A Numerical Investigation of the Performance Analysis of Ammonia Based SCR Aftertreatment Systems

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Emissions Control – Background

- Emissions from engines are major sources of urban air pollution.
- The gasoline engine exhaust gases contain oxides of nitrogen (NOx), carbon monoxide (CO), and partially burned or unburned hydrocarbons (HC).
- The major pollutants from diesel engines are particulate matter (PM) and NO_x.
- These pollutants have hazardous effects on environment and living beings.
- These pollutants are removed from the exhaust gases by employing aftertreatment devices.

Background

- The development of an efficient NO_x reduction technology is essential in spreading the use of diesel engines.
- Since the three-way catalyst has poor NO_x conversion efficiency in the lean environment, it cannot be used in diesel applications.
- Selective Catalytic Reduction (SCR) of NO_x with N-containing agents (ammonia and urea) is a promising technology for controlling NO_x from diesel exhaust.
- Due to difficulties associated with handling and higher toxicity of ammonia, urea is a preferred carrier for ammonia for mobile applications.

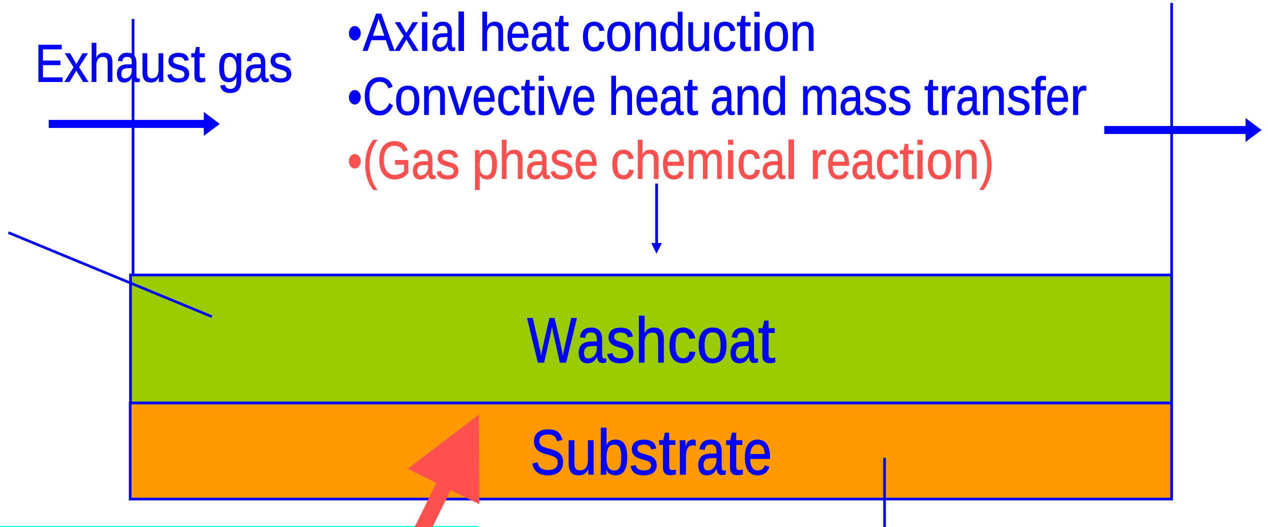
Objectives

- To investigate the physiochemical processes and develop a quantitative predictive understanding of the SCR systems for diesel engines

- To make recommendations for design changes to improve the performance of SCR systems

Physical Formulation

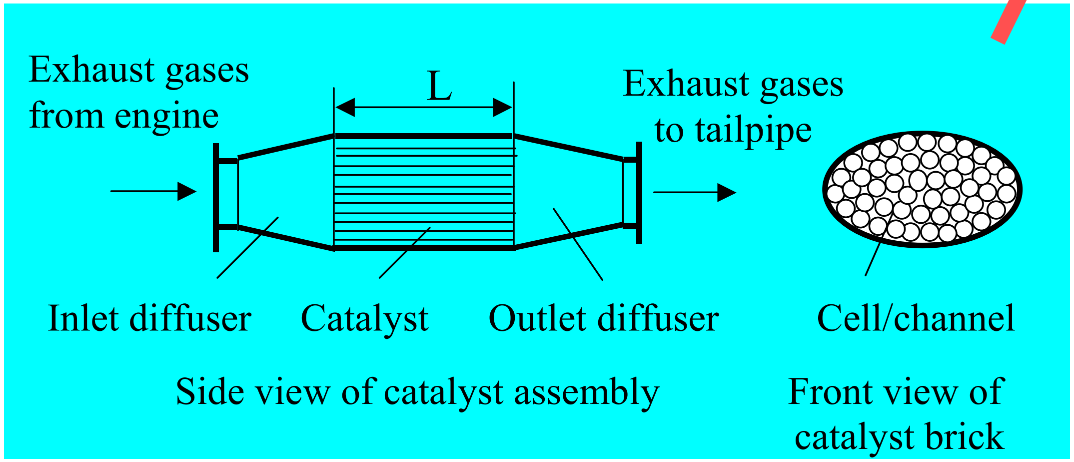
- Heterogeneous chemical reaction
- Heat generation
- Pore diffusion
- Surface adsorption and desorption
- Diffusion
- (Radial conduction)



- Axial heat conduction
- Convective heat and mass transfer
- (Gas phase chemical reaction)

- Axial heat conduction
- Heat losses to ambient
- (Radiation heat losses)

Physical & chemical phenomena in a single channel



Side view of catalyst assembly

Front view of catalyst brick

Governing Equations

- Conservation of energy (gas phase)

$$\rho_g C_{pg}\left(\varepsilon \frac{\partial T_g}{\partial t} + v_g \frac{\partial T_g}{\partial z}\right) = -h_g G_a (T_g - T_s)$$

