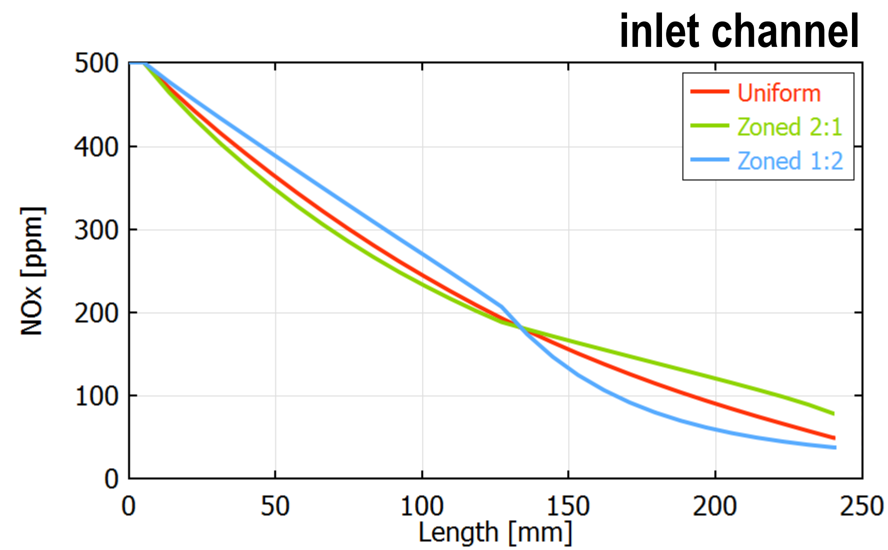


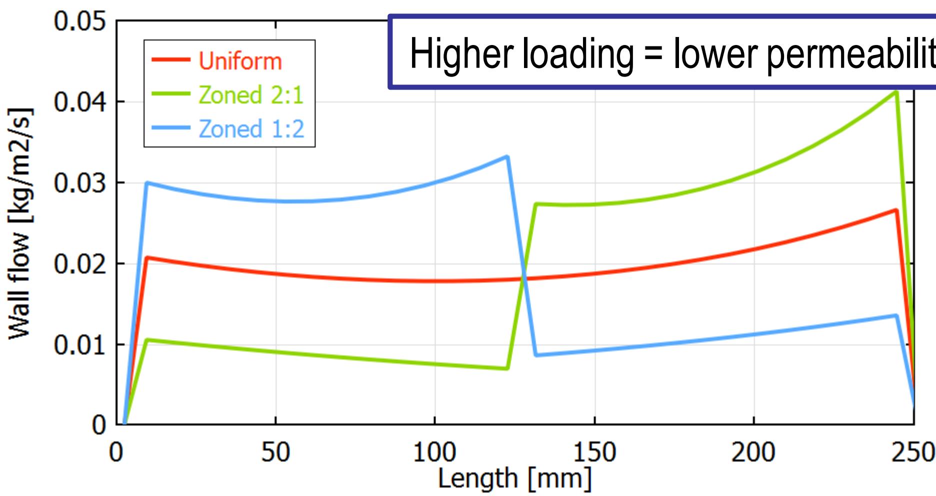
The inert part of the wall acts as a 'diffusion-barrier' NOx conversion decreases

Effect of catalyst 'zoning'

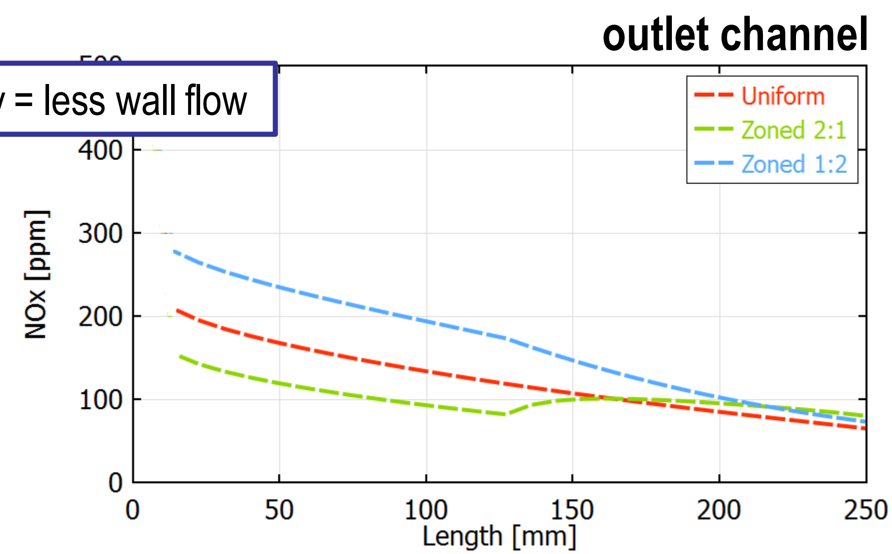
NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}



