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Optimal NH_3 Storage in SCR Catalysts

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Motivation

- Progress in advanced combustion technologies has made the exhaust temperature cooler
- Urea-SCR performance during low temperature operating conditions is a concern in meeting future emission legislation
- Maintaining optimum set levels of NH_3 in the catalyst is critical for higher NO_x control performance during low temperatures
- One approach is to design an optimal urea injection control strategy that minimizes NO_x and NH_3 emissions simultaneously.

Presentation Overview

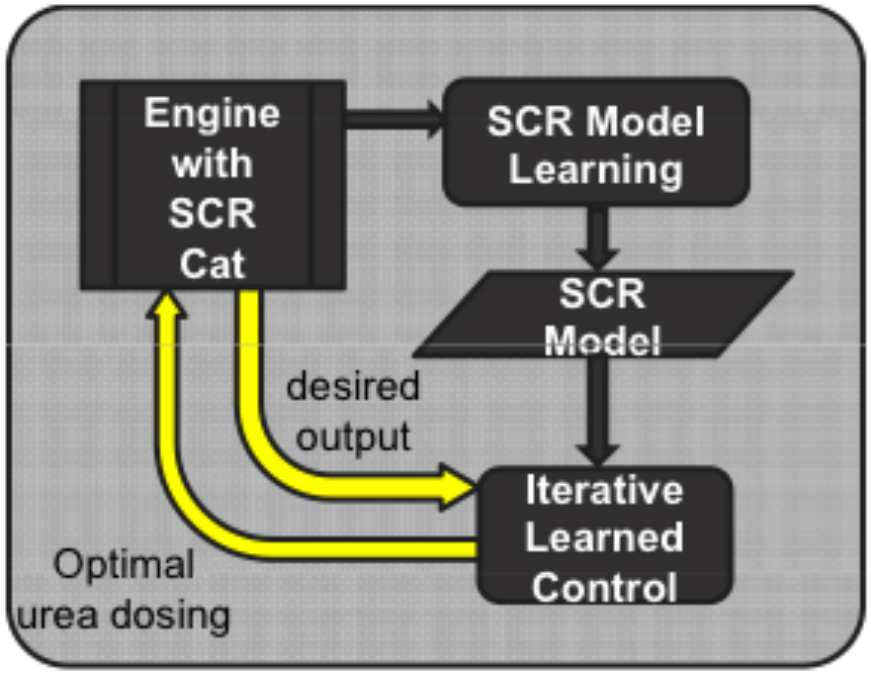
- Data Driven Control Strategies
- ILC Strategy using Hammerstein-Wiener Models
  - Learning H-W Models
  - Iterative Learning Control Strategy
- Model Free Adaptive Control Strategy
- Simulation Results
- Summary & Future Work

Why Data Driven Control Strategies?

- Reducing high fidelity models into accurate lower order models is complex and sometimes an impossible task.
- Uncertainty in model parameters (pre-exponentials, activation energies) can be avoided.
- Data driven control strategies can be easily plugged in with the hardware and offer real time optimal solutions.
- Two data driven control strategies are presented
    - Iterative Learned Control (ILC)
    - Model Free Adaptive Control (MFAC)

Process Flow for ILC Strategy

- Catalyst data measured on the engine is used to train the model.
- Learned model is then integrated with iterative learned control (ILC) strategy, optimal on H-W models, to design an urea injection strategy.