- Conservation of species (gas phase)

$$\varepsilon \frac{\partial C_g^j}{\partial t} + v_g \frac{\partial C_g^j}{\partial z} = -km^j G_a (C_g^j - C_s^j)$$

- Conservation of energy (solid phase)

$$(1-\varepsilon)\rho_s C_{Ps} \frac{\partial T_s}{\partial t} = (1-\varepsilon)\lambda_s \frac{\partial^2 T_s}{\partial z^2} + h_g G_a (T_g - T_s) - h_\infty S_{ext} (T_s - T_\infty)$$
$$+ G_a \sum_{j=1}^{n_{reaction}} R^j (T_s, C_s^1, \cdots\cdots, C_s^{n_{species}}) \cdot \Delta H^j$$

- Conservation of species (solid phase)

$$(1-\varepsilon)\frac{\partial C_s^j}{\partial t} = km^j G_a (C_g^j - C_s^j) - G_a R^j (T_s, C_s^1, \cdots\cdots, C_c^{N_{species}})$$

Governing Equations

- Accumulation and depletion of NH_3 on the catalyst surface

$$\Omega_j \cdot \frac{\partial \vartheta_j}{\partial t} = R_j$$

- where

Ω is catalyst NH_3 adsorption capacity [mol/m^3]

ϑ is NH_3 surface coverage [-]

ε is void volume fraction [-]

Reaction Kinetics

- NH_3 adsorption-desorption

$$NH_3 \leftrightarrow NH_3^*$$

$$r_{ads} = k_{ads} \cdot C_s^{NH_3} \cdot (1 - \vartheta)$$

$$r_{des} = k_{des}^o \cdot \exp\left[-\frac{E_{des}^o}{\Re \cdot T_s} \cdot (1 - \gamma \cdot \vartheta) \right] \cdot \vartheta$$

Reaction Kinetics

- NH_3 oxidation

$$4NH_3^* + 3O_2 \rightarrow 2N_2 + 6H_2O$$

$$r_{ox} = k_{ox}^{o} \cdot \exp\left(-\frac{E_{ox}}{\Re \cdot T_s} \right) \cdot \left(\frac{p_{O_2}}{0.02} \right)^{\beta} \cdot \vartheta$$

Reaction Kinetics

- Standard SCR reaction

$$4NH_3^* + 4NO + O_2 \rightarrow 4N_2 + 6H_2O$$

$$r_{NO} = k_{NO}^{o} \cdot \exp\left(-\frac{E_{NO}}{\Re \cdot T_s}\right) \cdot \frac{C_s^{NO} \cdot \vartheta}{1 + K_{LH} \cdot \dfrac{\vartheta}{1-\vartheta}} \cdot \left(\frac{p_{O_2}}{0.02}\right)^{\beta}$$

Solution Procedure

- The equations were discretized by using the control volume approach with a non uniform grid, and the central implicit difference scheme for space variable.
- A standard tridiagonal matrix algorithm with a successive line under relaxation method was used to solve the equations.
- The boundary and initial conditions were obtained experimentally.

Validation of Results

- Two experiment types:
  - Temperature programmed desorption
  - SCR performance with NO-NH_3 in the infeed gas

- SCR catalyst geometry
  - Hydrothermally aged catalyst 64 hrs @ 670°C
  - Cell density (cells/inch^2): 400
  - Wall thickness (mil): 6.5
  - Sample diameter (inch): Ø1
  - Length (inch): 1

Simulation of TPD performance

- GHSV (hr^{-1}) = 30,000 and 60,000
- Adsorption Temperatures (°C):
  50, 100, 150, 250, 350
- Desorption Ramp Rate (°C/min): 10
- Infeed NH_3 (PPM): 175, 260, 350
- Infeed O_2, CO_2, H_2O (%): 14, 5, 5
- Measured outlet PPMs: NH_3