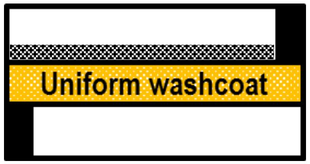




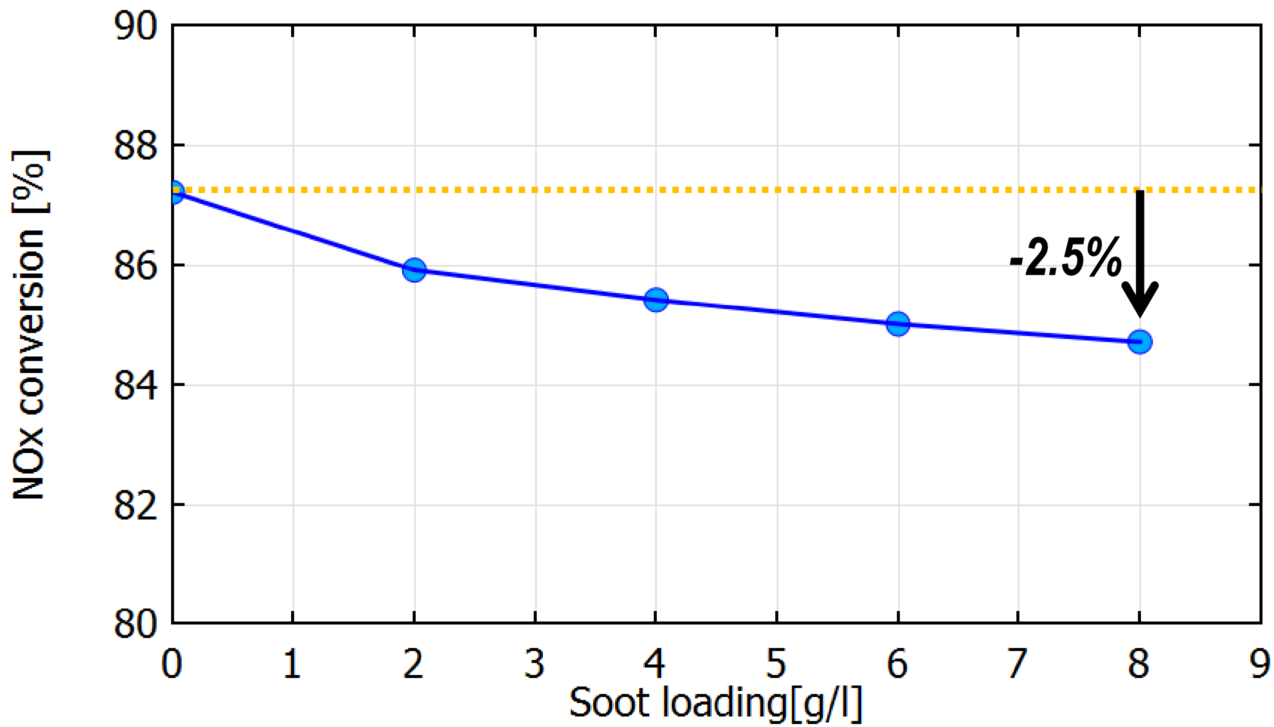
Higher loading = lower permeability = less wall flow



