

Model-Based Estimation and Control System Development in a Urea-SCR Aftertreatment System

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ABSTRACT

In this paper, a model-based linear estimator and a non-linear control law for an Fe-zeolite urea-selective catalytic reduction (SCR) catalyst for heavy duty diesel engine applications is presented. The novel aspect of this work is that the relevant species, NO, NO₂ and NH₃ are estimated and controlled independently. The ability to target NH₃ slip is important not only to minimize urea consumption, but also to reduce this unregulated emission. Being able to discriminate between NO and NO₂ is important for two reasons. First, recent Fe-zeolite catalyst studies suggest that NO_x reduction is highly favored by the NO₂ based reactions. Second, NO₂ is more toxic than NO to both the environment and human health. The estimator and control law are based on a 4-state model of the urea-SCR plant. A linearized version of the model is used for state estimation while the full nonlinear model is used for control design. An experimentally validated, higher order simulation is used to evaluate the performance of the closed loop system. For the cases considered, the control strategy uses less urea, produces less NH₃ slip, and less tailpipe NO_x than a similar strategy where NO and NO₂ are assumed as all NO during estimation and control law implementation.

INTRODUCTION

Turbocharged diesel engines operate in lean conditions at A/F ratios greater than 25 and produce significant amounts of NO and NO₂. Exhaust aftertreatment is required to meet the 2010 US EPA diesel emission standards in NO_x and particulate matter (PM). PM standards of 0.01 gm/bhp-hr can be met using existing filter technologies [1] while a variety of NO_x aftertreatment technologies are being investigated to meet the NO_x emission requirement of 0.2 gm/bhp-hr. Out of these technologies,

urea-SCR is a potential candidate to meet the emission standards. It is a proven technology in Europe in meeting Euro IV heavy duty NO_x standards [2]. A high level illustration of a urea-SCR aftertreatment system is shown in Figure 1. Urea solution spray is injected into the ex-

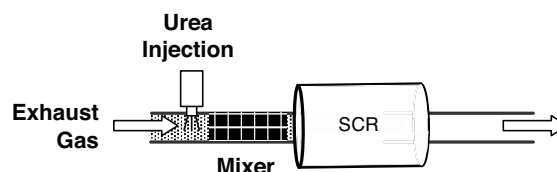


Figure 1: High-level illustration of the urea-SCR aftertreatment system.

haust gas upstream of the SCR catalyst. At sufficiently high exhaust gas temperature, the urea droplets evaporate and mix with the exhaust gas. NH₃ is formed as a result of urea decomposition and H₂NCO hydrolysis reactions in the exhaust pipe and in the SCR catalyst. NO_x is reduced to N₂ due to several SCR reactions aided by the catalyst. The urea-SCR catalyst needs to be actively controlled to ensure high NO_x reduction, low NH₃ slip and low urea consumption. The majority of SCR controllers in the literature are conservatively tuned, map-based open-loop controllers as described in reference [3]. As the authors in references [4] and [5] point out, achieving high NO_x reduction and low NH₃ slip will require model based control strategies incorporating a closed-loop controller.

Several model-based closed-loop controllers are discussed in references [2]-[3] and [5]-[6] which are designed assuming all NO_x quantity as NO in the catalyst. However, it is well known that the reaction rates change based on the NO and NO₂ concentrations entering the catalyst. Additionally, high rates of reactions involving NO₂ on a

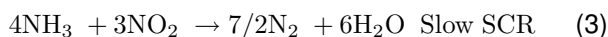
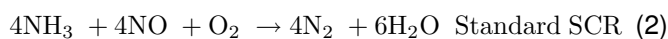
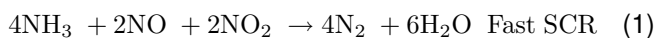
Fe-zeolite catalyst motivates the need for a control strategy that considers the chemistry of NO and NO₂. This paper focuses on the development of a model based estimator and a closed-loop control strategy that estimates and exploits NO and NO₂ states individually.

