Development and Validation of A Model for 2-way DPF/SCR

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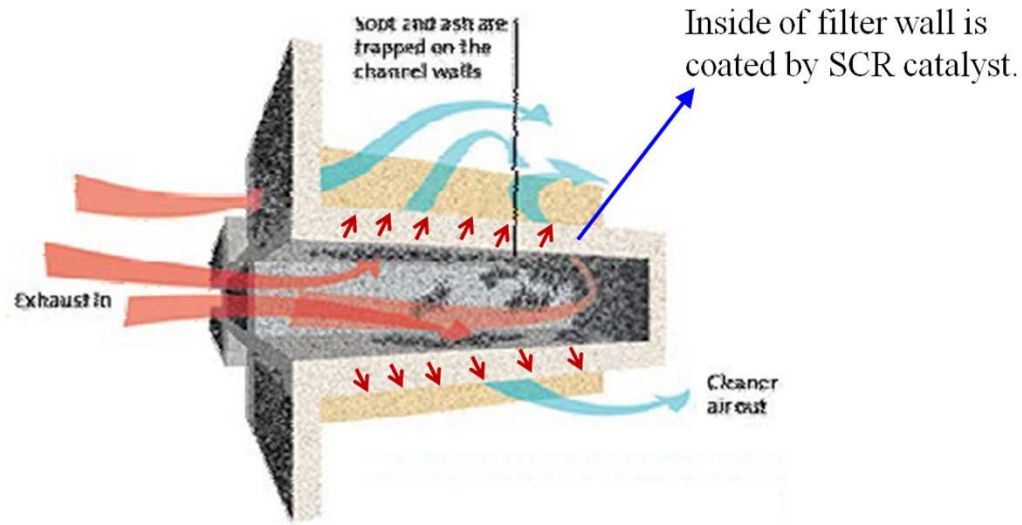
Contents

Introduction

What is a 2-way DPF/SCR aftertreament device?

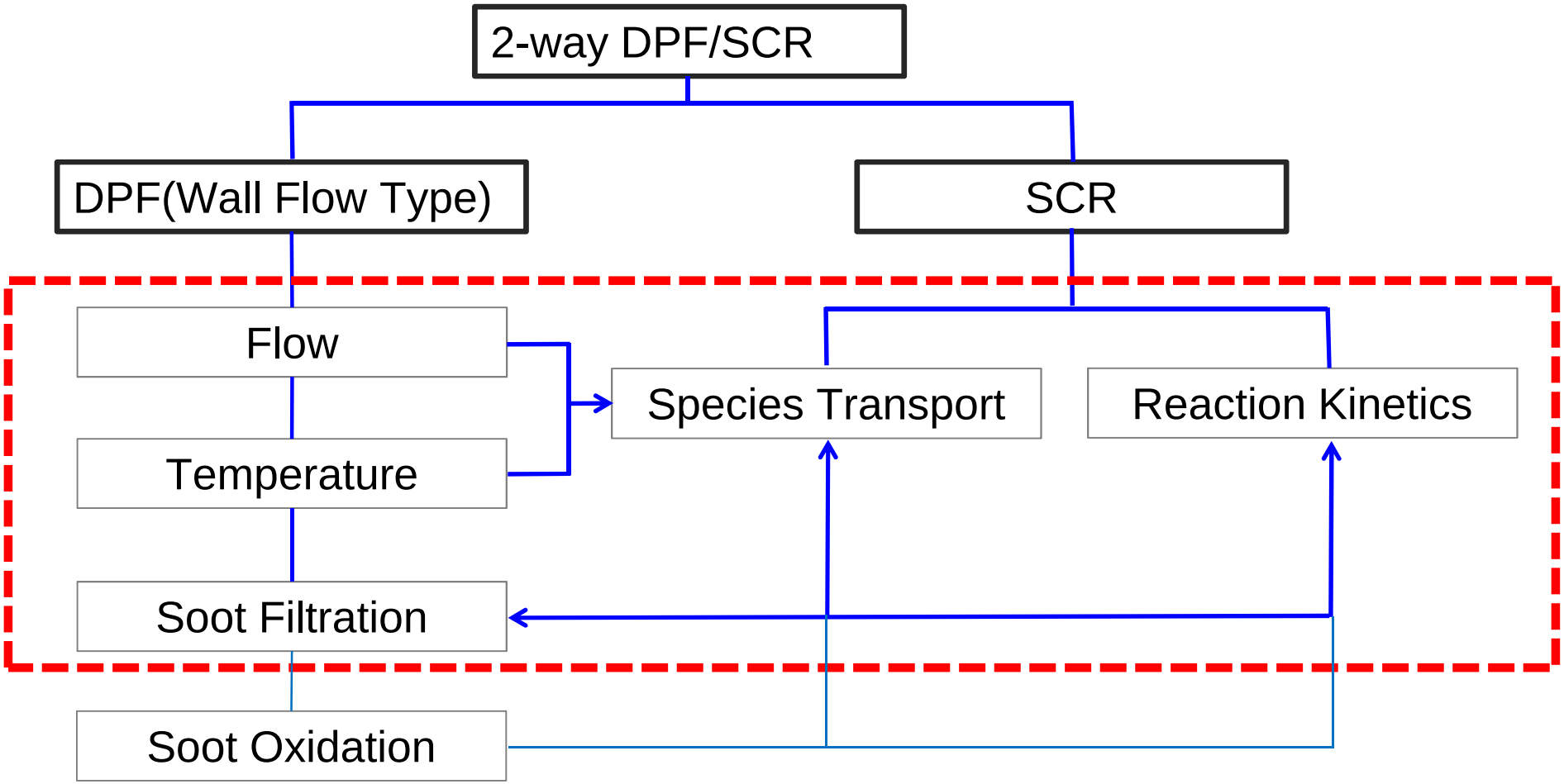
- Multifunctional device combining DPF and SCR
  a. Wall flow type DPF for soot trapping and oxidation
  b. SCR for NOx reduction



Issues

- DeNOx by SCR under wall flow type substrate
  - Model development for wall flow type SCR
  - Comparison study between flow channel type and wall flow type SCR
- Soot Filtration – DeNOx performance
- Soot Oxidation – DeNOx performance
  - NO_2 as a soot oxidizer and a reactant species in SCR

Model Structure



Model Development for Wall Flow Type SCR

Species Transport Equation(S.T.E)-General

$$\frac{\partial C_i}{\partial t} = -\nabla \cdot \mathbf{N_i} + R_{v,i}$$

where

$$\mathbf{N}_i = C_i \mathbf{v} + \mathbf{J_i}$$

$$\mathbf{J_i} = -D_i \nabla C_i$$

To solve this equation directly

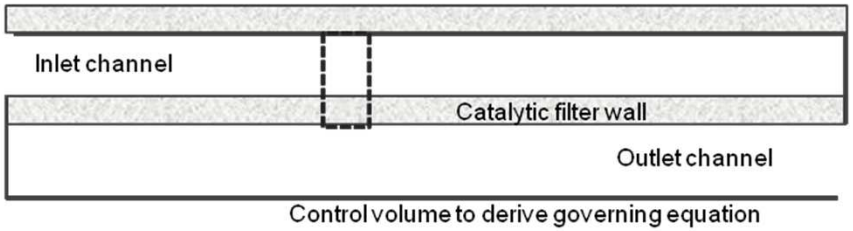
1. needs a lot of computer resources or solving time
2. is not an appropriate way for system simulation