Soot effect on NOx conversion

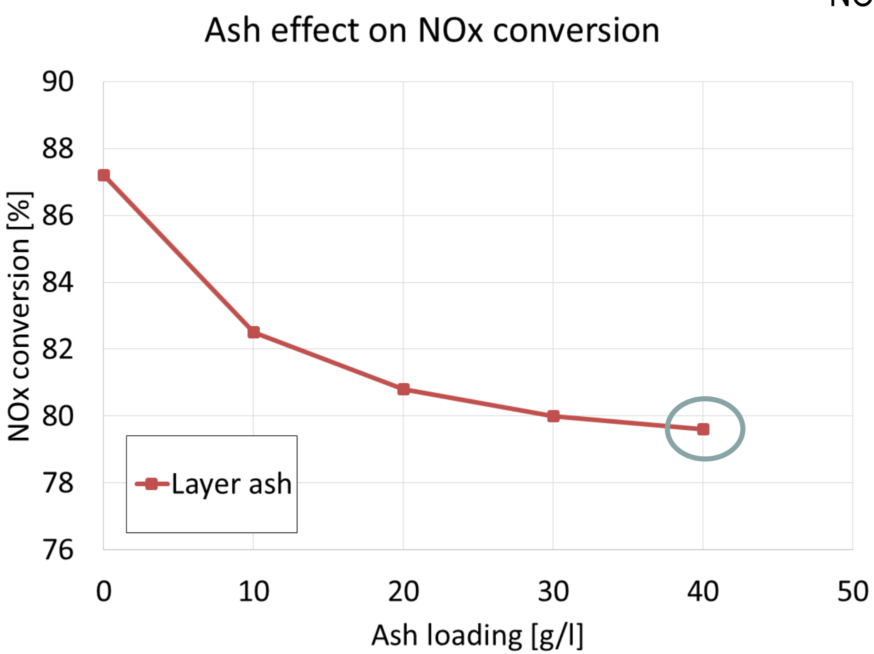


NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

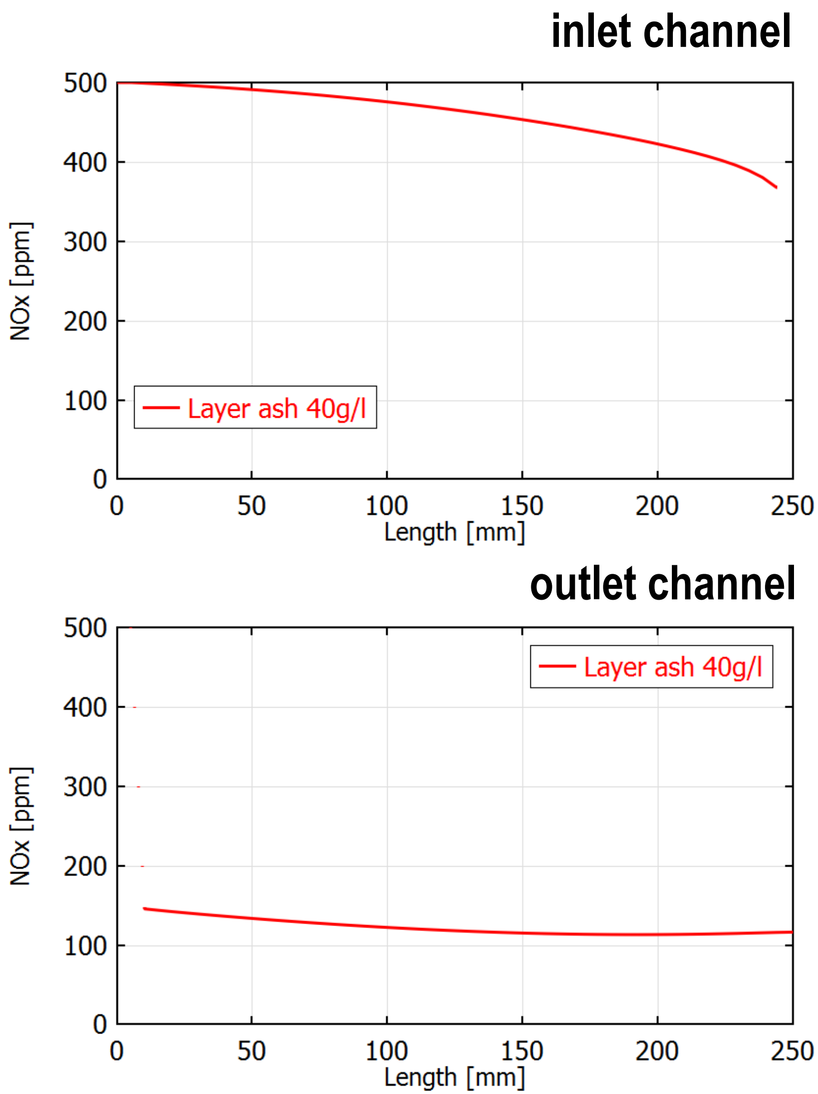


Soot has a **negative effect** on standard SCR acting as diffusion barrier

Ash effect on NOx conversion



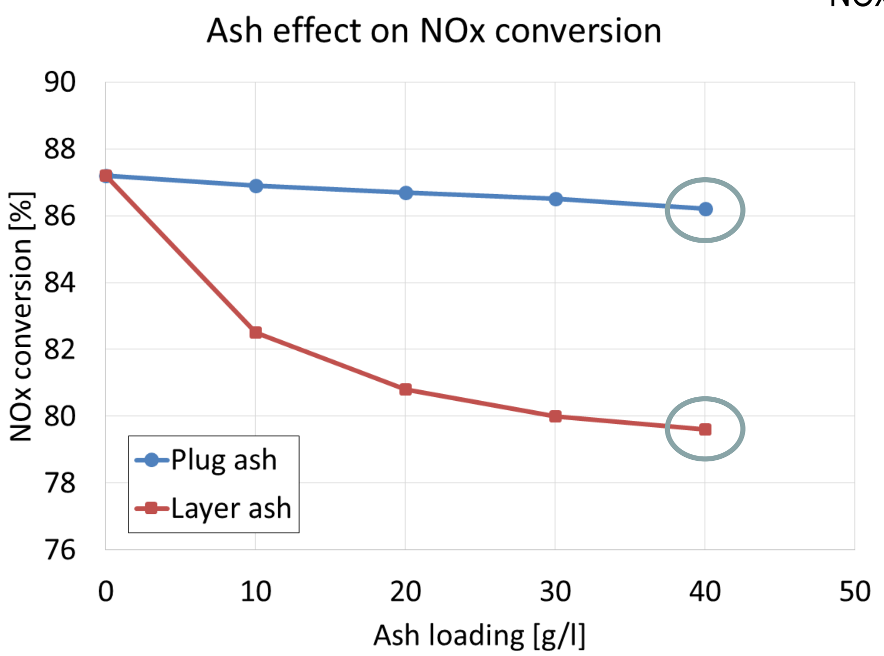
NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}