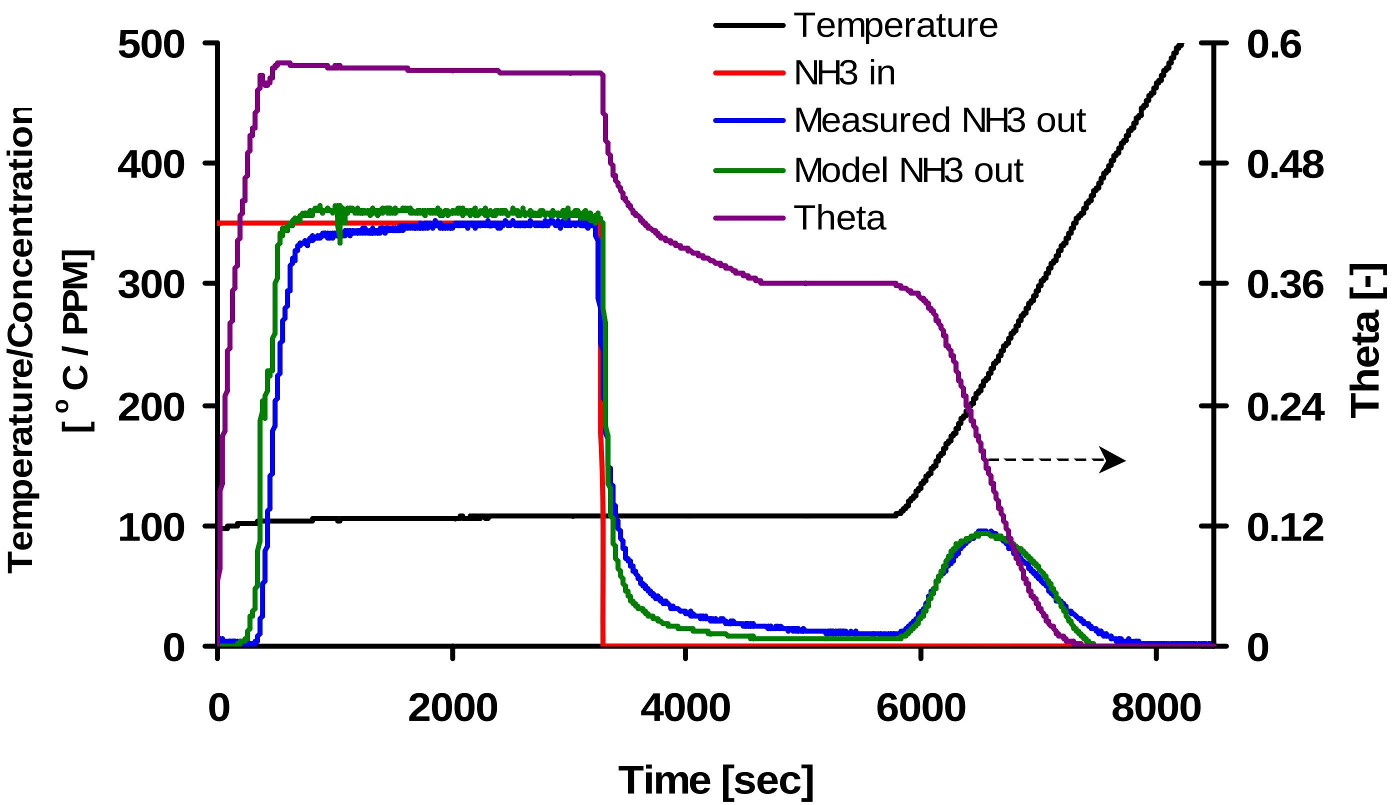
Kinetic Parameters

k_{ads} [1/s]	1.3E03
k^{o}_{des} [mole/m^3/s]	3.9E10
E^{o}_{des} [J/mole]	117E03*0.9
γ [-]	0.51
Ω [mol/m^3]	70

Reference: SAE paper 2005-01-0965

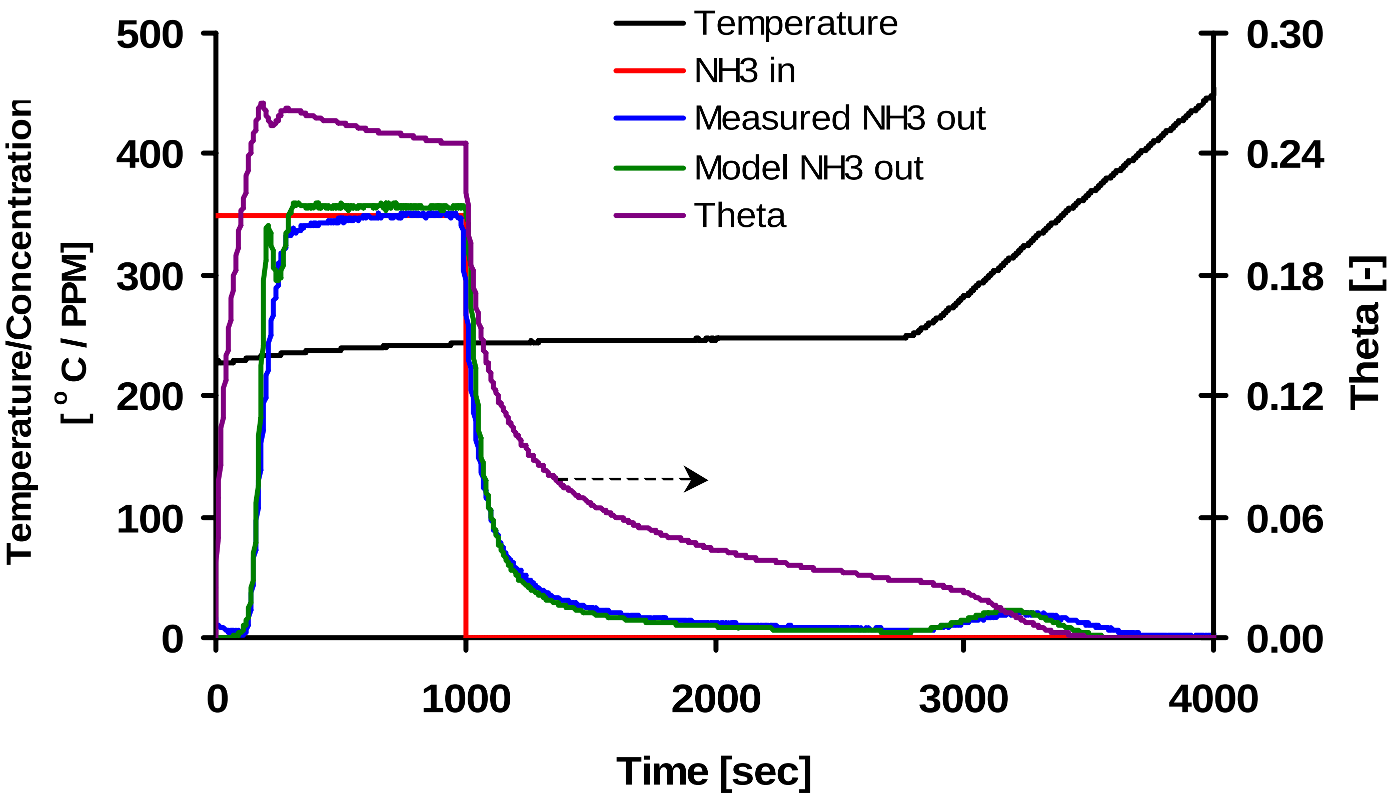
Chatterjee et al.