Model Learning for H-W Models

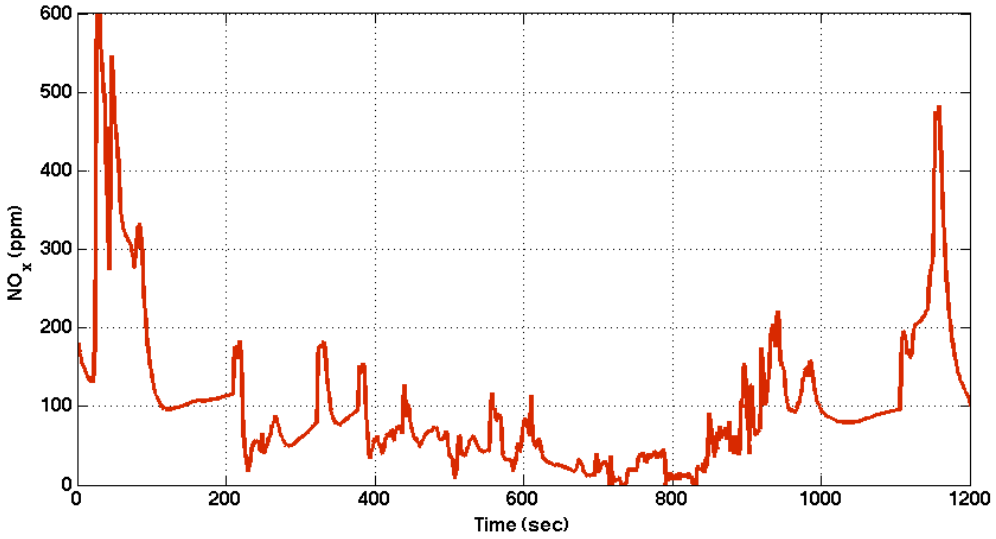
- Multiple input single output (MISO) models are developed for NO_x and NH_3.
- Both models use the following inputs

$$u_1 = c_{NH_3,in}$$
$$u_2 = c_{NO_X,in}$$
$$u_3 = \dot{m}$$
$$u_4 = e^{-1e4/T_{in}}$$

- Identification uses a recursive prediction error method (RPEM) with a restricted black box parametrization[1].
- Algorithm identifies the coefficients of a polynomial in states, inputs and derivative of inputs.

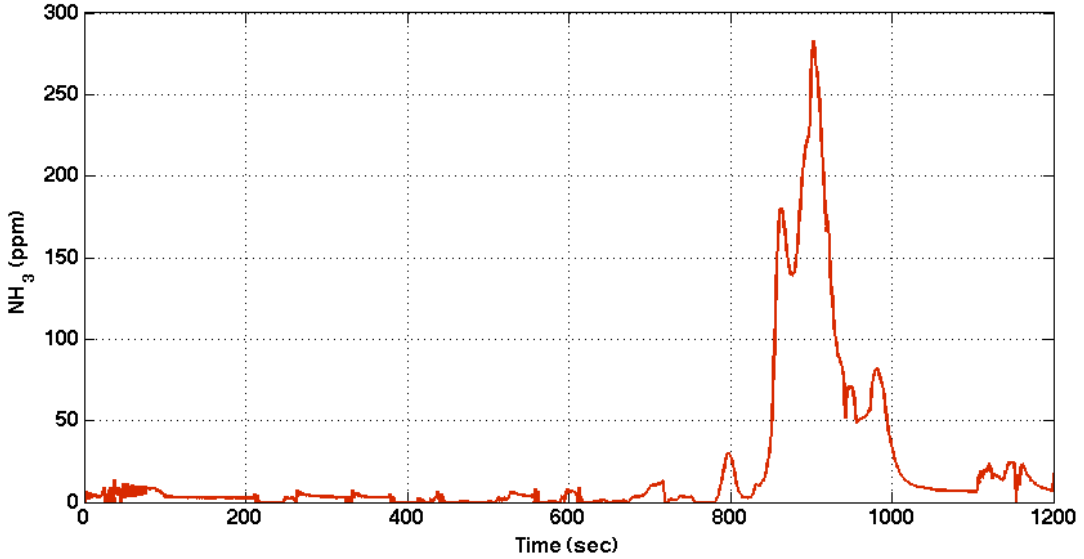
[1] Tayamon, S., and Wigren, T., American Control Conference, 2010

Learned Models Validation



- Learned model fits the measured FTP data well (not shown here)
- Low root mean square error in NO_x and NH_3 models

Need to validate on multiple data sets for more confidence in the model.