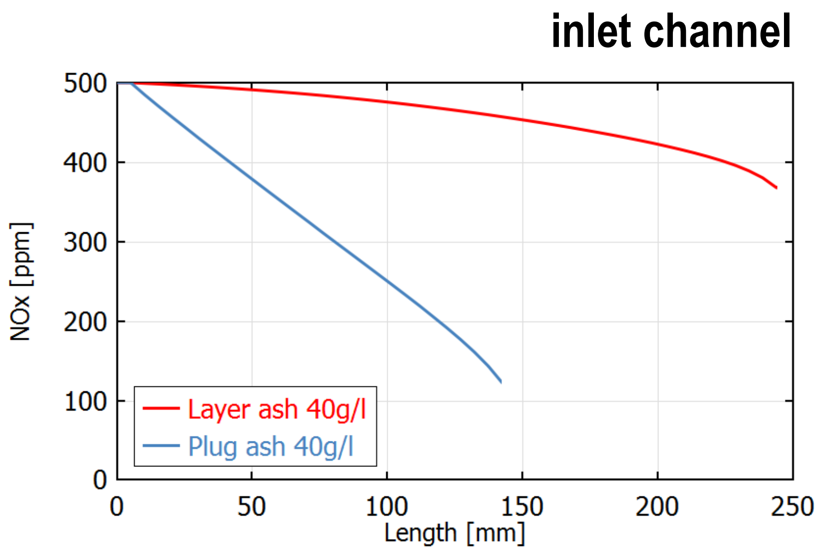


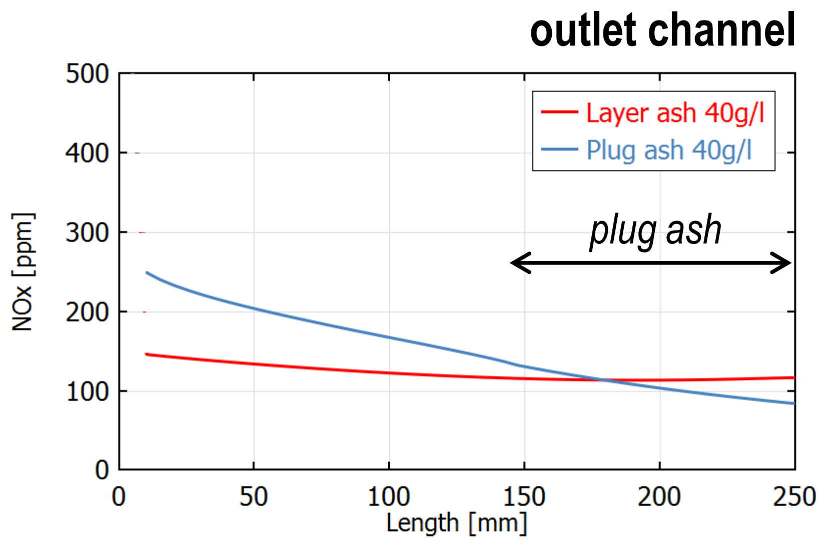
Ash property	Value
Packing density	300 kg/m^3
Permeability	5e-14 m^2
Pore diameter	100 nm
Pore volume fraction	90 %