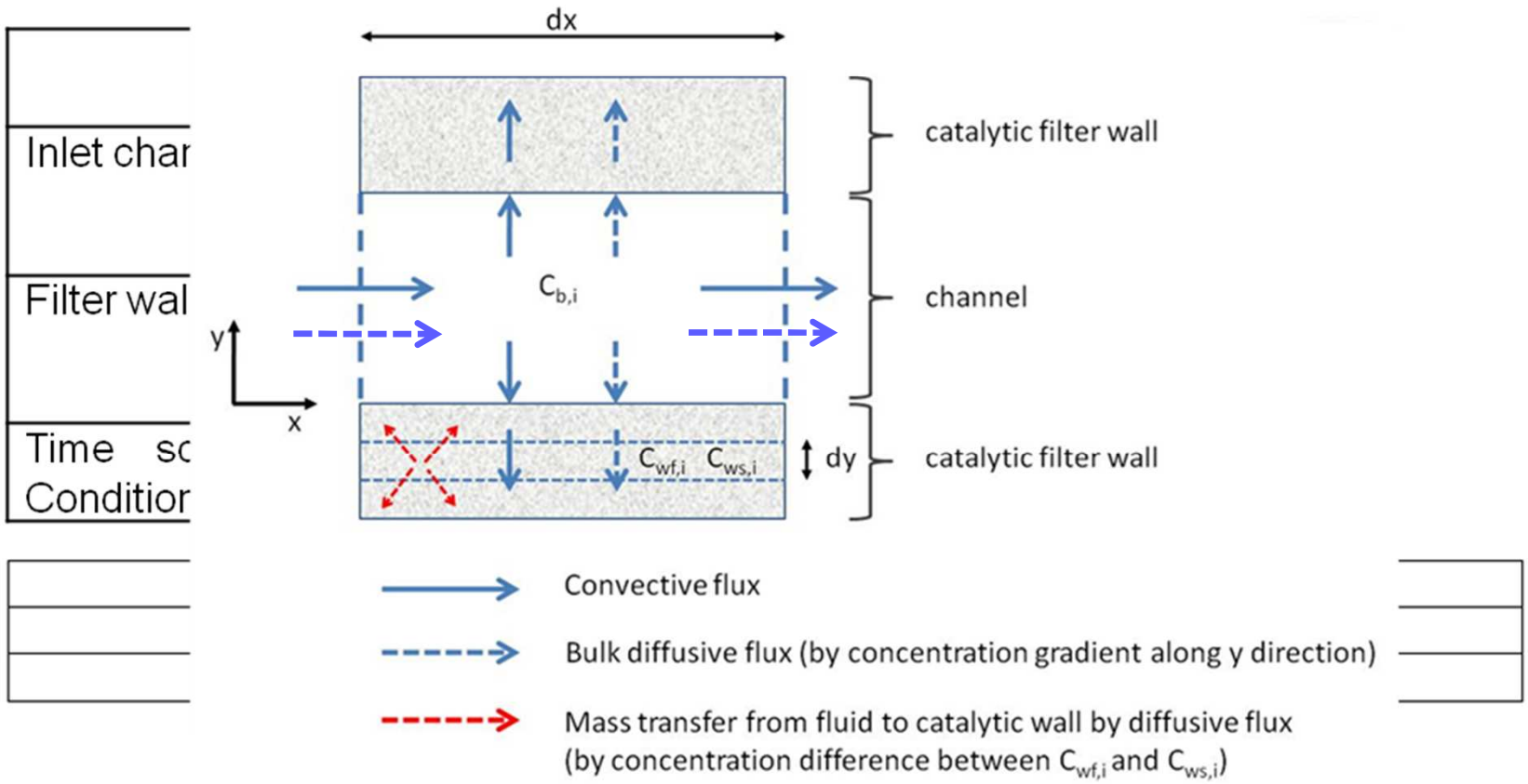
Scaling Analysis

	Transport by	Time scale	Order of Value (s)
Inlet channel	Convection along channel	L/U_{in}	~1E-2
	Diffusion along channel	L^2/D	~1E+2
	Diffusion to filter wall	a/k	~1E-2
Filter wall	Convection	w/u_w	~1E-2
	Diffusion along wall thickness	w^2/D_w	~1E-3
	Diffusion to catalyst surface	d_p/k_w	~1E-3
Time scale for external perturbation (Boundary Condition variation)			1~10

	Definition	Value
Pe_{ch}	$L \cdot U_{in}/D$	~1E+4
Pe_w	$w \cdot u_w/D_w$	~10

Scaling Analysis





Species Transport Equation (S.T.E.) - Simplified

$$\cancel{\frac{\partial C_i}{\partial t}} = -\nabla \cdot \mathbf{N_i} + R_{v,i}$$

Quasi steady assumption at each time step even in transient problem

where

$$\mathbf{N}_i = C_i \mathbf{v} + \cancel{\mathbf{J_i}}$$

In the inlet channel and outlet channel (axial direction)

$$\mathbf{J_i} = -D_i \nabla C_i$$

Final Form of S.T.E.

Gas Phase

I. $$\frac{\partial}{\partial x}(uC_{b,i}) + \frac{4}{a}(u_w C_{b,i}) + \frac{4k_c}{a}\left(C_{b,i} - C_{wf,i}\big|_{y=0}\right) = 0$$

B.C. $x = 0, \quad C_{b,i} = C_{b,i,inlet}$

Inlet Channel

II. $$\frac{\partial}{\partial y}(u_w C_{wf,i}) - D_w \frac{\partial^2 C_{wf,i}}{\partial y^2} + k_w S(C_{wf,i} - C_{ws,i}) = 0$$

B.C. $y = 0, \quad C_{wf,i} = C_{wf,i}\big|_{y=0} \qquad y = w, \quad \frac{\partial C_{wf,i}}{\partial y} = 0$

Catalytic filter wall

III. $$k_w S(C_{wf,i} - C_{ws,i}) = a_s \sum_k RR(k)$$

Catalytic filter wall

IV. $$C_b u_w + k_c\left(C_{b,i} - C_{wf,i}\big|_{y=0}\right) = C_{wf,i}\big|_{y=0} \cdot u_w - D_w \frac{\partial C_{wf,i}}{\partial y}\bigg|_{y=0}$$

Interface of Inlet channel and Catalytic filter wall

Adsorption phase (NH_3 only)

$$\frac{d\theta}{dt} = RR(1) - RR(2) - 2RR(3) - 4RR(5) - 2RR(6) - 4RR(7) - 2RR(8)$$

= net adsorption – ammonia oxidation – SCR reaction

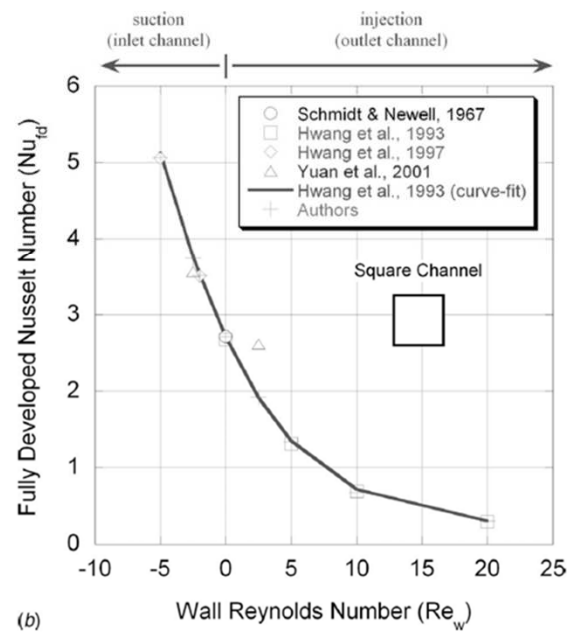
Final Form of S.T.E.

$$\frac{\partial}{\partial x}\left(u^o C_{b,i}{}^o\right) + \frac{4}{a}\left(u_w C_{wf,i}\Big|_{y=w}\right) = 0$$

$$C_{b,out} = \frac{1}{a \cdot u_{out}} \int 4\left(u_w C_{wf,i}\Big|_{y=w}\right) dx$$

Outlet channel

- Diffusion through boundary layer can be ignored(wall injection)
- Convective flux only → Channel out concentration can be found by simple mass balance



Depsick et. al., Journal of Engineering for Gas Turbines and Power 2008 vol.130

Solver Development

- Total 4n+1 coupled equations and 4n+1 unknowns of $C_{b,i}$ $C_{wf,i}$, $C_{ws,i}$, $C_{wf,i}(y=0)$ and θ
  - 2n equations of differential equations
  - 2n equations of algebraic equations
  - One equation of differential equation ($d\theta/dt$)

- 2n differential equations are converted to algebraic equation by FVM(Finite Volume Method).

SCR Reaction and its Rate Expression

Description	Reaction Rate	Rate Expression
ammonia adsorption ammonia desorption	RR(1) RR(2)	$RR(1) = A_1 C_{NH3}(1-\theta)$ $RR(2) = A_2 \exp\left(\frac{-E_{A,2}}{R_u T}\right)\theta$
ammonia oxidation	RR(3)	$RR(3) = A_3 \exp\left(\frac{-E_{A,3}}{R_u T}\right) C_{O2} \cdot \theta$
NO oxidation	RR(4)	$RR(4) = A_4 \exp\left(\frac{-E_{A,4}}{R_u T}\right) C_{NO}\sqrt{C_{O2}} \cdot \theta - k_{4,b} C_{NO2}$ where $k_{4,b} = A_4 \exp\left(\frac{-E_{A,4}}{R_u T}\right) / K_{eq}$ and $K_{eq} = \exp\left(\frac{-\Delta S}{R_u}\right) \exp\left(\frac{-\Delta H}{R_u T}\right)$
Standard NH_3 SCR	RR(5)	$RR(5) = A_5 \exp\left(\frac{-E_{A,5}}{R_u T}\right) C_{NO} \cdot \theta$
Rapid NH_3 SCR	RR(6)	$RR(6) = A_6 \exp\left(\frac{-E_{A,6}}{R_u T}\right) C_{NO} C_{NO2} \cdot \theta$
NH_3 SCR with NO_2	RR(7)	$RR(7) = A_7 \exp\left(\frac{-E_{A,7}}{R_u T}\right) C_{NO2} \cdot \theta$
N_2O formation by SCR	RR(8)	$RR(8) = A_8 \exp\left(\frac{-E_{A,8}}{R_u T}\right) C_{NO2} \cdot \theta$