The remainder of the paper is organized as follows. Two reduced order models which are used for model based estimation and closed-loop control system design are discussed. One model uses an approach of assuming all NO_x as NO and the closed-loop strategy based on this model is compared to the closed-loop strategy based on a model containing NO and NO₂ individually. Parameter identification and model validation is discussed in the next section, followed by estimator and control system design. Simulation results demonstrating the control system performance are discussed and then conclusions are presented.

REDUCED ORDER MODELS

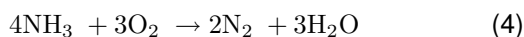
In model-based control system design, the controller should be implementable in real-time. Therefore, there is a need for reduced order models which must capture the essential phenomena and maintain the accuracy. The first model, referred to as '3 state model', follows the reference [7].

A higher order model with detailed modeling methodology and major assumptions pertaining to both the higher order and reduced models is discussed in [8] and is not discussed here for compactness. The three main SCR reactions used in the model are the 'fast' SCR, the 'standard' SCR and NO₂ based SCR ('slow' SCR) respectively shown in Eq. 1 to Eq. 3:



The authors in references [9] and [10] report that the reaction rate of the NO₂ based SCR reaction shown in Eq. 3 has a reaction rate equivalent to the fast SCR reaction rate and greater than standard SCR reaction rate at high temperatures. This is a unique feature of Fe-zeolite catalysts and is not observed on vanadium catalysts.

Only NH₃ oxidation to N₂ is used in the model given by Eq. 4.



NH₃ oxidation to NO is not considered as the literature [11] and [12] suggests that the direct oxidation of NH₃ to N₂ is more relevant than the oxidation of NH₃ to NO on a Fe-zeolite catalyst formulation which is used in this work. The NH₃ oxidation reaction to N₂ is not too temperature dependent like the NH₃ oxidation to NO which is fast at high temperatures on a vanadium catalyst [13].

Fe-zeolites also exhibit distinct NO oxidation capability to NO₂ as reported in references [12] and [10] which is also a

unique feature of zeolites. This reaction is also neglected in the model as NO₂, even if produced by NO oxidation reaction, is reduced by NH₃ through the SCR reactions.

NH₃ adsorption and desorption reactions are the reactions included in the models. The adsorption of NH₃ onto the active sites of the catalyst from the gas phase is given by Eq. 5.



Here S is the active free sites available in the catalyst and NH_3^* is the adsorbed NH₃ site in the catalyst. The desorption of NH₃ from the active sites is given by Eq. 6.



3 State Model

Upadhyay et al. in references [6] and [7] propose a 3 state control-oriented lumped parameter model that contains the gas phase concentrations of NO_x and NH₃ and ammonia storage as the states and it is shown in Eq. 7. The model has been modified to account for 100% selectivity of NH₃ oxidation to N₂. Four reactions shown in Eq. 2 and Eq. 4-Eq. 6 are considered in the model. The following are the assumptions for the 3 state model in addition to the major assumptions discussed in [8]:

1. Mass transfer is neglected. That means the chemical kinetics in the catalyst are reaction controlled.
2. Only the standard SCR reaction is considered.
3. Surface phase concentrations of the species are neglected.
4. All NO_x in the exhaust gas is assumed to be NO.
5. Reaction rates are assumed to be a function of the gas phase concentration of NO_x and ammonia storage.

The reaction rate constants k_i s of the reactions in Eq. 7 correspond to the reactions shown in Eq. 1 to Eq. 6 respectively.

$$\begin{aligned} \dot{C}_{\text{NO}_x} &= -\bar{Q}C_{\text{NO}_x} - k_2\Omega\theta C_{\text{NO}_x} + \bar{Q}C_{\text{NO}_x,in} \\ \dot{\theta} &= -(k_6 + k_4)\theta + k_5C_{\text{NH}_3} - k_1C_{\text{NO}_x}\theta \\ &\quad - k_5\theta C_{\text{NH}_3} \\ \dot{C}_{\text{NH}_3} &= k_6\Omega\theta - (k_5\Omega + \bar{Q})C_{\text{NH}_3} + k_5\Omega\theta C_{\text{NH}_3} \\ &\quad + \bar{Q}C_{\text{NH}_3,in} \end{aligned} \quad (7)$$

Here $\bar{Q}$ is the normalized flow rate given by Eq. 8 in $\frac{1}{\text{sec}}$.

$$\bar{Q} = \frac{Q}{V} \quad (8)$$

Q is the volumetric flow rate in actual $\frac{m^3}{\text{sec}}$ and V_e is the volume of the exhaust gas in the substrate channel in m^3 given by Eq. 9.