Simulation of TPD performance



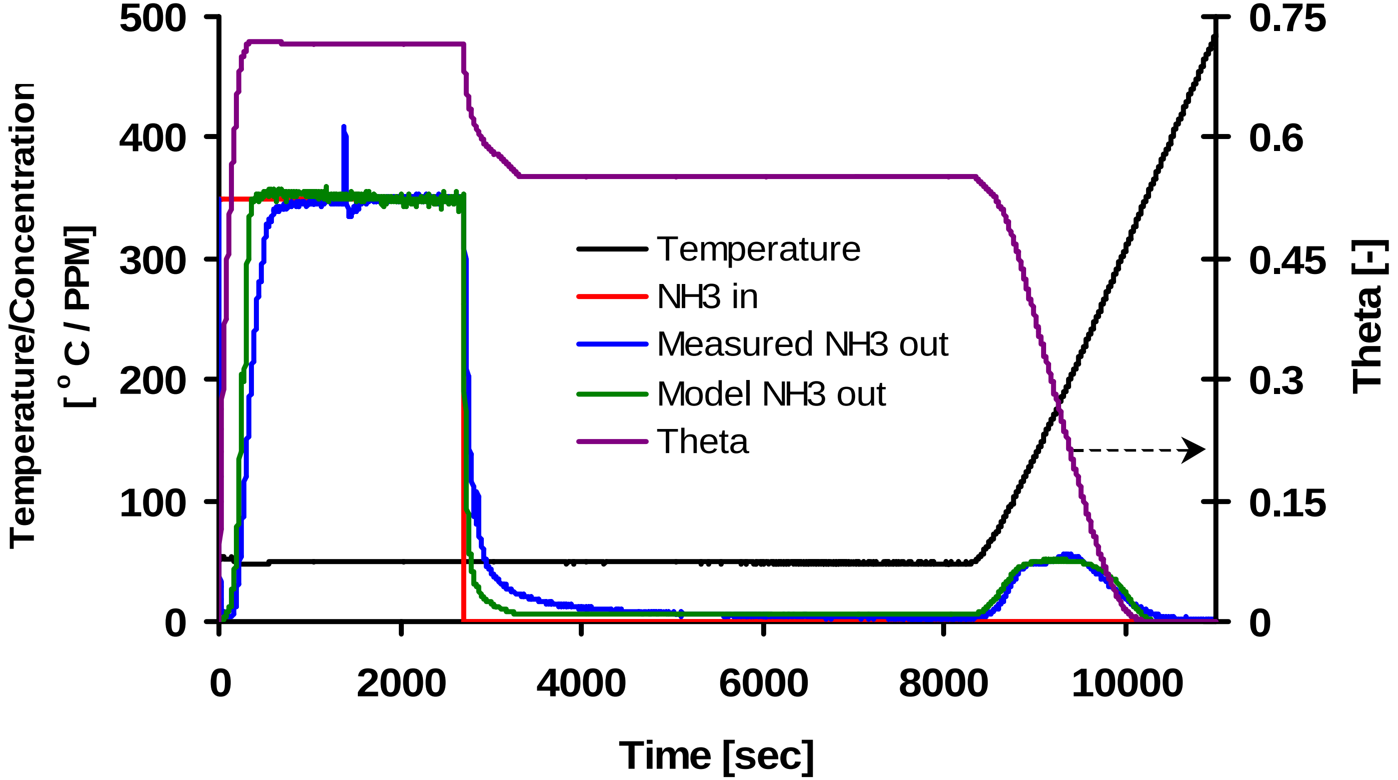
GHSV = 30,000 hr $^{-1}$; Temperature = 100°C; Inlet NH_3 = 350 PPM