Modeling Ea

Non-Linear

Non-Linear

Activation Energy for Desorption

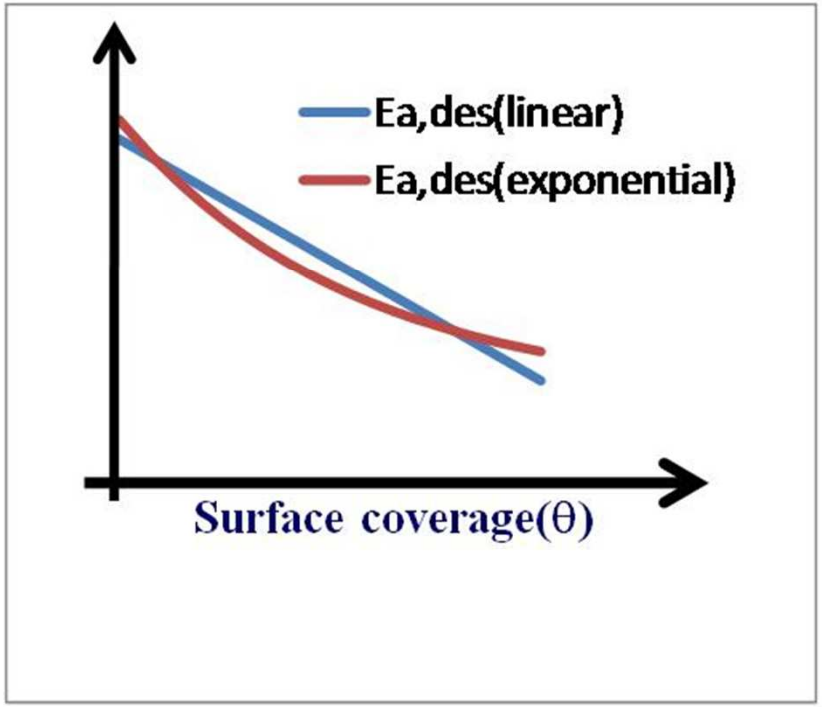
Dependence of activation energy on surface coverage