Ash effect on NOx conversion

NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}





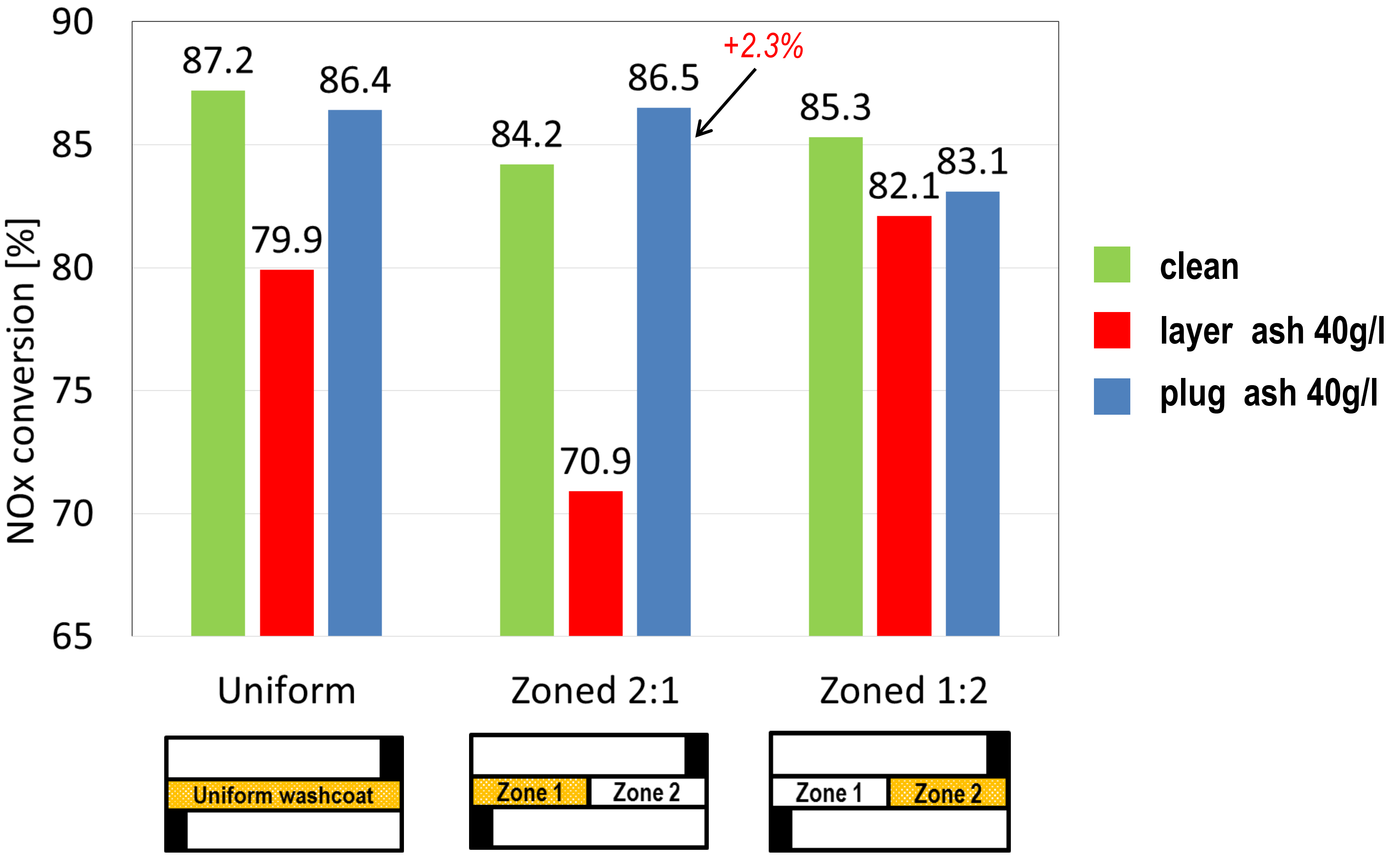


Ash property	Value
Packing density	300 kg/m^3
Permeability	5e-14 m^2
Pore diameter	100 nm
Pore volume fraction	90 %

Ash effect on NOx conversion
Impact of catalyst zoning

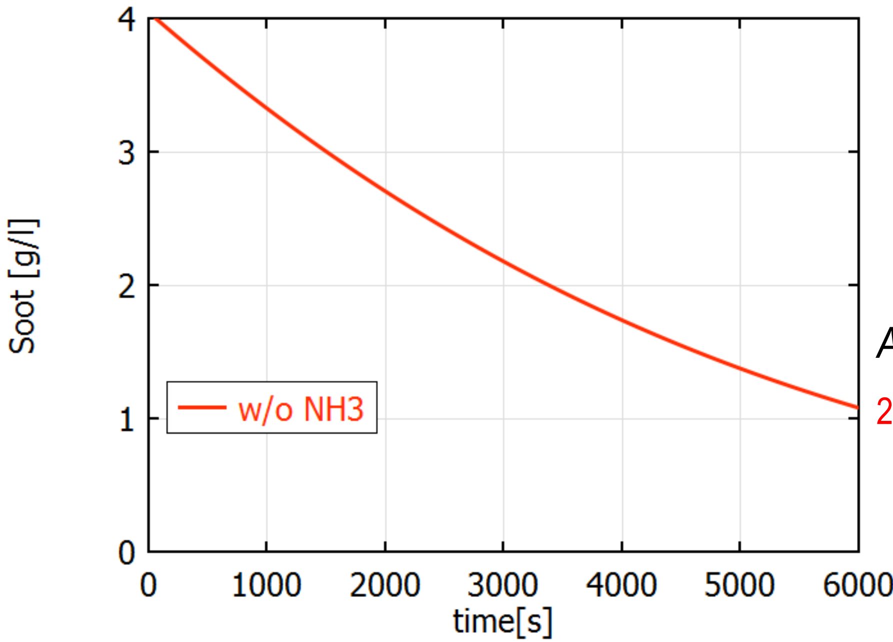
Exothermia

NOx=500ppm, NO_2/NOx=0, NH_3/NOx=1, T=250°C, GHSV=50,000 h^{-1}



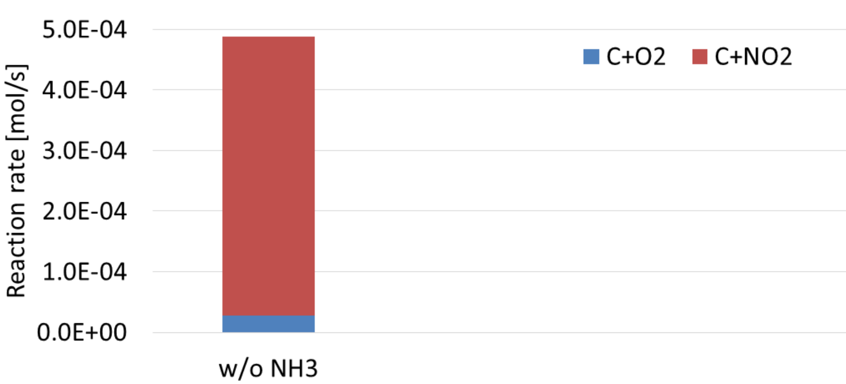
Effect of NH_3/SCR on soot oxidation

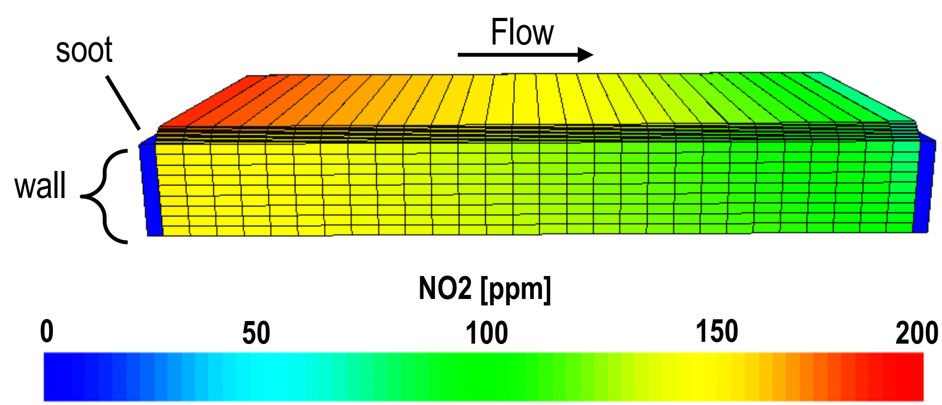
NOx=600ppm, NO_2/NOx=0.3, T=350°C, GHSV=90,000 h^{-1}