ϵ is the open frontal area, A_c is the converter frontal area in m^2 . L_{cat} is the length of the catalyst in m . A total of 9 parameters need to be identified in this reduced order model which include the pre-exponential factors and activation energies of the 4 reactions and the total NH_3 adsorption capacity, Ω .

4 State Model

Incorporating NO and NO_2 individually into the model enables the tracking of NO_2 slip from the tail pipe which is a major concern to many environmental agencies such as CARB and MSHA and a recent reference [14] shows the concern about NO_2 levels increasing in the atmosphere while NO is decreasing. Also, improving the control system performance by modeling NO and NO_2 individually is the motivation behind the development of the 4 state model. The assumptions in the 4 state model in addition to the major assumptions discussed in [8]:

1. Mass transfer is neglected in the model meaning that the chemical kinetics in the catalyst are reaction controlled.
2. Surface phase concentrations of the species are neglected.
3. Reaction rates are defined as a function of ammonia storage (θ) and gas phase concentrations of NO and NO_2 .

The 4 state model contains the gas phase concentrations of NO, NO_2 and NH_3 and ammonia storage as the states shown in Eq. 10. All the 6 reactions considered in the higher order model discussed in [8] are considered in the 4 state model.

$$\begin{aligned}\dot{C}_{NO} &= -\bar{Q}C_{NO} - k_1\Omega\theta C_{NO}C_{NO_2} - k_2\Omega\theta C_{NO}n_{O_2} \\ &\quad + \bar{Q}C_{NO,in} \\ \dot{C}_{NO_2} &= -\bar{Q}C_{NO_2} - k_1\Omega\theta C_{NO}C_{NO_2} - k_3\Omega\theta C_{NO_2} \\ &\quad + \bar{Q}C_{NO_2,in} \\ \dot{\theta} &= -(k_6 + k_4)\theta + k_5C_{NH_3} - k_1C_{NO}C_{NO_2}\theta \\ &\quad - k_2C_{NO}n_{O_2}\theta - k_3C_{NO_2}\theta - k_5\theta C_{NH_3} \\ \dot{C}_{NH_3} &= k_6\Omega\theta - (k_5\Omega + \bar{Q})C_{NH_3} + k_5\Omega\theta C_{NH_3} \\ &\quad + \bar{Q}C_{NH_3,in}\end{aligned}\quad (10)$$

A total of 13 parameters need to be identified in this reduced order model which include the pre-exponential factors and activation energies of the 6 reactions and the total NH_3 adsorption capacity.

TEST DATA AND MODEL VALIDATION

This section discusses the parameter identification and model validation approach followed for a 4 state model based on the test data. The adequacy of the 4 state model for model-based control system design is briefly

discussed. For detailed procedure on parameter identification and adequacy of the reduced order models, the reader is referred to reference [8].

Experiments were conducted on a Navistar I6 7.6L engine. The experimental set-up with a Horiba emission bench and an MKS FTIR analyzer at locations denoted as α and β is shown in Figure 2. Tests were designed to

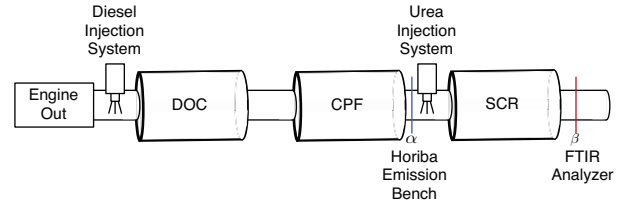


Figure 2: Schematic of the aftertreatment test set-up used for experimental testing.