1. **Linear form**

$$E_{a,des} = E_{a,des0}(1-\alpha\theta)$$

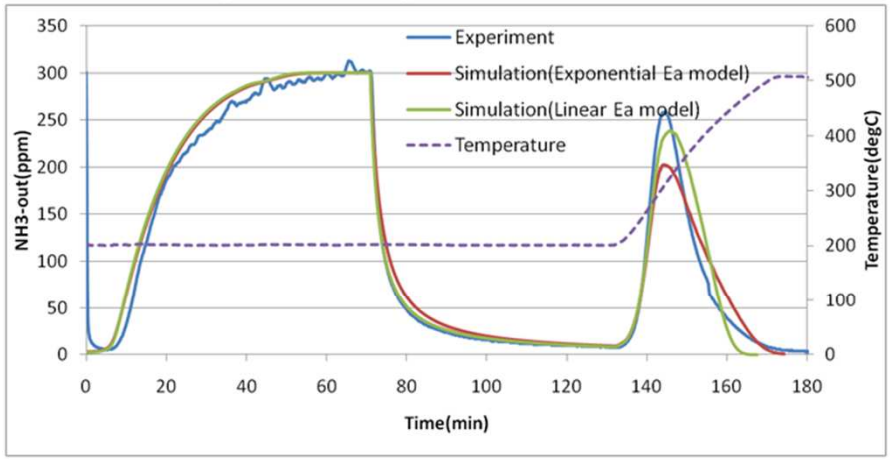
2. **Exponential form**

$$E_{a,des} = E_{a,des1}\exp(-\beta\theta) + E_{a,des2}$$

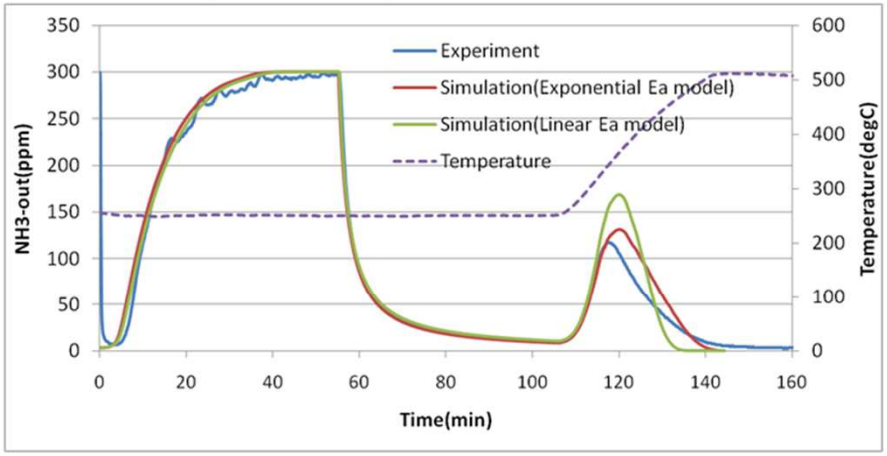


Validation of Model -TPD

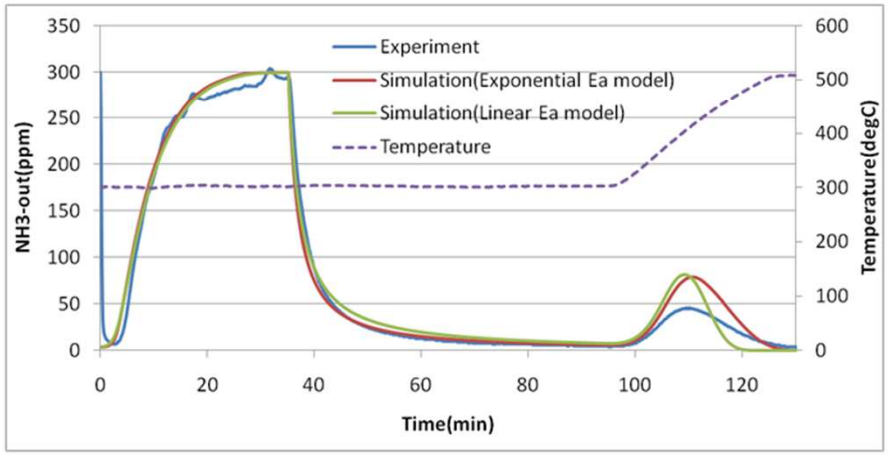
Initial Temp. : 200degC



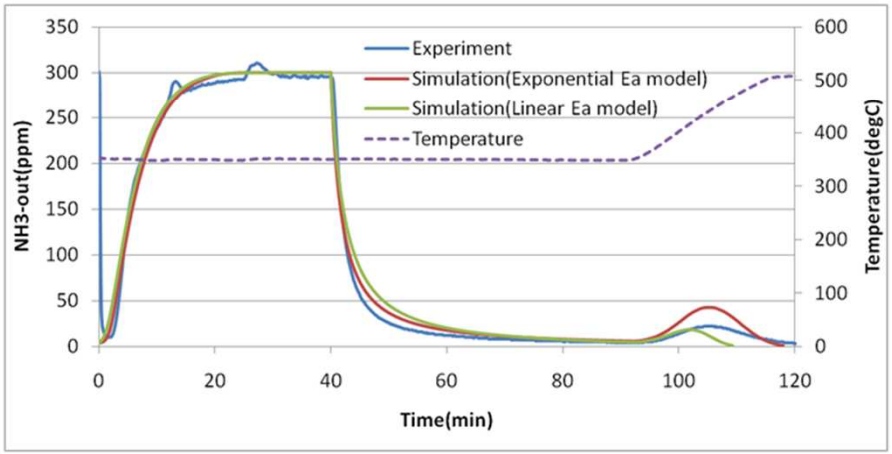
Initial Temp. : 250degC



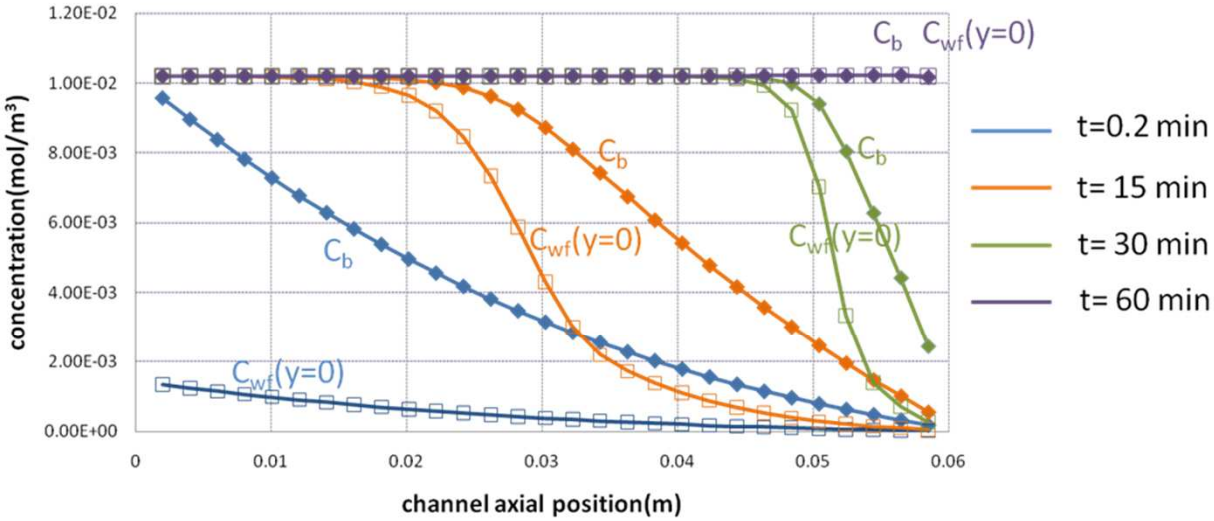
Initial Temp. : 300degC

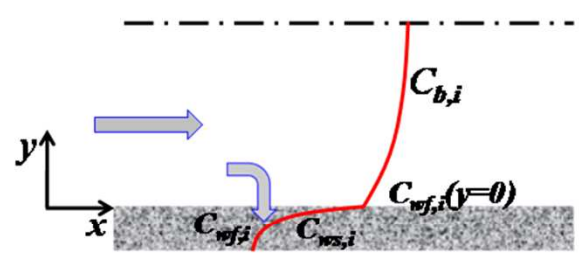


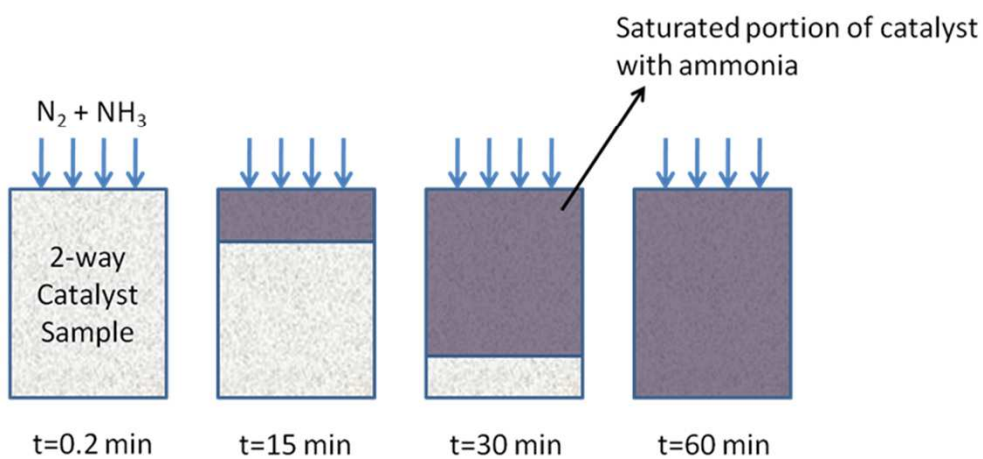
Initial Temp. : 350degC

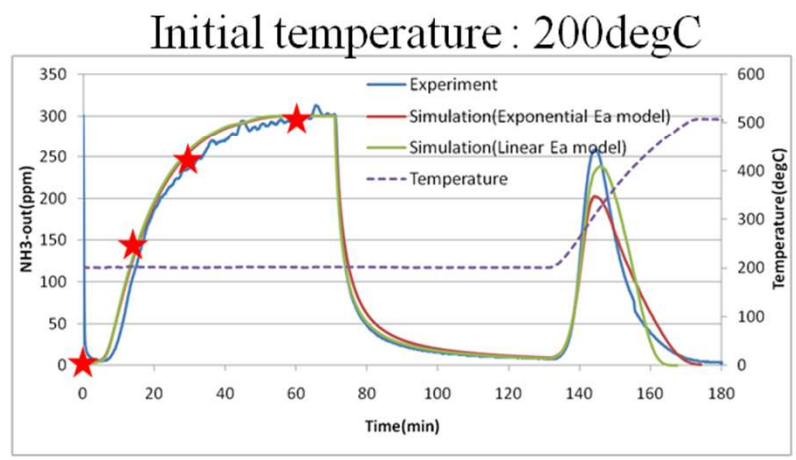


TPD-NH_3 Concentration Distribution







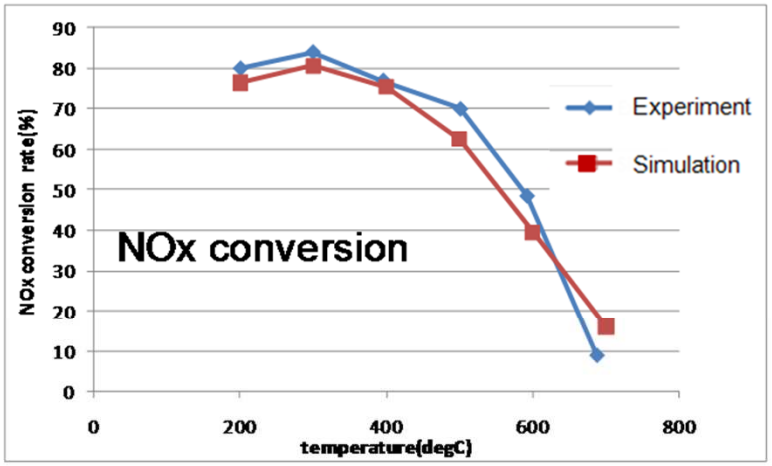


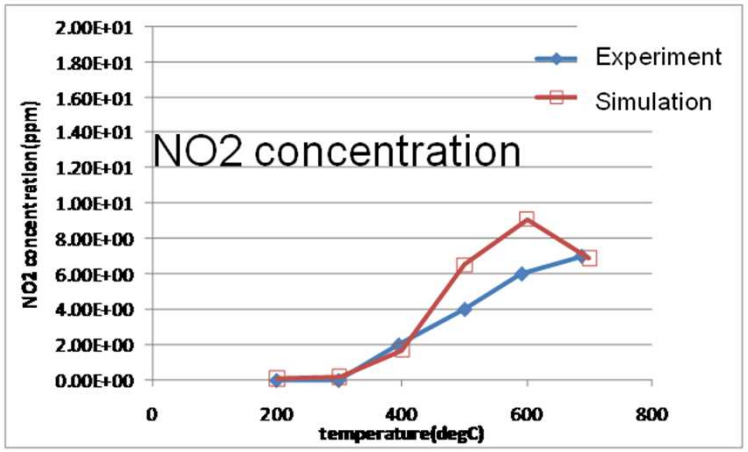
Model Validation –DeNOx (Steady State)

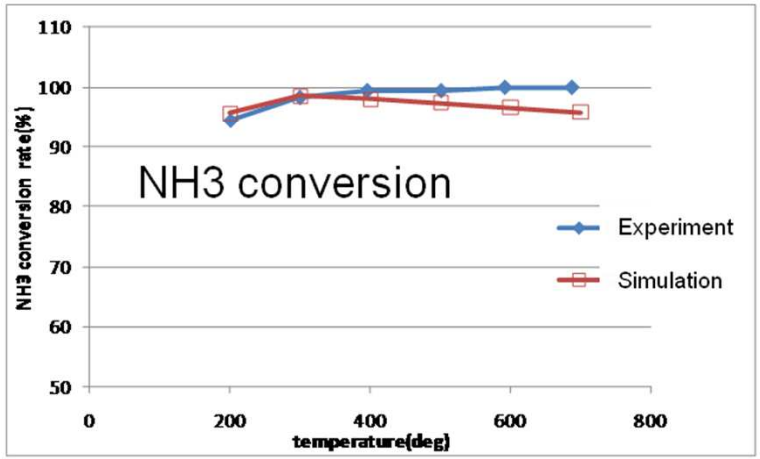
1. Model is calibrated under no soot loading and α=0.9 condition

2. Reaction constants for adsorption and desorption are carried over from TPD results.

3. Reaction constants for RR(3) to RR(8) are calibrated to match the simulation results to experiment's

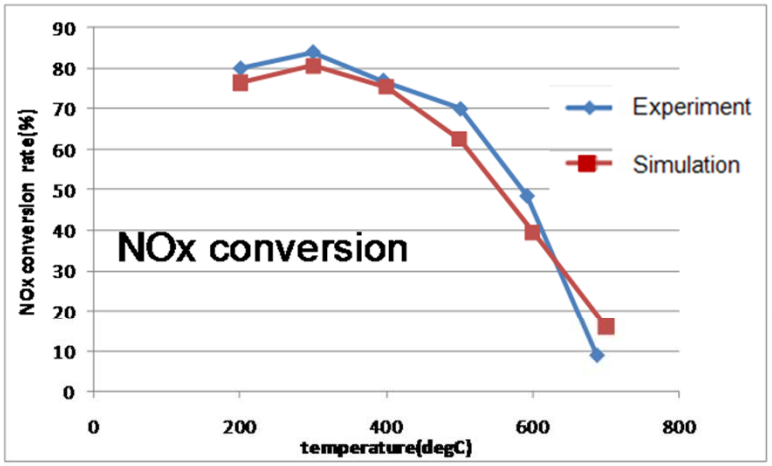


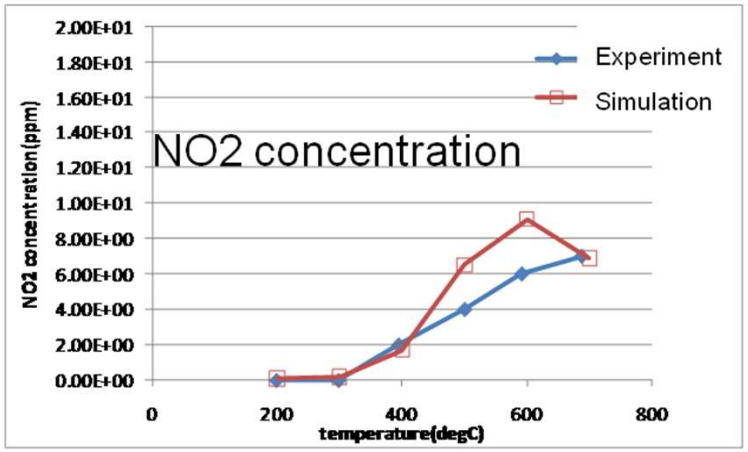


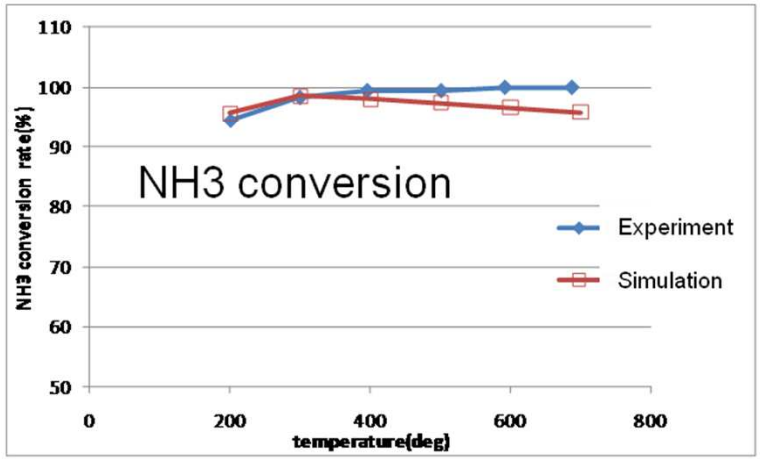


Model Validation –DeNOx (Steady State)