Iterative Learned Control Strategy

- ILC guarantees optimality for H-W models.
- Algorithm is simple and well grounded in stochastic control theory

Denote the tracking error by $e_k(t) = y_d(t) - y_k(t)$, where y_d is the desired output and y_k is the output at the k-th iteration. For each t, $u_0(t)$ is arbitrarily chosen.

The control signals at odd steps are given by

$$u_{2k+1}(t) = u_{2k}(t) + c_k \Delta_k(t),$$

The control signals at even steps are given recursively by

$$\bar{u}_{2(k+1)}(t) = u_{2k}(t) - \frac{a_k}{c_k \Delta_k(t)} (|e_{2k+1}(t+1)|^2 - |e_{2k}(t+1)|^2),$$

$$u_{2(k+1)}(t) \cdot I_{\left[|\bar{u}_{2(k+1)}| \le M_{\sigma_k(t)}\right]},$$

$$\sigma_k(t) = \sum_{l=1}^{k-1} I_{\left[|\bar{u}_{2(l+1)}| > M_{\sigma_l(t)}\right]},$$

$$\sigma_0(t) = 0.$$

Model Free Adaptive Control (MFAC)

A data driven control strategy that uses dynamic linearization to linearize a nonlinear system

$$y(k+1) = f(Y(k), U(k))$$

as

$$\Delta y(k+1) = \Phi^T(\mathrm{k})\Delta U(k)$$

where Y and U are matrices containing signals from time k to time $k-L$, $\Phi_{n_u \times L} = [\phi_1 | \phi_2 | \dots | \phi_L]$ is a time-varying pseudo-Jacobi matrix, and n_u is the number of input signals. Starting with the control law

$$J\big(u(k)\big) = \|y^*(k+1) - y(k+1)\|^2 + \lambda \|\Delta U(k)\|^2$$

we derive an update rule for $u(k)$,

$$u(k) = u(k-1) + \frac{\phi_1(k)}{\lambda + \|\phi_1(k)\|^2}\left[\rho_1\big(y^*(k+1) - y(k)\big) - \sum_{i=2}^{L} \rho_i \phi_i \Delta u(k-i+1)\right],$$

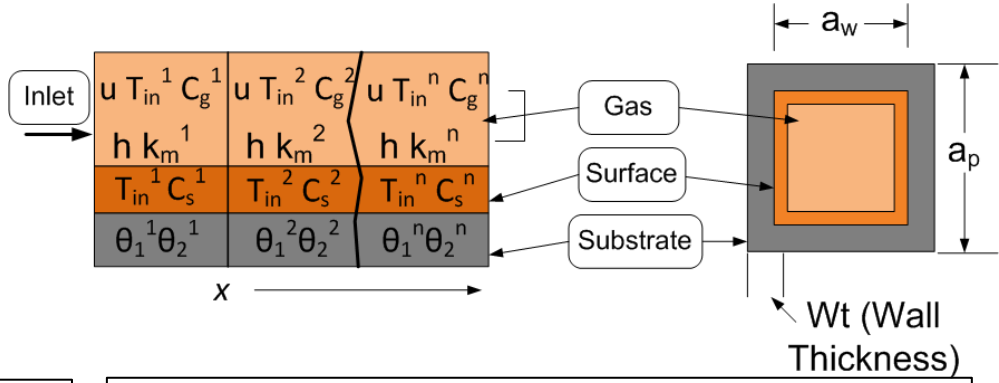
Energy criterion is used to find the update rule for Φ

$$J\big(\Phi(k)\big) = \|\Delta y(k) - \Phi^T(\mathrm{k})\Delta U(k-1)\|^2 + \mu \|\Delta \Phi(k)\|^2$$

Overview of MTU's SCR Model

MTU's SCR model was run using the MFAC strategy and the results are compared.