facilitate parameter identification and validate the models. The urea flow rate was manually set to cover a wide range of NH_3/NO_x ratios in the test. The experiments were conducted at various steady state engine operating points. The step changes from point to point are used to capture the transient effects of temperature and mass flow rates on catalyst dynamics. An $m - T$ curve for one such test at different temperatures and flow rates is shown in Figure 3. The various speed and load conditions in the test are shown in Figure 4. The input concentration profiles of the species at the inlet of the SCR catalyst are shown in Figure 5. The various speed-load conditions at which the experiment is conducted are denoted by A_1, B_1 , etc in the figures. NH_3 concentration at the inlet of the catalyst is not measured and is calculated from the urea injection flow rate as shown in Eq. 11. It is based on the assumption that one mole of urea forms two moles of NH_3 and is available for NO_x conversion in the catalyst.

$$C_{NH_3} = 2.0 * \frac{\dot{m}_{urea}}{\dot{m}_{exh}} \frac{MW_{exh}}{MW_{urea}} * 1E6 \quad (11)$$

Here, C_{NH_3} is the concentration of NH_3 in ppm. $\dot{m}_{urea}$ is the mass flow rate of urea in kg/sec , MW_{urea} is the molecular weight of urea in $gram/mole$ ($MW_{urea} = 60 gram/mole$) and MW_{exh} is the molecular weight of exhaust gas in $gram/mole$ ($MW_{exh} = 28.8 gram/mole$)

The parameter identification exercise for both models is formulated as an optimization problem and Matlab's simplex method based optimization function $fminsearch$ is used to identify the parameters while minimizing the cost function in the species concentrations. The optimization problem is defined as:

Find the model parameters (x_i) where x_i are the pre-exponential factors and activation energies of the reactions, which minimize the cost function in Eq. 12.

$$J = \frac{1}{N} \sum_{i=1}^N (C_{i,s} - C_{i,m})^2 \quad (12)$$

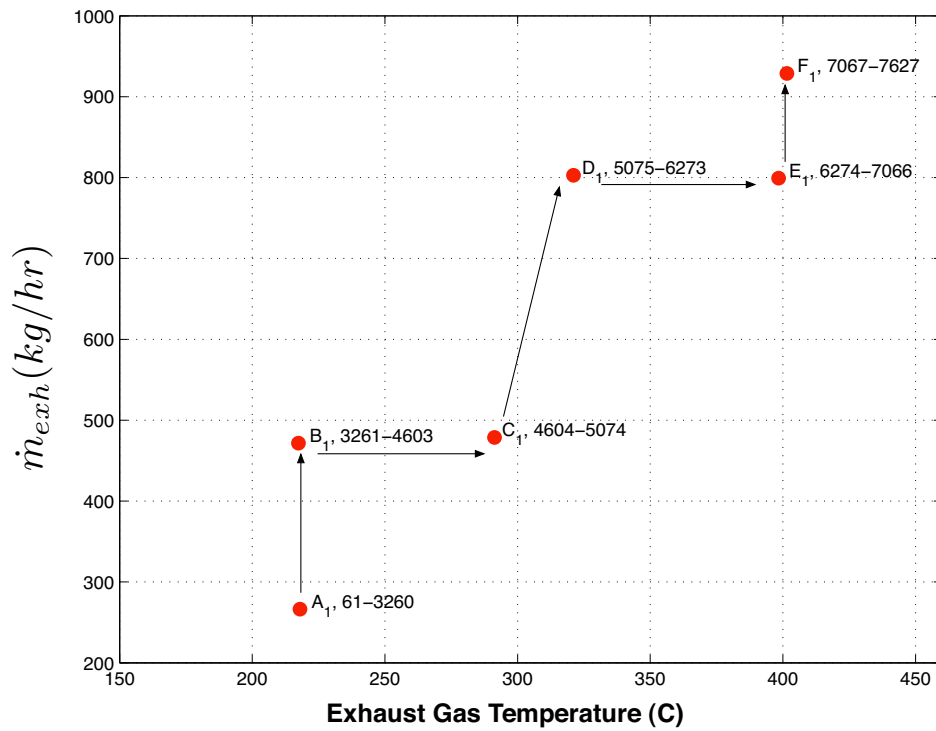


Figure 3: $\dot{m}$ -T Curve for the test. The numbers next to the data points indicate the time for which the test is run at that point in seconds. The arrows indicate the sequence of step changes in operating conditions.