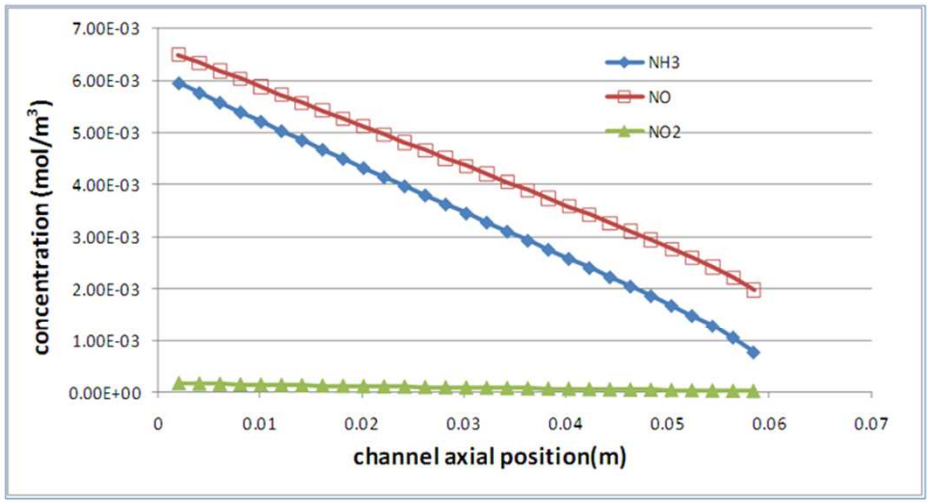
Parameter	Value
NO	195 ppm
NO_2	5 ppm
NH_3	180 ppm
S.V.	23000 (1/hr)



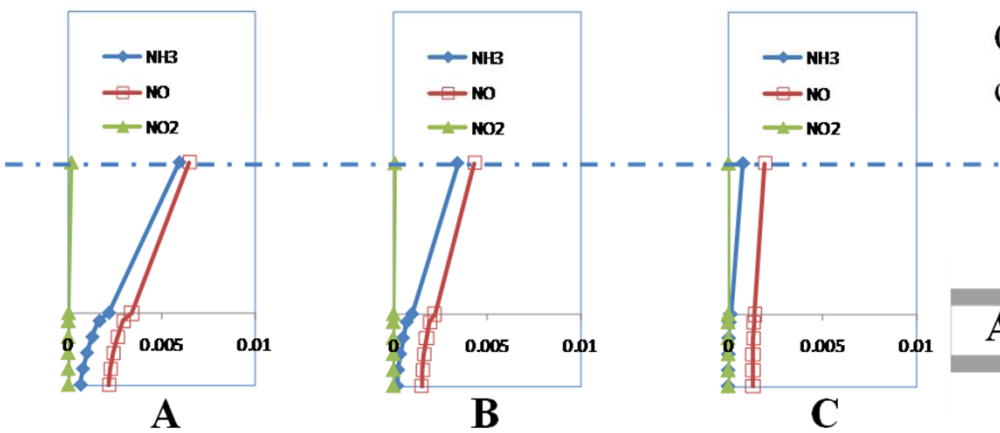




DeNOx-Species Spatial Distribution

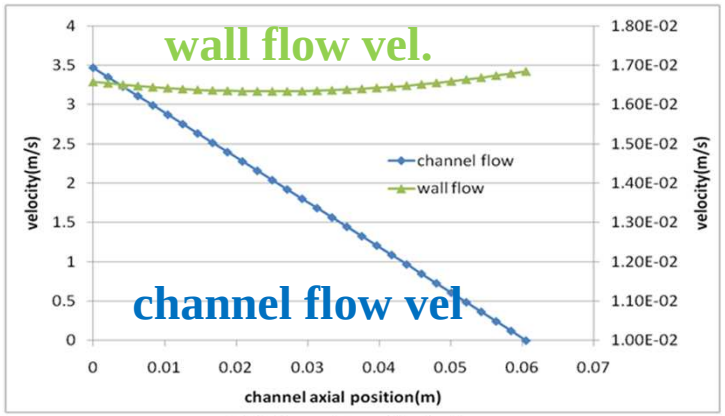


Concentration distribution along the channel axial position

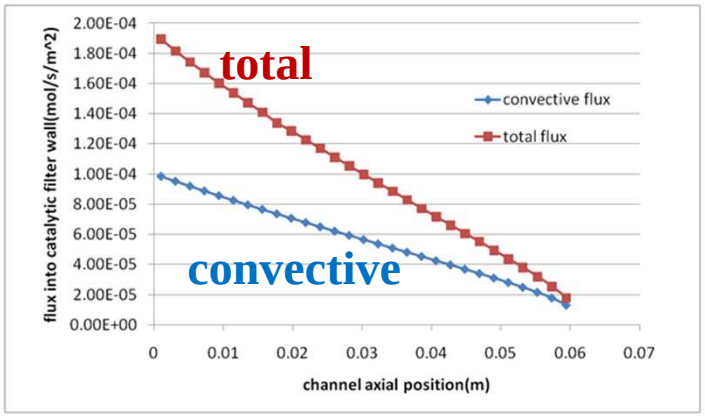


Concentration distribution through catalytic filter wall

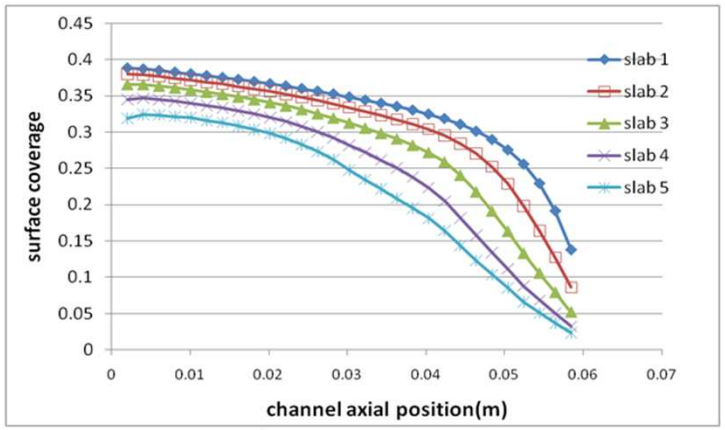
Vel., Flux, and Surface coverage



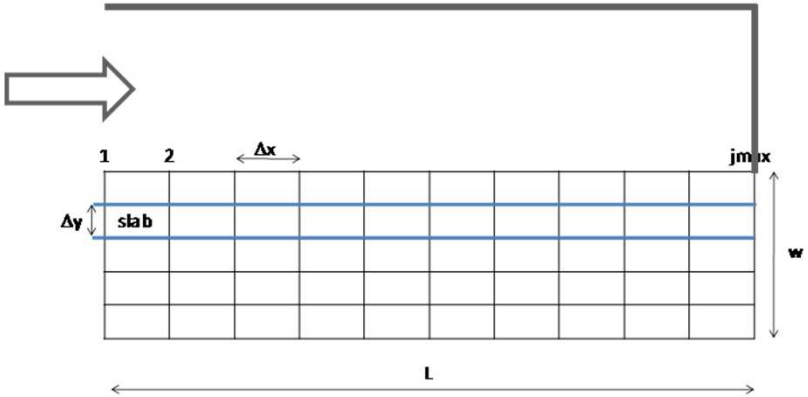
Velocity field



Mass flux to catalytic filter wall



Surface Coverage



Monitoring Ammonia Balance

Manipulating the S.T.E for ammonia

$$\dot{N}_{in,NH3} = \underbrace{\dot{N}_{out,NH3}}_{A} + \underbrace{\int a_s \frac{d\theta}{dt} dV}_{B} + \underbrace{\int a_s \cdot 2RR(3) dV}_{C} + \underbrace{\int a_s (4RR(5) + 2RR(6) + 4RR(7) + 2RR(8))\, dV}_{D}$$

A: rate of ammonia slip
B : net ammonia adsorption rate
C : rate of ammonia oxidation
D : SCR reaction rate