- 1D single channel model with gas and surface phase
- 2 NH_3 storage sites
- All reactions take place of the surface of the catalyst



Chemical Reactions in the Model

$$NH_3 + S_i \rightarrow NH_{3,i}^*$$ — NH_3 Adsorption

$$NH_{3,i}^* \rightarrow S_i + NH_3$$ — NH_3 Desorption

$$i = Site1, Site2$$

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

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

$$4NH_3^* + 2NO + 2NO_2 \rightarrow 4N_2 + 6H_2O$$ — Fast SCR

$$4NH_3^* + 3NO_2 \rightarrow \frac{7}{2}N_2 + 6H_2O$$ — Slow SCR

$$6NH_3^* + 8NO_2 \rightarrow 7N_2O + 9H_2O$$ — N_2O Formation

Gas and Surface Phase Species Balance

$$\varepsilon \frac{\partial C_{g,i}}{\partial t} = -u \frac{\partial C_{g,i}}{\partial x} - \beta_i A_g (C_{g,i} - C_{s,i})$$

$$(1-\varepsilon)\frac{\partial C_{s,i}}{\partial t} = \beta_i A_g (C_{g,i} - C_{s,i}) - \sum_j n_{i,j} R_j$$

$$i = NH_3, NO, NO_2, N_2O \,\&$$

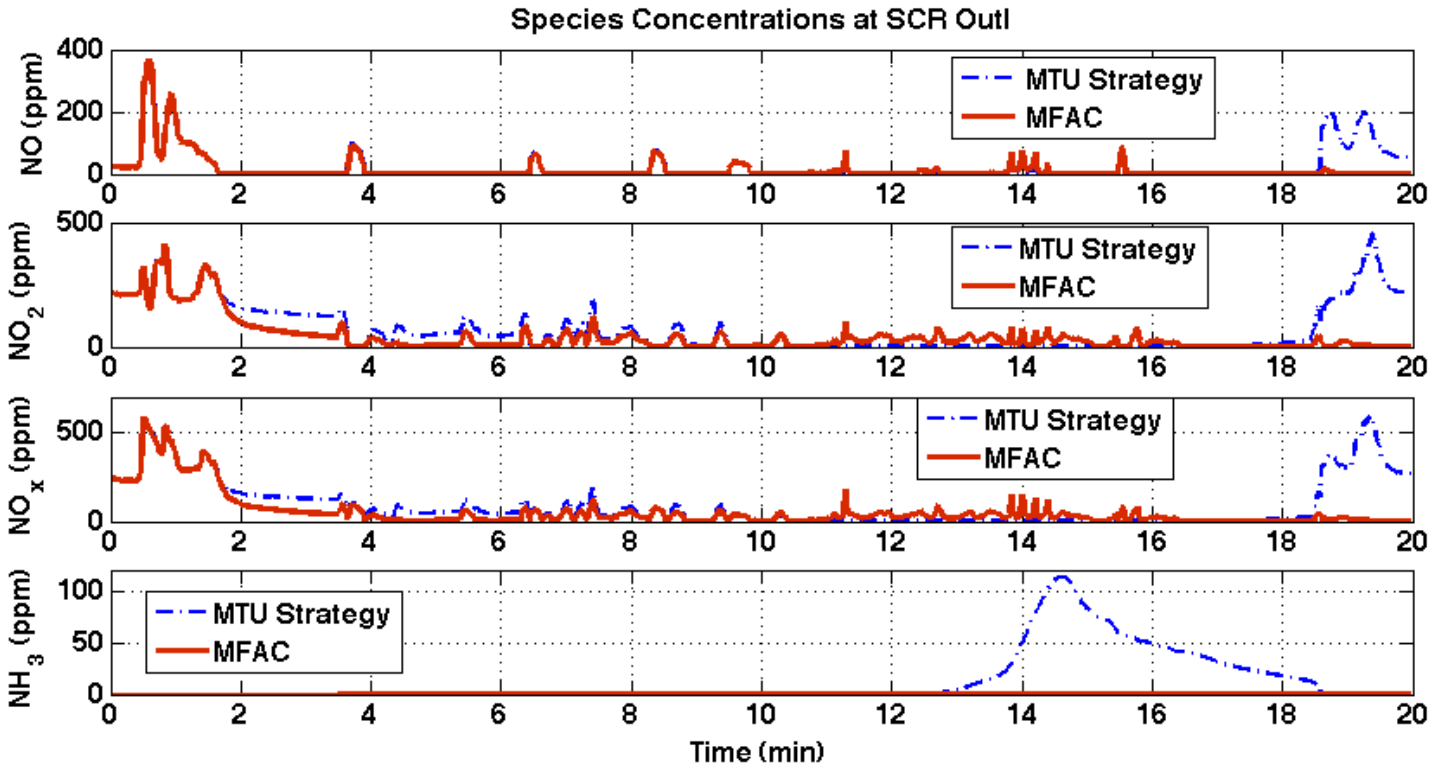
$$j = Ads, Des, std, Fst, slo, oxi$$

NH3 Storage Equations For Site 1 and 2

$$\Omega_1 \dot{\theta}_1 = R_{Ads,1} - R_{Des,1} - 4R_{Oxi} - 4R_{std} - 4R_{Fst} - 4R_{slo}$$

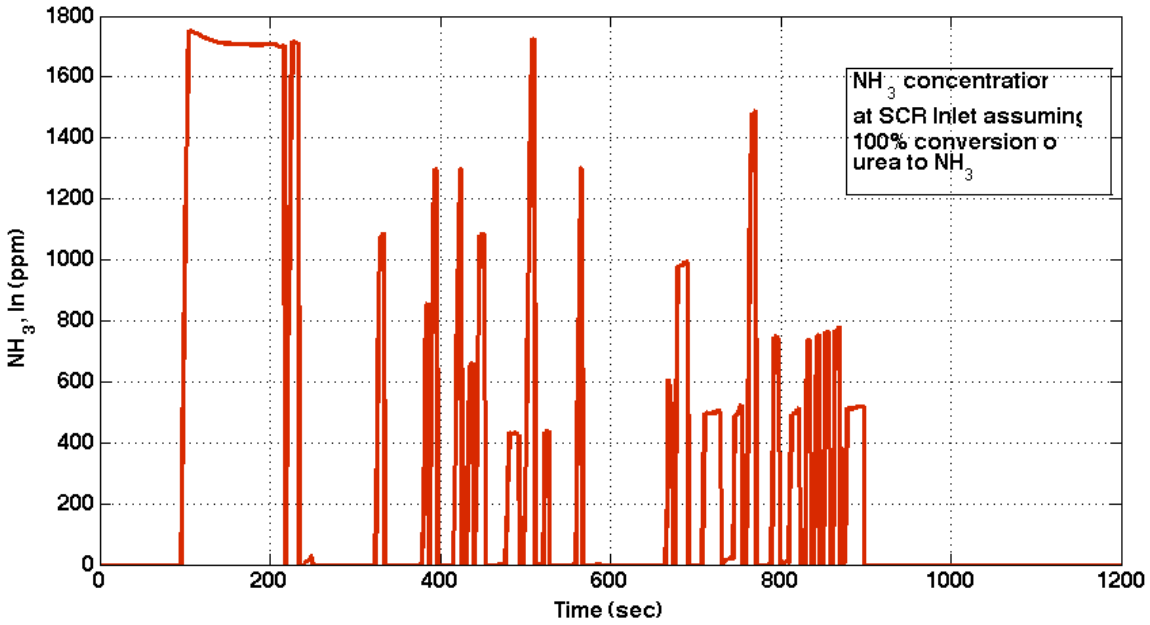
$$\Omega_2 \dot{\theta}_2 = R_{Ads,2} - R_{Des,2}$$

MFAC strategy results in greater NO_x reduction while eliminating NH_3 slip



- Benefits in overall NO_x reduction and NH_3 slip (completely eliminated in the catalyst)

MFAC strategy also results in lesser cumulative urea usage



- Cumulative urea injection is 4% lesser than the existing strategy
- Cumulative SCR out NO_x is reduced by 30% using this strategy.
- Further efforts to improve the control performance (urea injection rate) are underway.