Simulation of TPD performance



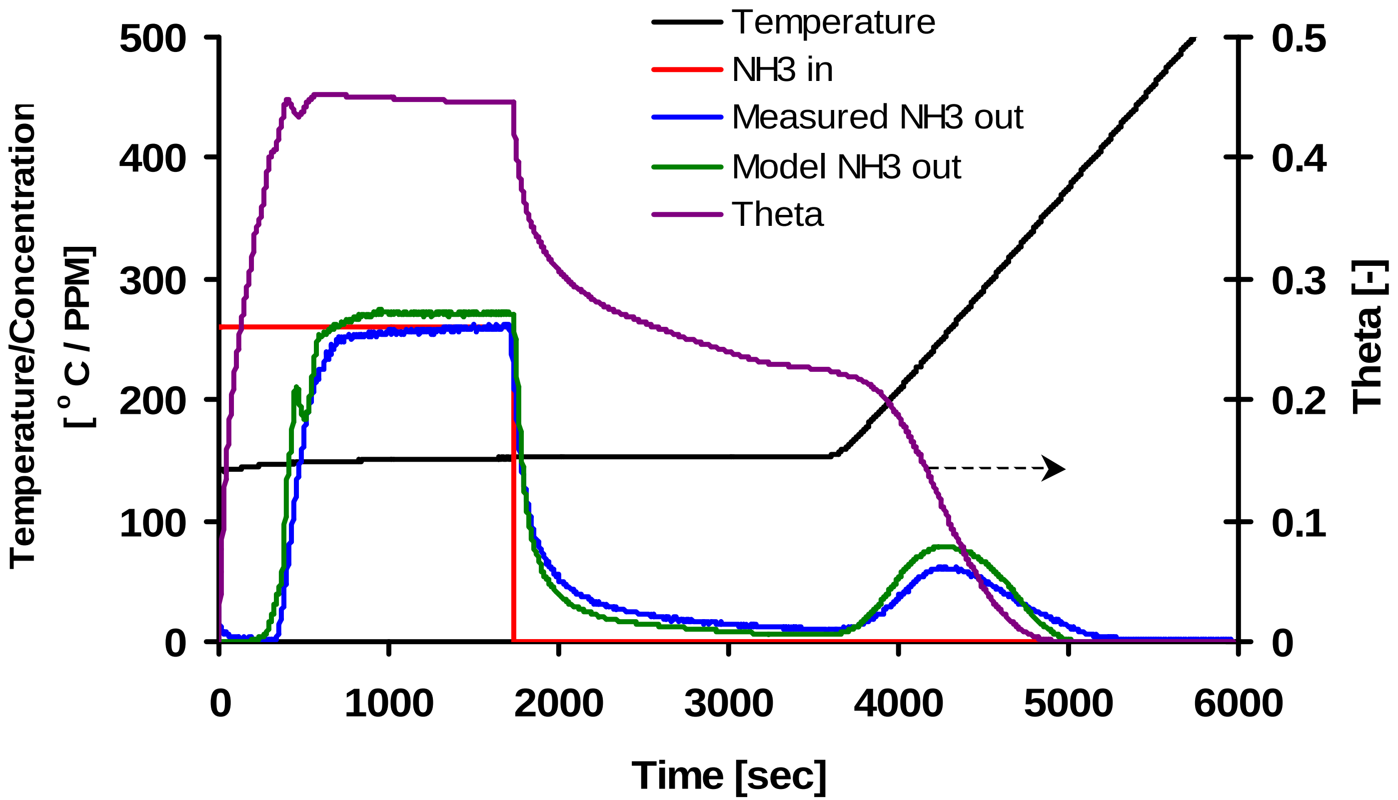
GHSV = 30,000 hr $^{-1}$; Temperature = 250°C; Inlet NH_3 = 350 PPM

Simulation of TPD performance



GHSV = 60,000 hr $^{-1}$; Temperature = 50^{o}C; Inlet NH_3 = 350 PPM

Simulation of TPD Performance



GHSV = 30,000 hr $^{-1}$; Temperature = 150°C; Inlet NH_3 = 260 PPM

Simulation of SCR Performance

- GHSV (hr $^{-1}$) = 30,000, 60,000, and 120,000
- Temperatures (°C):
  150, 200, 250, 300, 350, 400, 450, 500
- Infeed NO, and NH_3 (PPM): 350, 350
- Infeed O_2, CO_2, H_2O (%): 14, 5, 5
- Measured outlet PPMs: NH_3, NO, N_2O, NO_2