Test condition–NH3 Balance

Test Condition

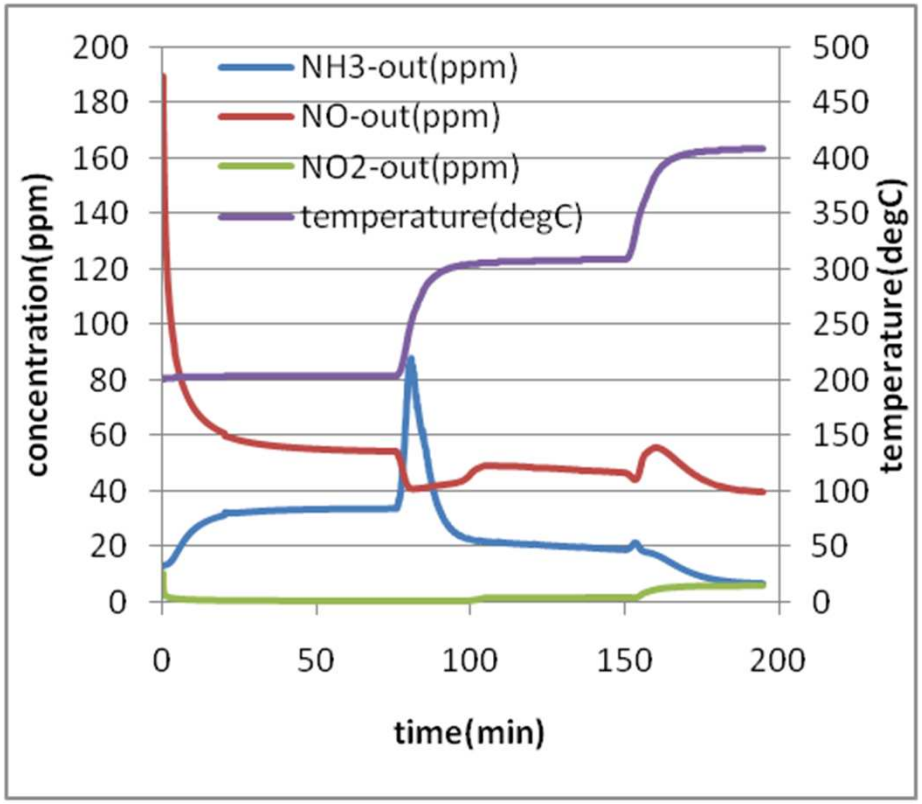
1. No adsorbed ammonia initially
2. Constant rate of ammonia injection
3. Step temperature increasing at certain points

Parameter	Value
Space Velocity	23000(1/hr)
NO	200 ppm
NO2	0 ppm
NH3	180 ppm

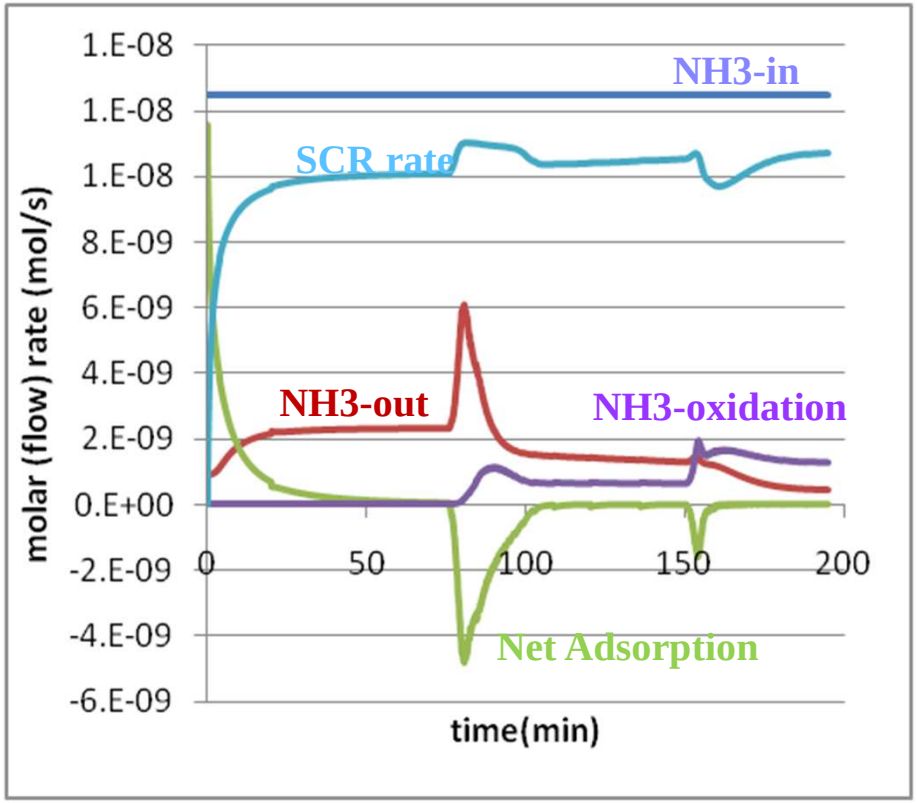
Inlet condition

Results –NH3 Balance

Outlet concentration

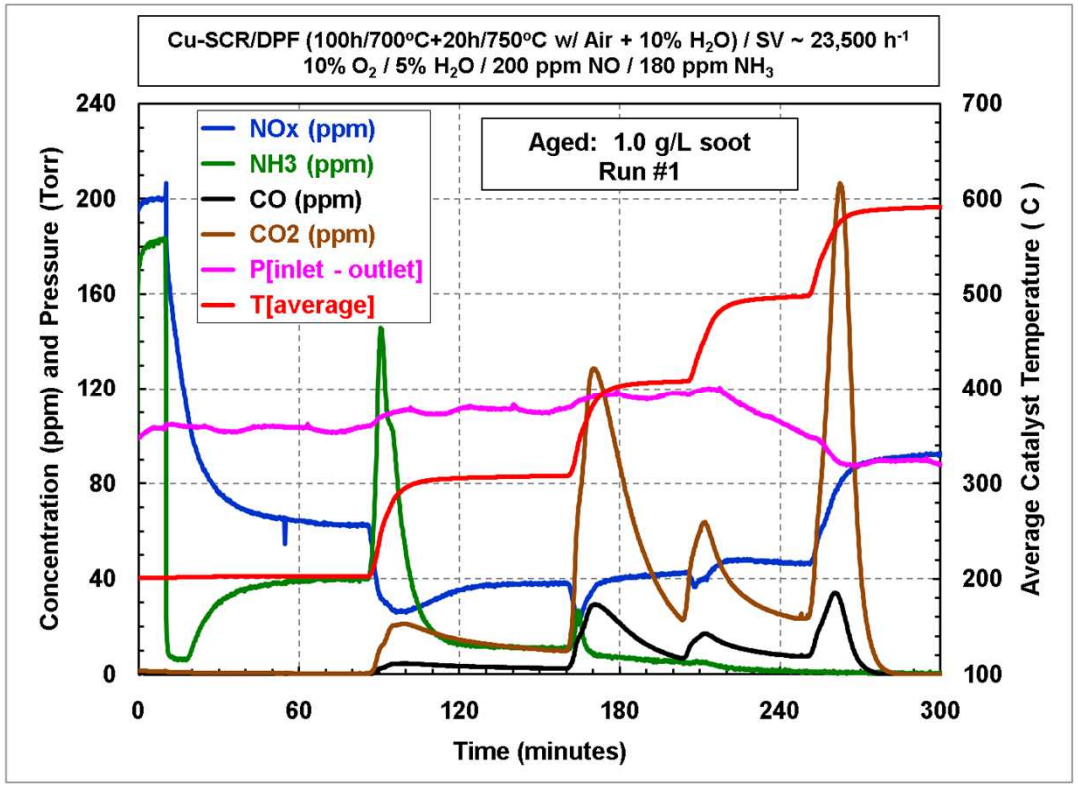


Ammonia Balance



Effect of Soot Deposit (Filtered Soot) on SCR Reaction

Review –Experiment(I)



Observation (I)

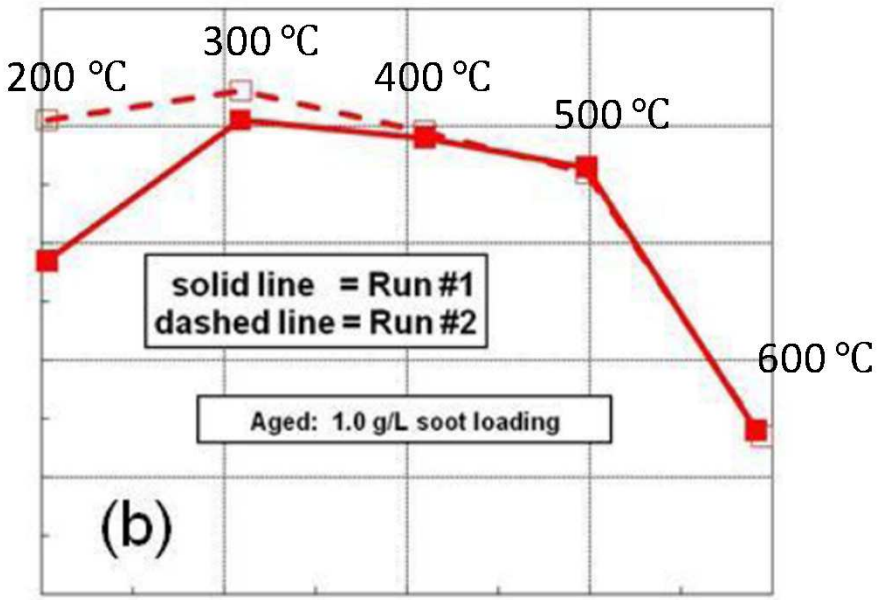
1. Regeneration starts at lower temperature comparing non catalyzed DPF
2. Soot oxidation rate decreases as temperature increases from 400 to 500 degC

Conclusion(I)

1. Catalytic oxidation at the temperature of 200 to 400 degC → soot inside the filter wall

2. Thermal oxidation(above 400 deg) → soot on the filter wall

Review – Experiment(II)



Observation (II)

1. Deterioration of NOx reduction performance
   1) Only occurs at 200 and 300 degC
   2) Can be recovered if soot is burned out.