Summary and Future Work

- Optimal controls literature was reviewed and data driven control strategies were chosen for optimal NH3 storage task
- ILC Strategy
  - H-W models for NO_x and NH_3 emissions were validated using FTP data
- MFAC Strategy
  - Benefits in overall NO_x control and urea consumption
  - NH_3 slip was completely eliminated

Future Work

- Further validation of H-W models and integrating with ILC strategy
- Comparison of ILC and MFAC strategies in NO_x and NH_3 control and urea usage.

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Modeling Aging Effects on Reaction Pathways in Cu-CHA Urea SCR Catalysts

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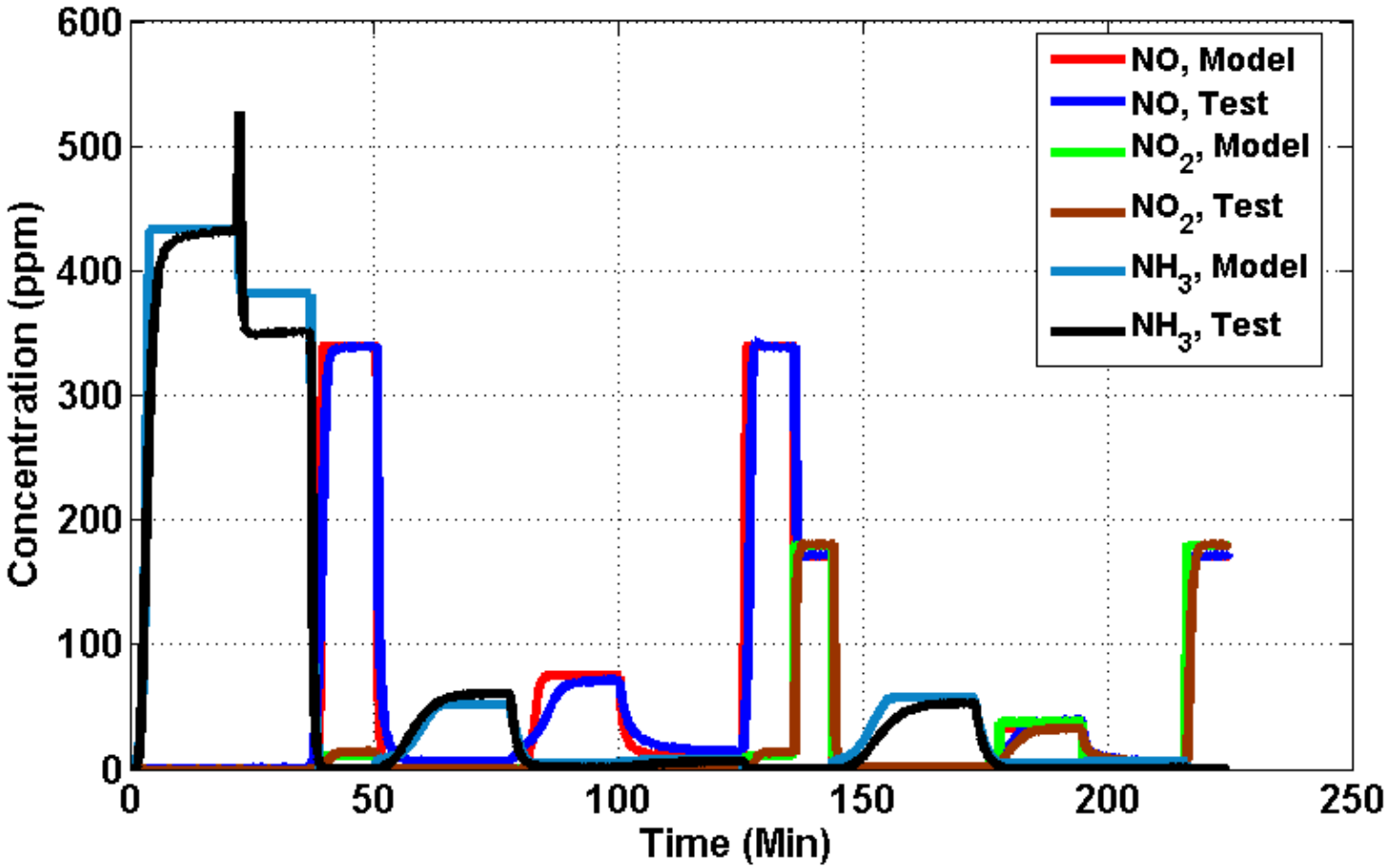
Motivation