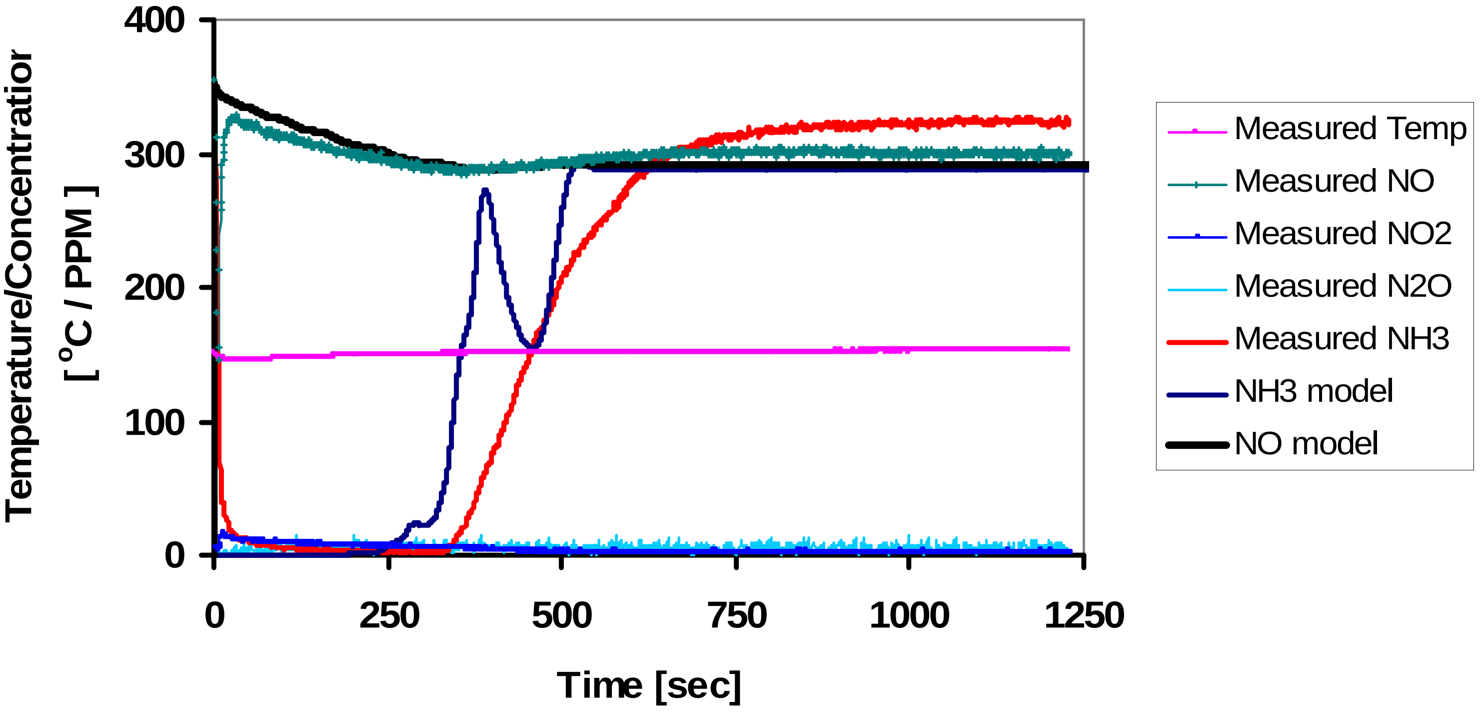
Kinetic Parameters

k^{o}_{ox} [mole/m^3/s]	1.1E09
E_{ox} [J/mole]	118E03
k^{o}_{NO} [1/s]	2.2E08
E_{NO} [J/mole]	55E03*1.039
β [-]	0.27
K_{LH} [-]	8.2

Reference: SAE paper 2005-01-0965

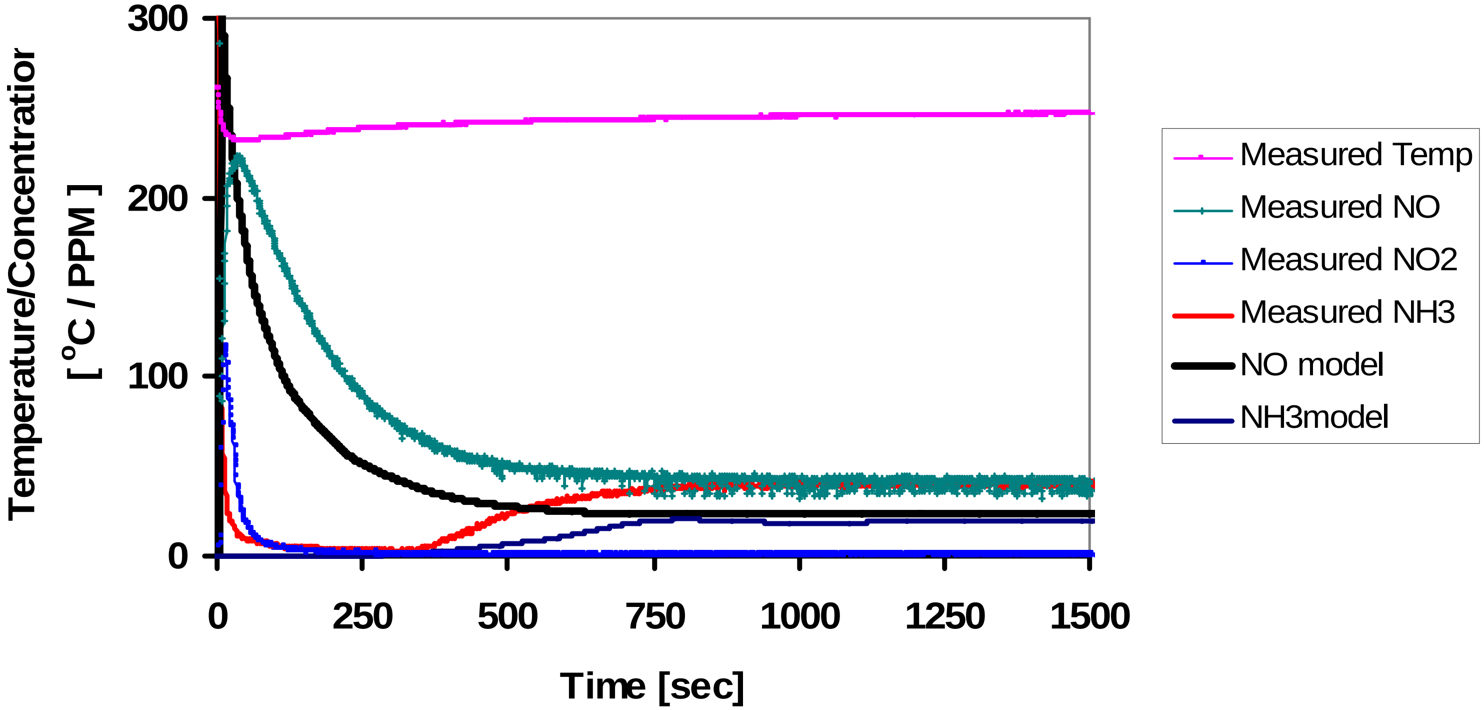
Chatterjee et al.