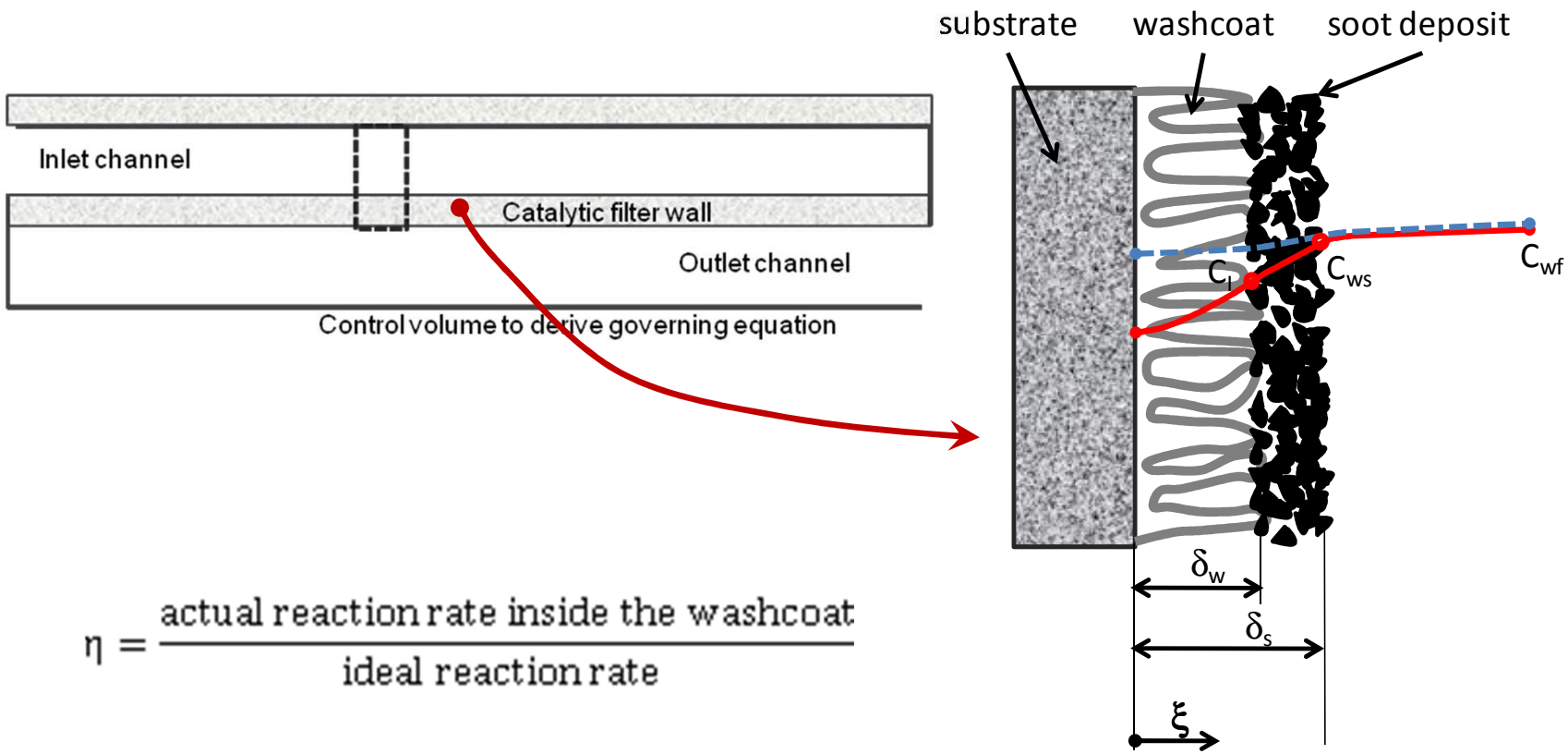
Conclusion (II)

1. At 400 degC or above, there is no soot inside the filter wall because of catalytic oxidation of soot.

2. Soot deposit inside the filter wall disrupts mass transfer from fluid side to catalytic site. → reason for poor NOx conversion efficiency at 200 and 300 degC when soot exists

Model for soot deposit effect on SCR

Idea of Thiele Modulus and Effectiveness Factor can be extended to soot loading case



$$\eta = \frac{\text{actual reaction rate inside the washcoat}}{\text{ideal reaction rate}}$$

Diffusion-Reaction Equation for soot deposited case

For catalyst washcoat

$$0 \le \bar{\xi} \le \delta_w \qquad \frac{\partial^2 \bar{C}}{\partial \bar{\xi}^2} - \Phi^2 \bar{C} = 0$$

$$\text{at } \bar{\xi} = 0, \qquad \frac{\partial \bar{C}}{\partial \bar{\xi}} = 0 \qquad \text{at } \bar{\xi} = 1, \qquad \bar{C} = 1$$

For soot deposit layer

$$\delta_w < \bar{\xi} \le \delta_s \qquad \frac{\partial^2 \bar{C}}{\partial \bar{\xi}^2} = 0$$

$$\text{at } \bar{\xi} = 1, \qquad \bar{C} = 1 \qquad \text{at } \bar{\xi} = \frac{\delta_w}{\delta_s}, \qquad \bar{C} = \frac{C_{ws}}{C_I}$$

where $\bar{C} = C/C_I$ $\quad \bar{\xi} = \xi/\delta_w$ and $\Phi = \sqrt{\frac{k_v \delta_w^{\,2}}{D_{eff,w}}}$ Thiele Modulus

Solution & Effectiveness Factor

Solution

$$\bar{C}(\bar{\xi}) = \frac{\sinh(\Phi_w \bar{\xi})}{\sinh(\Phi_w)} \qquad \text{for } 0 \le \bar{\xi} \le \delta_w$$

$$\bar{C}(\bar{\xi}) = \frac{\delta_c (C_{ws} - C_I)}{C_I(\delta_s - \delta_w)}\bar{\xi} + \frac{C_I \delta_s - C_{ws}\delta_w}{C_I(\delta_s - \delta_w)} \qquad \text{for } \delta_w < \bar{\xi} \le \delta_s$$

where $$C_I = \frac{\dfrac{D_{eff,s}}{(\delta_s - \delta_w)}}{\sqrt{D_{eff,w} k_v}\tanh(\Phi_w) + \dfrac{D_{eff,s}}{(\delta_s - \delta_w)}} C_{ws}$$