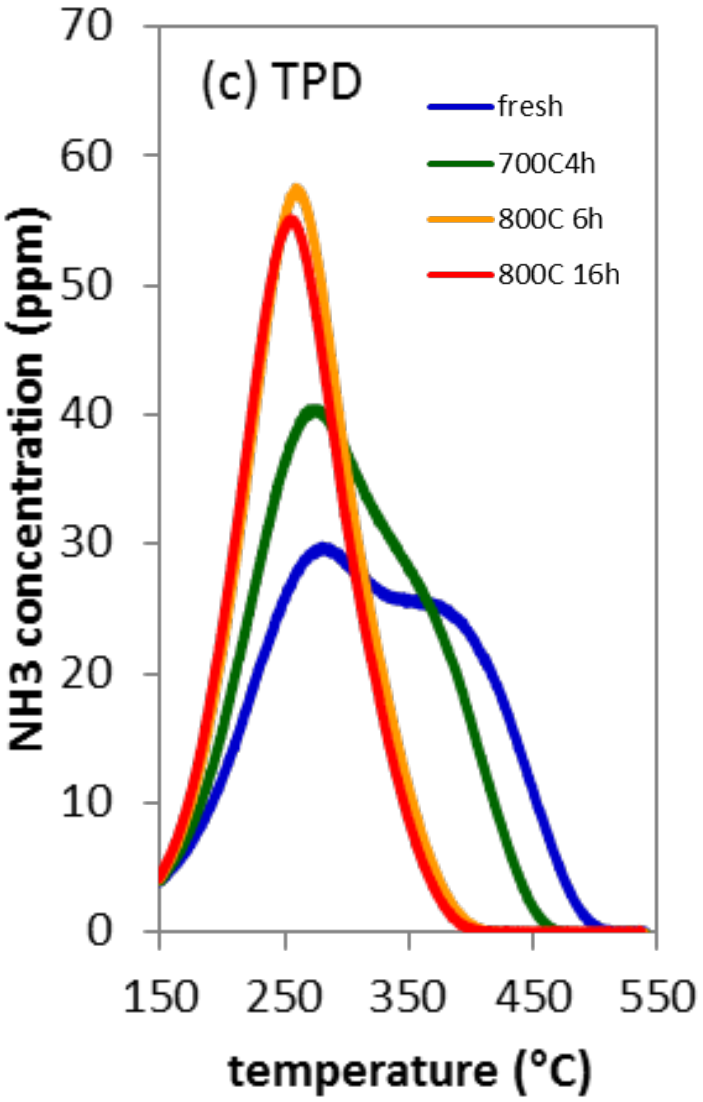
- Overall goal is to develop catalyst aging factors, essential for model based control adaptation, using 1D SCR models.
- Transient protocol and TPD data collected on Cu-CHA samples at ORNL were used to develop the SCR model.

SCR model considering a single NH_3 storage site was developed and validated



- SCR Model validation shown at 90k SV at T = 300°C
- Model was successfully validated for various cases: $0.8 \leq NH_3/NOx \leq 1.2$ and $30k \leq SV \leq 90k$

Recent TPD data on a fresh Cu-CHA sample showed more than one storage site



- Recent data on a fresh catalyst sample (NH_3 desorption vs temperature during TPD shown on the left) shows two peaks indicating the possibility of more than one active site with different stabilities in the catalyst.
- The two peaks convolute into one as the sample is degreened and aged as shown in the figure.
- This has motivated us to develop a model with two NH_3 storage sites so that the aging effect on NH_3 storage and other reaction pathways can be accurately predicted.