Simulation of SCR Performance



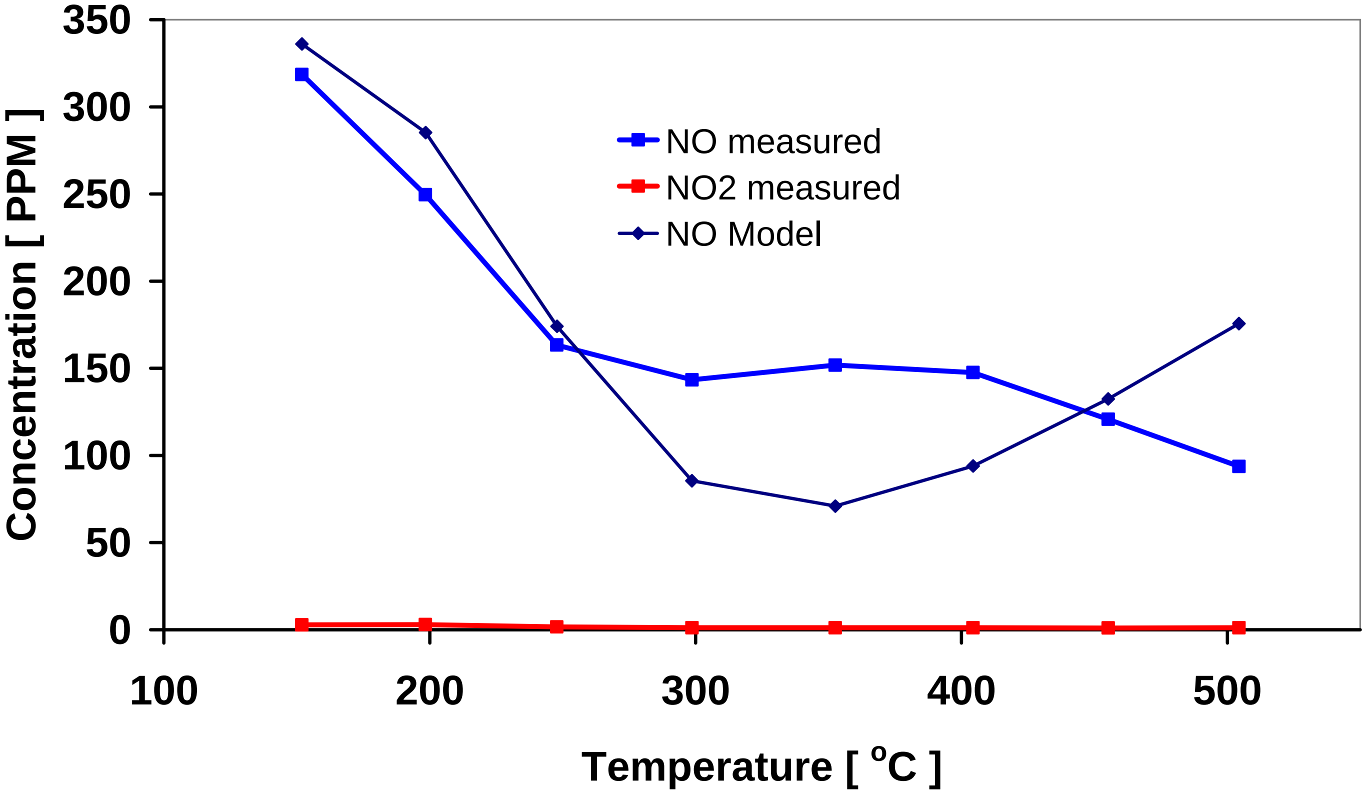
GHSV = 30,000 hr $^{-1}$; Temperature = 150°C; Inlet NH_3 and NO = 350 PPM

Simulation of SCR Performance



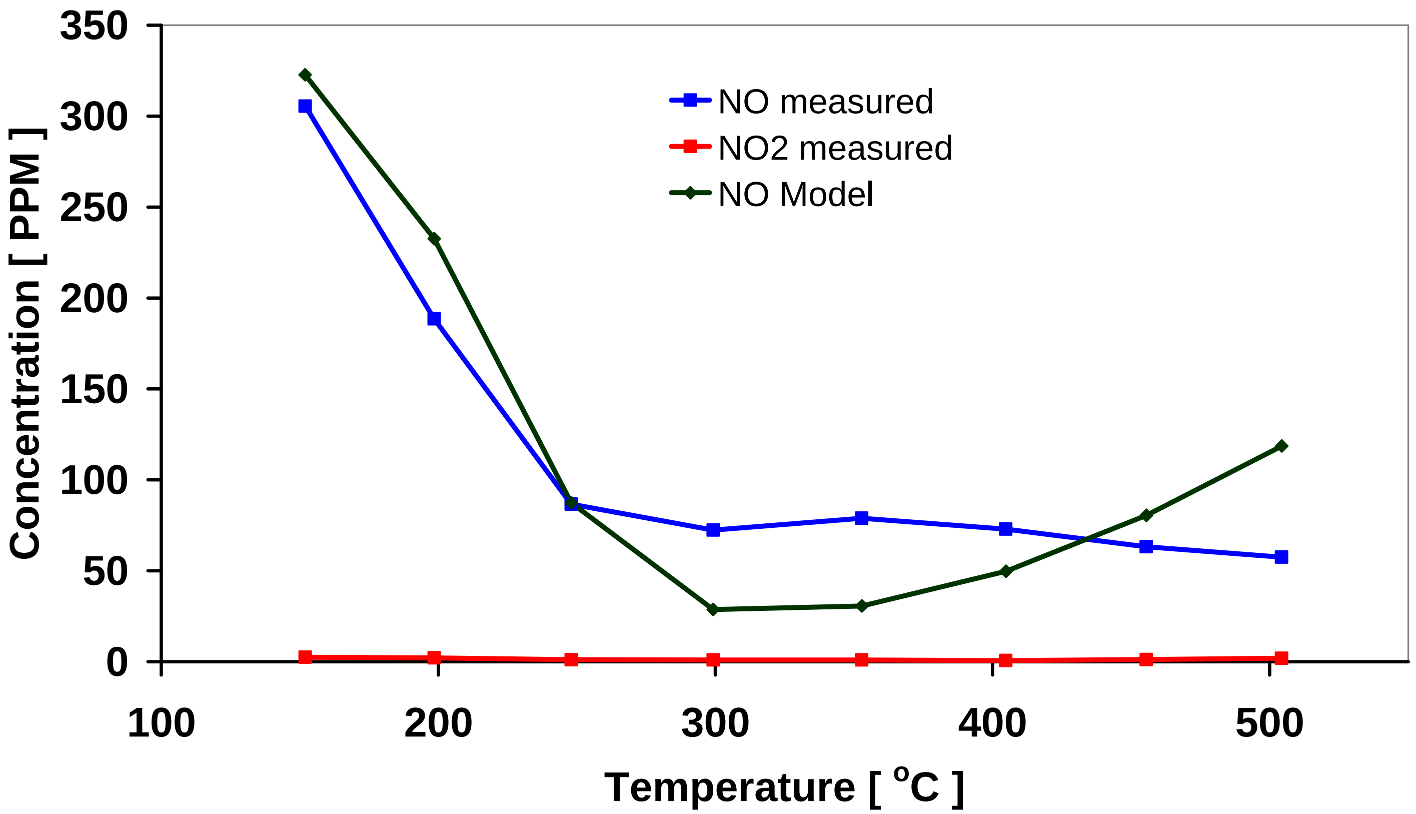
GHSV = 30,000 hr $^{-1}$; Temperature = 250°C; Inlet NH_3 and NO = 350 PPM