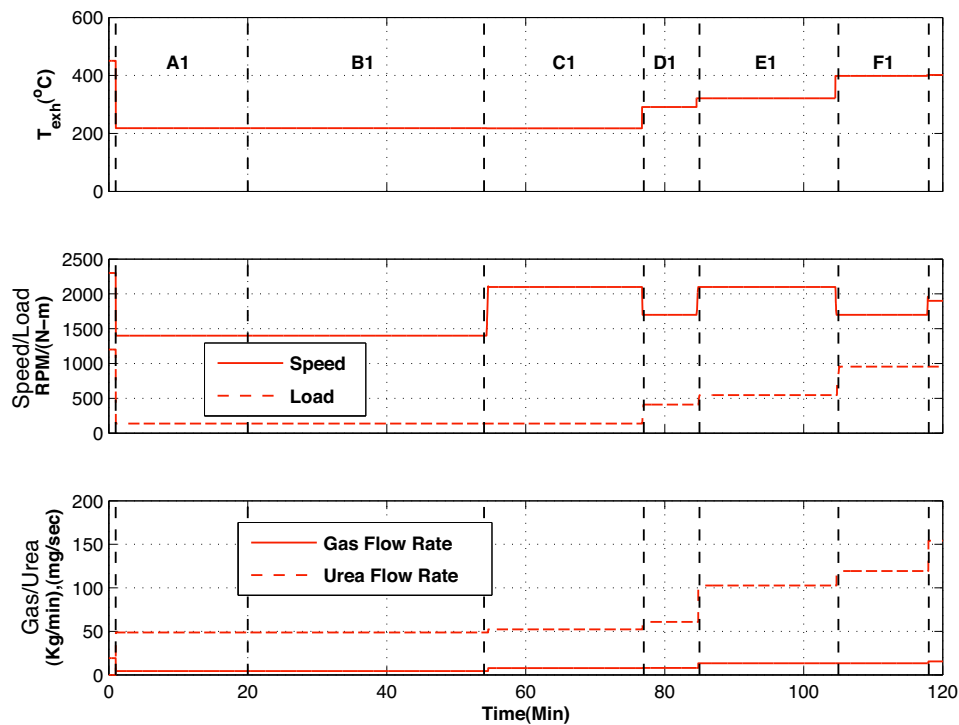


Figure 4: Input profiles of exhaust gas temperature, urea injection flow rate, speed, load and mass flow rate at various engine operating conditions during the test.