Dual Site NH_3 Storage Model

Site 1 (Weakly Adsorbed)

$$r_{ads,s1} = A_{ads,s1} c_{g,NH_3} (1 - \theta_{NH_3,s1})$$

$$r_{des,s1} = A_{des,s1} e^{\frac{-E_{des,s1}(1-\gamma\theta_{NH_3,s1})}{RT}} \theta_{NH_3,s1}$$

Site 2 (Strongly Adsorbed)

$$r_{ads,s2} = A_{ads,s2} c_{g,NH_3} (1 - \theta_{NH_3,s2})$$

$$r_{des,s2} = A_{des,s2} e^{\frac{-E_{des,s2}}{RT}} \theta_{NH_3,s2}$$

$$\frac{\partial c_{g,NH_3}}{\partial t} = -\frac{u}{\varepsilon}\frac{\partial c_{g,NH_3}}{\partial x} + \frac{\Omega_1}{\varepsilon}(r_{des,s1} - r_{ads,s1}) + \frac{\Omega_2}{\varepsilon}(r_{des,s2} - r_{ads,s2})$$

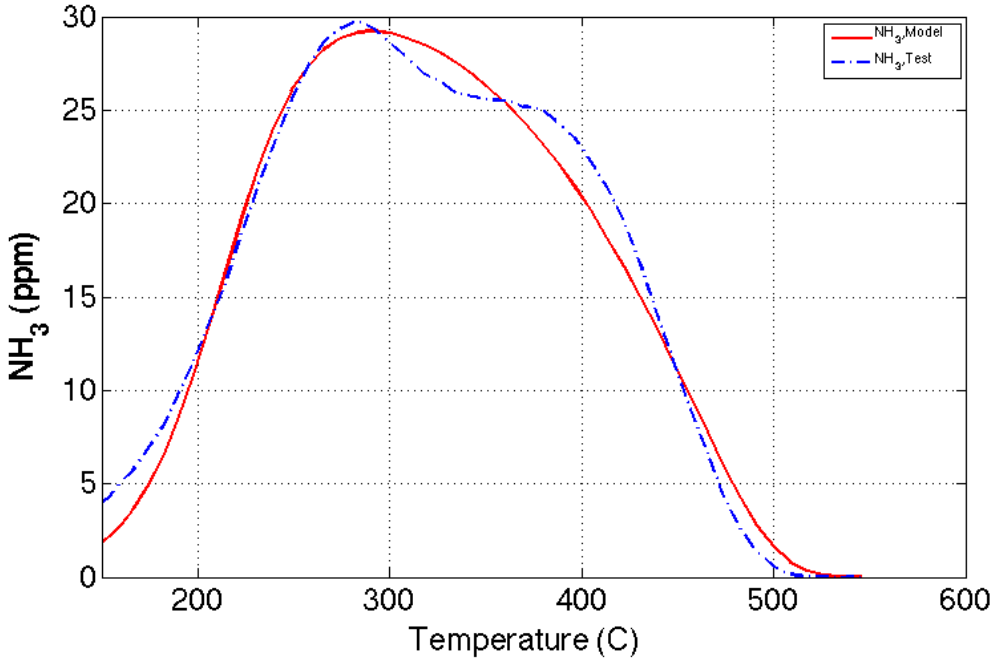
$$\frac{d\theta_{NH_3,s1}}{dt} = r_{ads,s1} - r_{des,s1}$$

$$\frac{d\theta_{NH_3,s2}}{dt} = r_{ads,s2} - r_{des,s2}$$

Rate equations are taken from Colombo et al.'s recent modeling work on Fe-Z catalyst

[1]Colombo et al., Catalysis Today(2011). doi: 10.1016/j.cattod.2011.09.002

Current dual site NH_3 storage model does not match the desorption peaks



- Number of storage sites is critical to predict aging effect on each of the reaction pathways in the SCR catalyst and to identify if 'a' site participates.
- Should more NH_3 storage sites be considered to predict this effect, as reported by Skarlis et al. (Journal of Physical Chemistry C, 2012)?
- How to estimate the activation energies of each of the sites?
- Will the multi-site model be suitable for controls adaptation during aging?