Simulation of SCR Performance



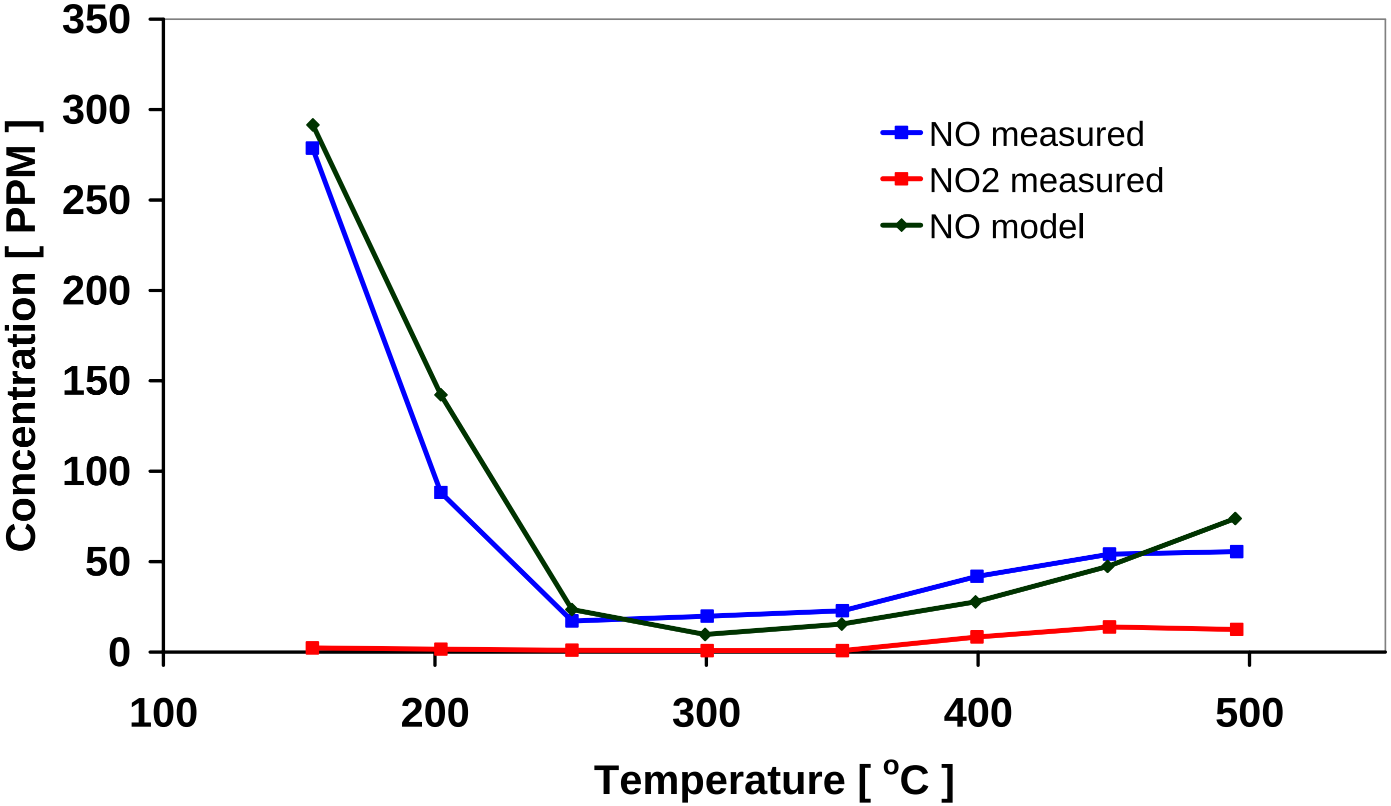
GHSV = 120k hr $^{-1}$; Inlet NH_3 and NO = 350 PPM

Simulation of SCR Performance



GHSV = 60k hr^{-1}; Inlet NH_3 and NO = 350 PPM

Simulation of SCR Performance



GHSV = 30k hr $^{-1}$; Inlet NH_3 and NO = 350 PPM

Conclusions

- Model results for TPD are in good agreement with measurement data
- Present kinetics shows increasing outlet NO PPM with temperature at high temperatures
- Smaller activation energy for NH_3 oxidation results in higher outlet NO PPM
- Decreasing activation energies for SCR reaction increase the conversion efficiency of NO
- At higher GHSV and higher temperatures, the catalyst can adsorb less NH_3
- Fast SCR reactions are being incorporated in the model

End

Thank you for the attention!