Average burn rate

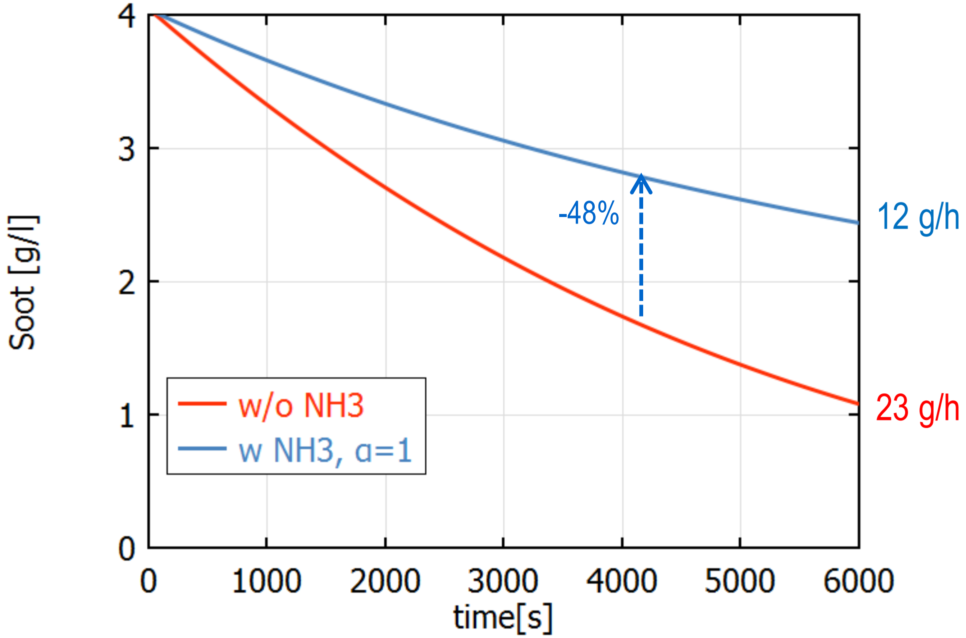
23 g/h



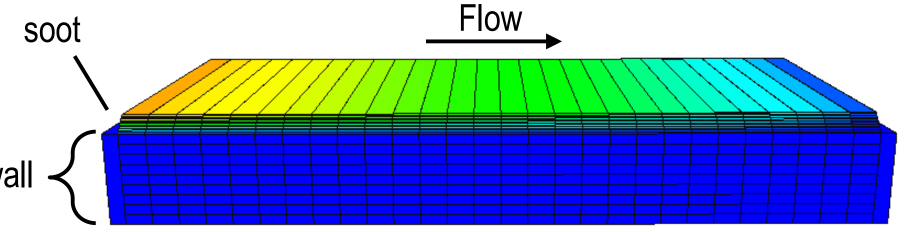


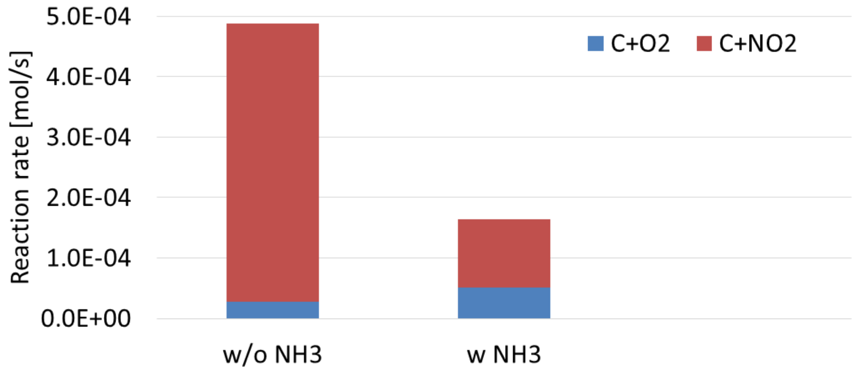
SCR effect on soot oxidation @ 350ºC

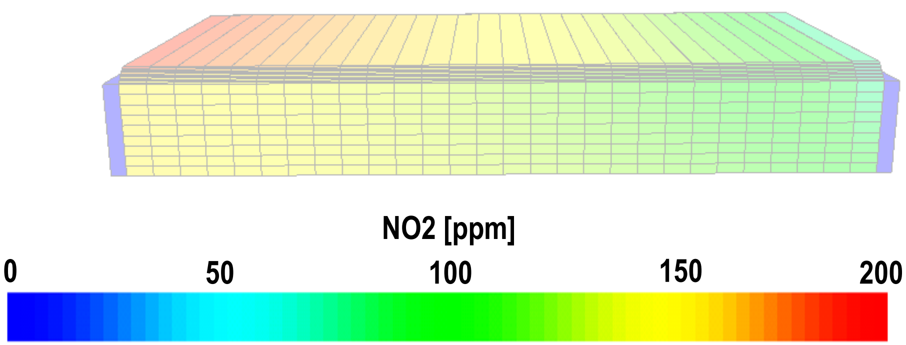
Exothermia