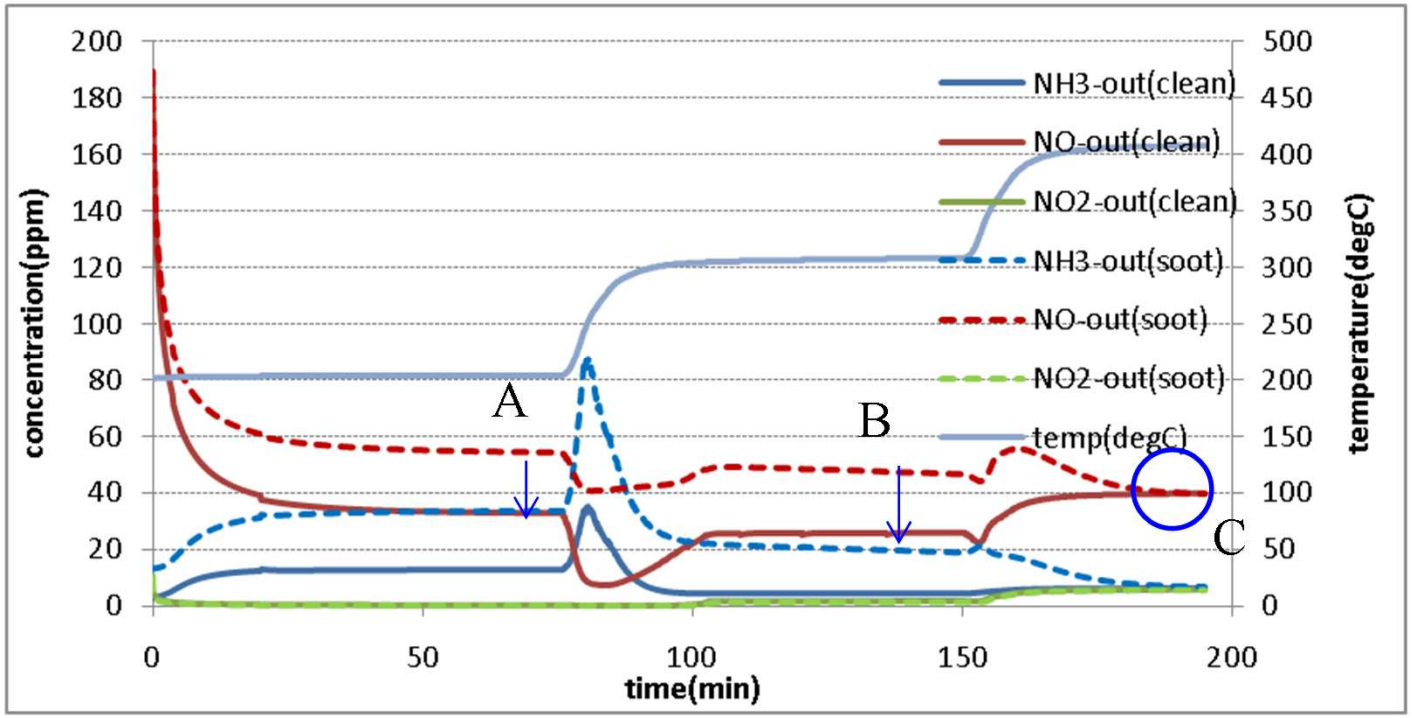
Effectiveness Factor

$$\eta_s = \frac{D_{eff,s}}{k_v \delta_w}\left(\frac{\sqrt{D_{eff,w} k_v}\tanh(\Phi_w)}{(\delta_s - \delta_w)\sqrt{D_{eff,w} k_v}\tanh(\Phi_w) + D_{eff,s}}\right)$$

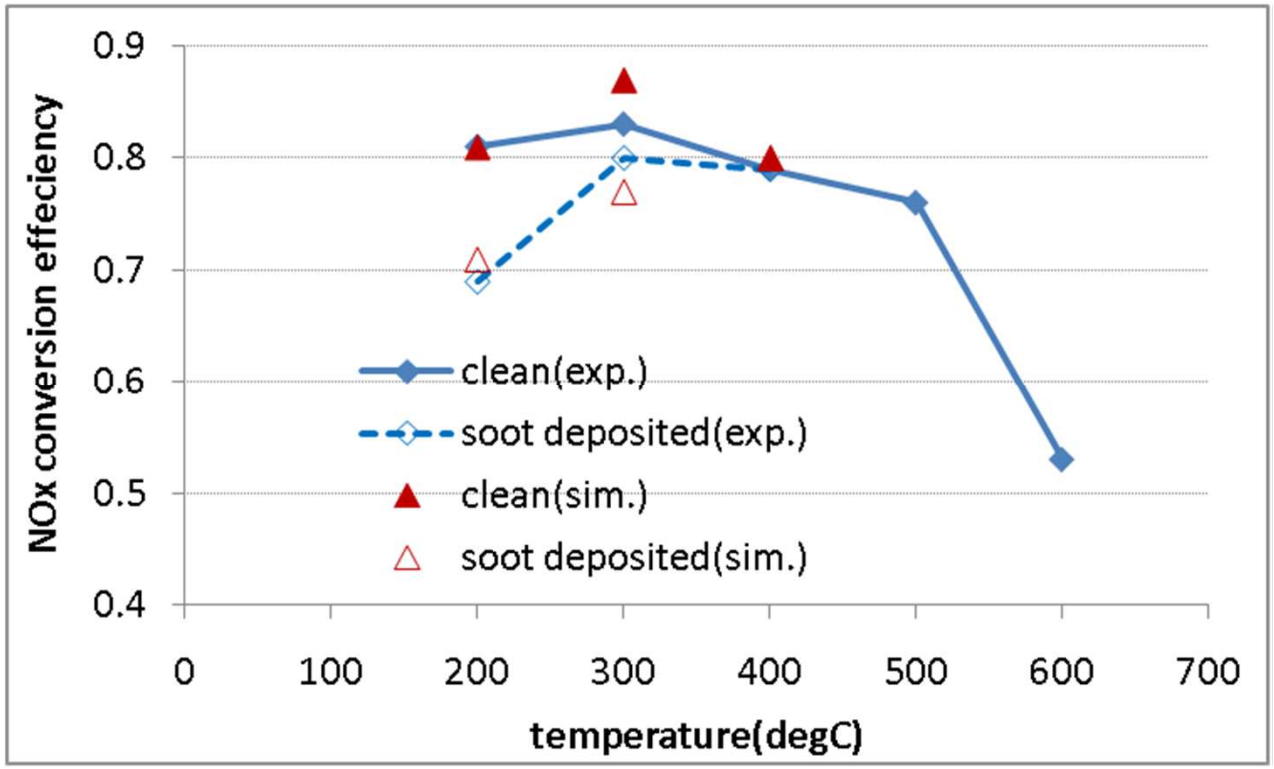
$$\lim_{\delta_s \to \delta_w} \eta_s = \frac{\sqrt{D_{eff,w} k_v}\tanh(\Phi_w)}{k_v \delta_w} = \frac{\tanh(\Phi_w)}{\Phi_w} = \eta_c \qquad k_{app,s} = \eta_s k_v$$

Implementation of Effectiveness Factor

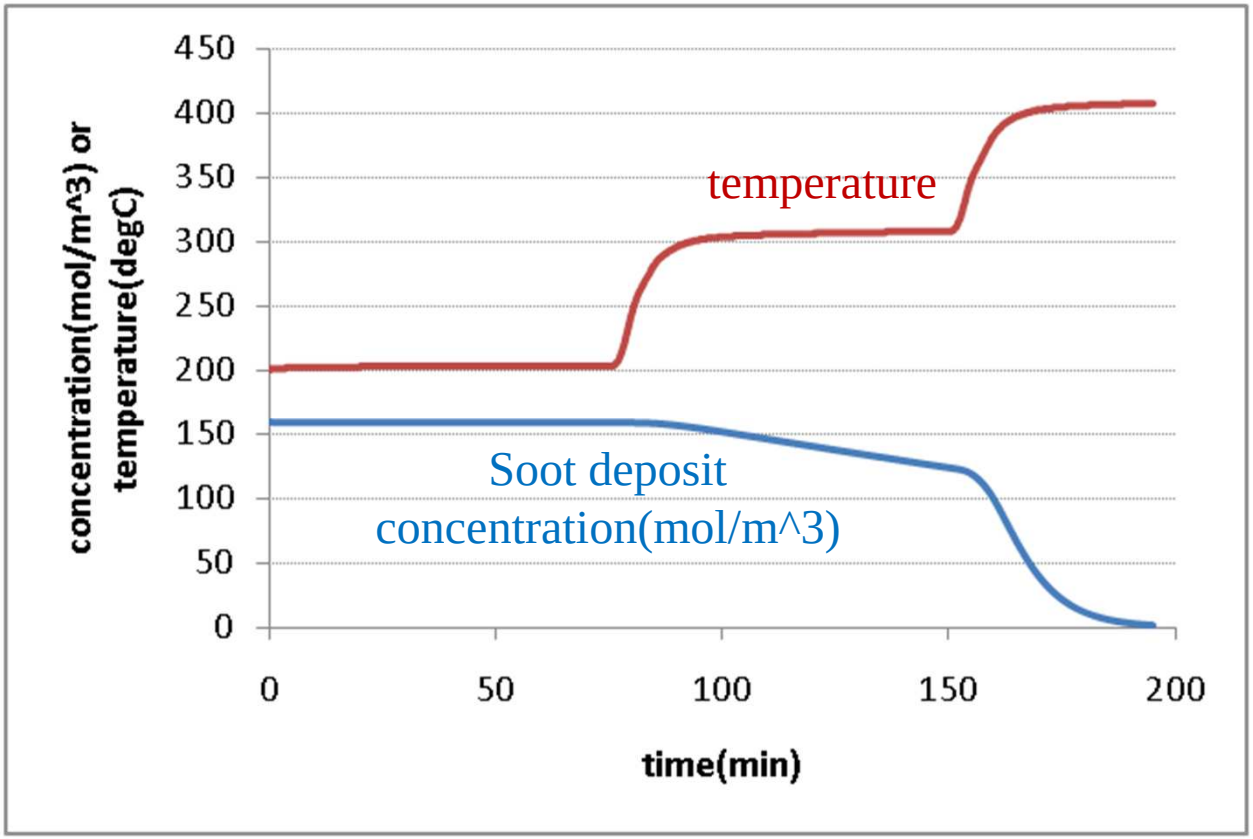
Comparison of clean and soot deposit case : Simulation



Simulation and Experimental Results



Concentration of soot deposit inside the filter wall



Concentration of soot deposit : only inside the filter wall

Conclusion

1. **Introduction of model development for wall flow type SCR**
2. **Model was validated over TPD, steady NOx reduction test case.**
3. **The results are investigated in space and they gives the insight for the utilization of catalyst.**
4. **Ammonia balance monitor → can be applied to control model**
5. **Soot deposit inside the filter wall → Resistance to mass transfer for catalytic reaction → Decreasing overall SCR performance**
6. **Extended Thiele modulus and effectiveness factor → For effect of soot loading on SCR performance**

Acknowledgement

This research work is supported by General Motors as UW-GM Collaborative Research Laboratory.

Experimental Work : Steven Schmieg

Thank You!