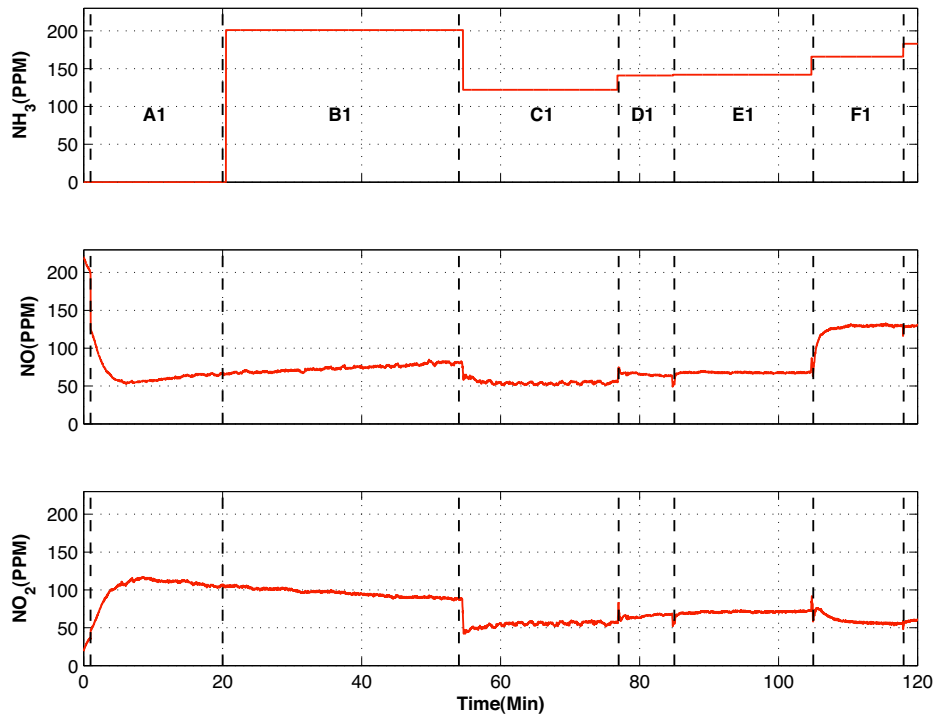


Figure 5: Concentrations of NO, NO₂ and NH₃ species at the inlet of the SCR catalyst during the test. A1, B1 etc are the steady state points shown in Figure 3. NH₃ is calculated from the urea injection flow rate as shown in Eq. 11.

$C_{i,s}$ and $C_{i,t}$ in Eq. 12 refer to the simulated and test concentrations of the species.

The reaction rate parameters identified (calibrated) for the 3 state model are tabulated in Table 1. The validation of this model is discussed in reference [8]. The total NH₃ adsorption capacity, Ω is identified as 158 moles of NH₃ per m^3 of exhaust gas volume.

Table 1: Reaction Rate Parameters Identified for the 3 State Model

Reaction	A	E(kJ/mol)
SCR Reaction	$3.32E9 \frac{m^3}{mol-sec}$	84
NH ₃ Oxidation	$4.33E6 \frac{1}{sec}$	114
NH ₃ Adsorption	$2.13E7 \frac{m^3}{mol-sec}$	43
NH ₃ Desorption	$3.28E11 \frac{1}{sec}$	112

The reaction rate parameters identified (calibrated) for the 4 state model are tabulated in Table 2. The total NH₃ adsorption capacity, Ω is identified as 157 moles of NH₃ per m^3 of exhaust gas volume. A comparison based on the rate parameters identified for the 4 state and 3 state models is shown for the NH₃ oxidation, NH₃ adsorption and NH₃ desorption reactions in the form of an Arrhenius plot by Devarakonda et al. in [8].

The 3 state model and the 4 state model have different SCR reactions. The fast, standard and NO₂ based SCR reactions considered in the 4 state model are considered as a single 'SCR Reaction' in the 3 state model. Also,

Table 2: Reaction Rate Parameters Identified for the 4 State Model

Reaction	A	E(kJ/mol)
Fast SCR	$4.50E14 \frac{m^6}{mol^2-sec}$	100
Standard SCR	$3.50E5 \frac{m^3}{mol-sec}$	75
NO ₂ SCR	$2.83E8 \frac{m^3}{mol-sec}$	85
NH ₃ Oxidation	$4.44E6 \frac{1}{sec}$	106
NH ₃ Adsorption	$5.24E6 \frac{m^3}{mol-sec}$	47
NH ₃ Desorption	$3.26E9 \frac{1}{sec}$	114

a comparison is made in the activation energies of all the reactions between the 3 state and the 4 state model. To compare the activation energy of the SCR reaction in both the models, an average of the activation energies of the fast, standard and NO₂ SCR reactions in the 4 state model from Table 2 is compared with that from the 3 state model from Table 1 and the % difference in activation energy is obtained as 3%. Magnitudewise, the pre-exponential factors and activation energies of NH₃ oxidation from both the models vary within 2% and 7% respectively. The activation energies of all the other reactions vary between 1-10% between the models.

Figure 6 shows the validation of the 4 state model based on the test data in NO, NO₂ and NH₃ concentrations. Figure 7 indicates the adequacy of the reduced order models for model-based control. The validation of various models and the adequacy of the 4 state model for model-based control is discussed in detail in reference [8].