NOx=600ppm, NO_2/NOx=0.3, T=350°C, GHSV=90,000 h^{-1}



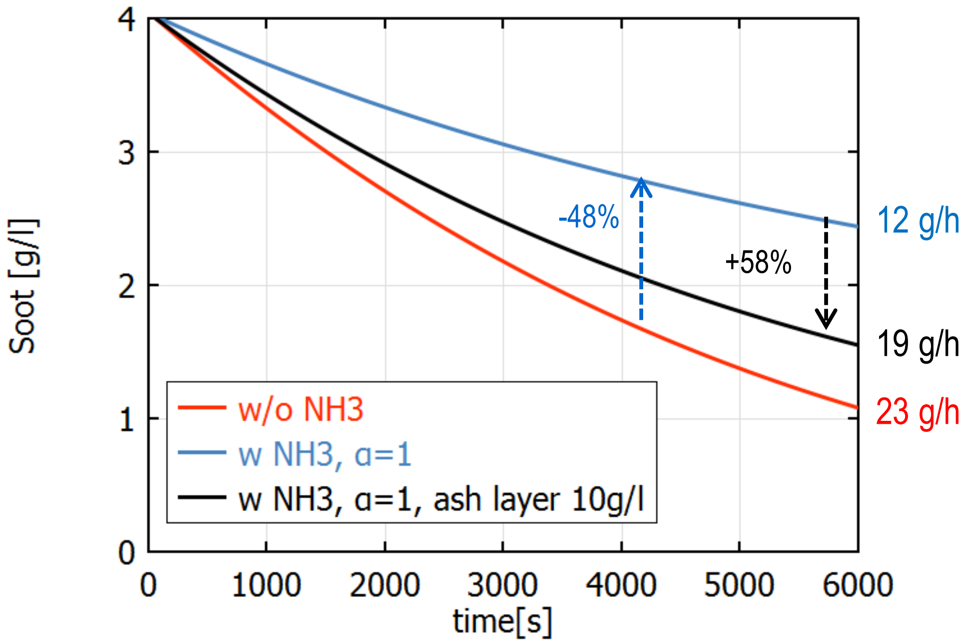


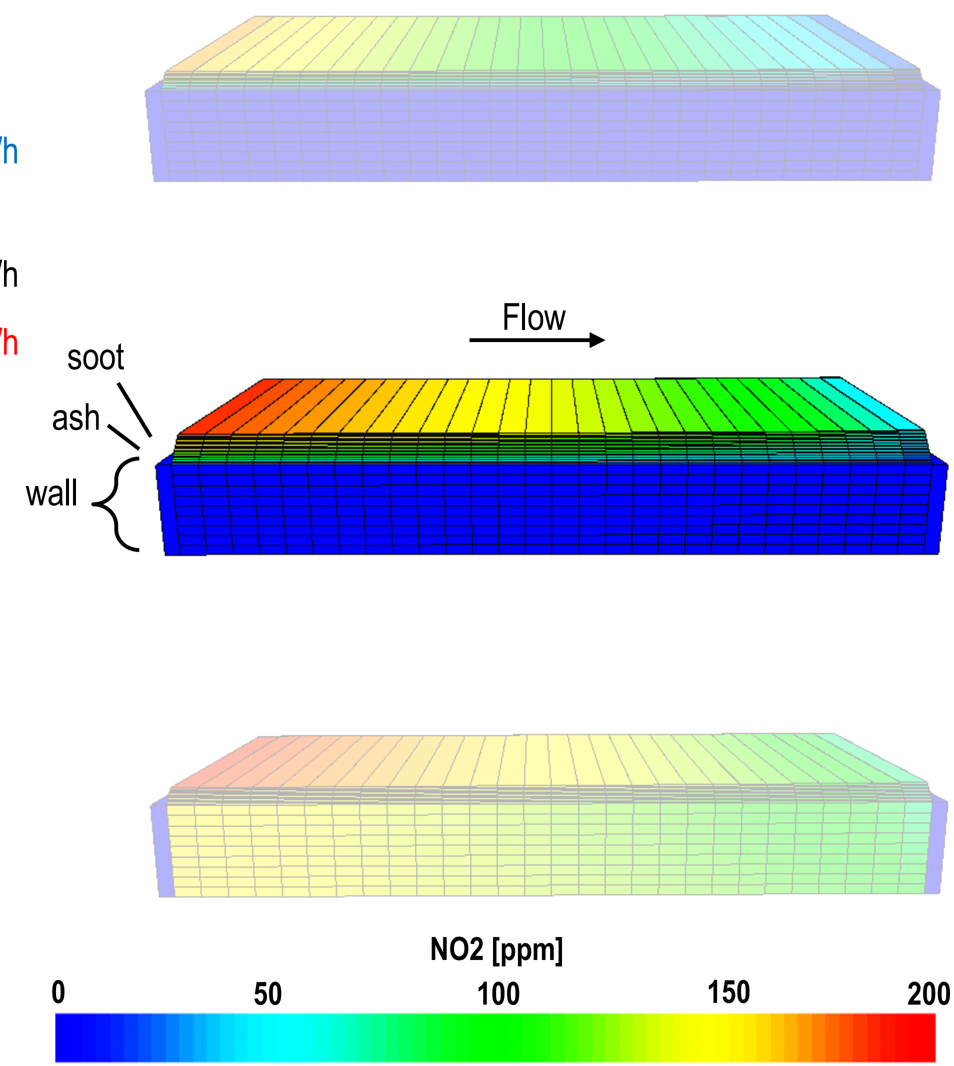


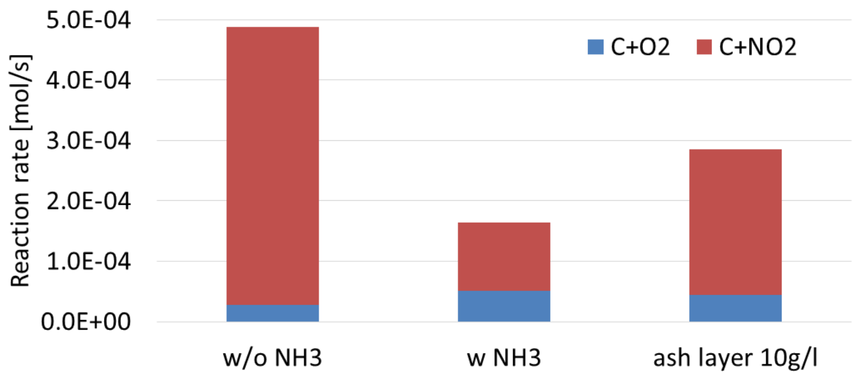
SCR effect on soot oxidation with ash @ 350ºC

Exothermia

NOx=600ppm, NO_2/NOx=0.3, T=350°C, GHSV=90,000 h^{-1}

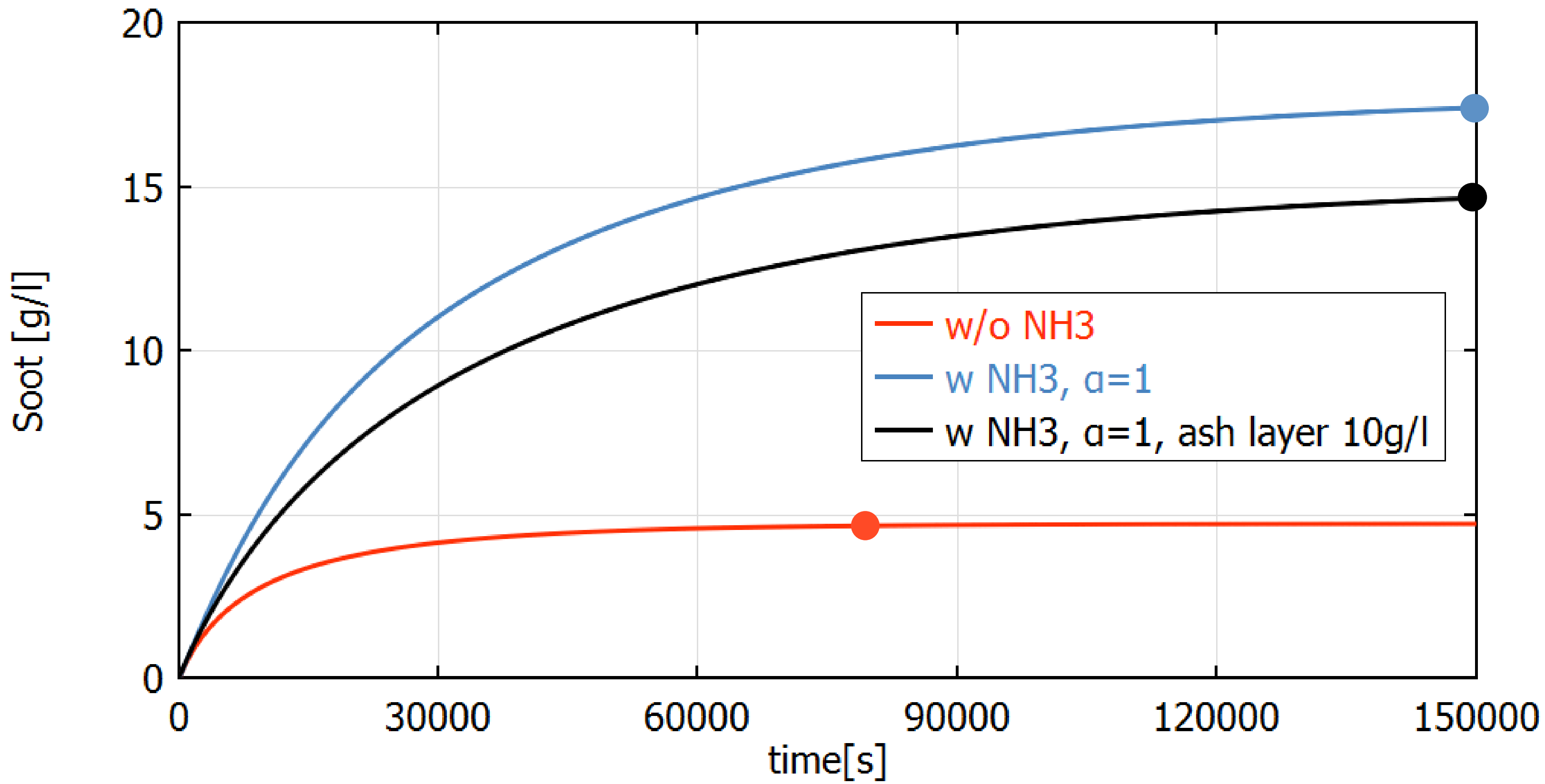






SCR and ash effect on soot balance point

T=350°C, Flow=1000kg/h, NO_2/NO_X=0.3, NO_X/soot=35 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!