

MEAN-VALUE MODEL OF THE SCR SYSTEM OF A MOBILE APPLICATION

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Abstract: This paper presents a mean-value model describing the SCR system of a heavy duty mobile application. The system consists of two catalysts, one for the preoxidation and one for the selective catalytic reduction (SCR) of NO_x . The model of the catalytic converter system is one-dimensional. Its core consists of three identical cells that describe the thermal and the chemical behavior of the SCR catalyst. The performance of the model is verified by comparing results of simulations and of measurements taken on a dynamic test bench. *Copyright © 2002 IFAC.*

Keywords: SCR catalyst, nitrogen oxide, urea, ammonia, mean-value model, mobile application

1. INTRODUCTION

Indications are that the emission limits defined by future legislation for heavy-duty Diesel vehicles no longer can be met by engine tuning only. An aftertreatment system thus will become mandatory in order to reduce nitrogen oxide (NO_x) and/or to abate particulate (PM) emissions. With an aftertreatment system in place, the engine can again be operated fuel-optimally, due to the NO_x -PM trade-off: The resulting high raw emissions of NO_x are considerably reduced in the aftertreatment system, whereas PM is already at a low level and is lowered even further by the aftertreatment system.

An SCR (Selective Catalytic Reduction) exhaust gas aftertreatment system which uses an urea solution as a reductant has a high NO_x reduction potential and is a well-known technique for stationary applications. In mobile applications, however, the volume of the catalytic converter is limited and the engine runs in transient operation mode most of the time. This causes certain problems due to the variations in exhaust gas temperatures and due to fast changes in the space velocity.

This paper presents a mean-value model that describes the SCR system of a mobile heavy duty application. Its output represents the exhaust gas temperature, and the molar flows of NO_x and NH_3 downstream of the aftertreatment system. The range of engine power is 10 to 110 kW. For the present investigations, the engine speed is held constant.

Section 2 presents the structure of the model. The experimental setup is described in section 3. Section 4 deals with the model parametrization. Finally, in section 5, the performance of the model is verified with measurements.

2. MODEL OF THE SCR SYSTEM

The plant to be modelled is the catalytic converter system in the exhaust gas system of a Diesel engine. It consists of an oxidation catalyst, a device to inject urea solution, and the SCR catalyst, as shown in Fig. 1. The catalytic converter system is modelled with $n+1$ cells. The first is called the oxidation cell. It describes the system between the measurement points 4 and 5. It is followed by n identical SCR cells, which together model the plant between the measurement points 5 and 6.

2.1 Oxidation Cell

The model assumptions for the oxidation cell are:

- The injected urea solution is heated to the temperature of the exhaust gas. No urea decomposition takes place.
- The molar flow of NO_x is not affected.
- The oxidation catalyst is a perfect heat exchanger and the exhaust gas leaving it has the same temperature as the catalyst.

The thermal part of the model takes into account the energy used to heat the urea solution, the conductive heat flux from the oxidation catalyst (OC) to the pipe wall (OCW), and the radiation from the pipe wall to the ambient air.

Based on the definitions $a_1 = \frac{c_{p,EG}}{c_{p,OC}m_{OC}}$,

$$a_2 = \frac{(1-c_U)c_{p,H_2O(g)} + c_Uc_{p,U}}{c_{p,OC}m_{OC}}, \quad a_4 = \frac{\alpha_{Cond}A_{Cond}}{c_{p,OC}m_{OC}},$$

$$a_3 = \left(\begin{array}{c} \frac{[(T_{BP} - T_{Amb})c_{p,H_2O(l)} + r_{v,H_2O}](1-c_U)}{c_{p,OC}m_{OC}} \\ \frac{(1-c_U)c_{p,H_2O(l)}T_{BP}}{c_{p,OC}m_{OC}} - \frac{c_Uc_{p,U}T_{Amb}}{c_{p,OC}m_{OC}} \end{array} \right),$$

$$a_5 = \frac{\alpha_{Cond}A_{Cond}}{c_{p,OCW}m_{OCW}}, \quad a_6 = \frac{\epsilon_{Rad,OCW}\sigma_{SB}A_{Rad,OCW}}{c_{p,OCW}m_{OCW}},$$

and $c_1 = \frac{c_U}{M_U}$, the state, input, and output vectors

$$\underline{x} = [x_1 \ x_2]^T = [T_{OC} \ T_{OCW}]^T$$

$$\underline{u} = [u_1 \ u_2 \ u_3 \ u_4]^T = [\dot{m}_{US,inj} \ \dot{n}_{NO_x,4} \ \dot{m}_{EG,4} \ T_4]^T,$$

$$\underline{y} = [y_1 \ \dots \ y_5]^T = [\dot{n}_{U,5} \ \dot{n}_{NH_3,5} \ \dot{n}_{NO_x,5} \ \dot{m}_{EG,5} \ T_5]^T$$

the equations for the oxidation cell read as follows:

$$\dot{x}_1 = a_1 u_3 (u_4 - x_1) - (a_2 x_1 + a_3) u_1 - a_4 (x_1 - x_2), \quad (1)$$

$$\dot{x}_2 = a_5 (x_1 - x_2) - a_6 (x_2^4 - T_{Amb}^4)$$

$$y_1 = c_1 u_1, y_2 = 0, y_3 = u_2, y_4 = u_1 + u_3, y_5 = x_1. \quad (2)$$

The output signal y_2 is the molar flow of ammonia, which is zero at location 5, upstream of the first SCR cell.

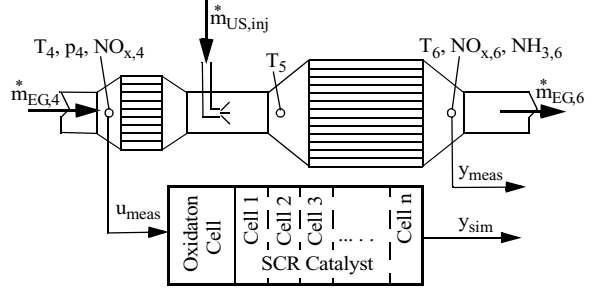


Fig. 1. The plant to be modelled is the exhaust gas system, consisting of the oxidation catalyst, the urea injection, and the SCR catalyst.

2.2 SCR Cell

The model assumptions for the description of the SCR catalyst are:

- The model includes one solid component, urea, and two gases only, NO_x and NH_3 .
- Only one reaction for urea decomposition is modelled:
 $(\text{NH}_2)_2\text{CO}(s) + \text{H}_2\text{O}(g) \rightarrow 2\text{NH}_3(g) + \text{CO}_2(g)$.
- Adsorption and desorption of NH_3 are modelled as taking place on the surface of the catalytic converter.
- The Eley-Rideal reaction kinetics for the SCR are:
 $4\text{NH}_3(ads) + 4\text{NO}(g) + \text{O}_2(g) \rightarrow 4\text{N}_2(g) + 6\text{H}_2\text{O}(g)$
- NO_2 is considered to react similarly to NO , consuming one NH_3 molecule for each NO_2 molecule.
- The oxidation of NH_3 adsorbed on the surface of the catalytic converter is described as follows:
 $4\text{NH}_3(ads) + 3\text{O}_2(g) \rightarrow 2\text{N}_2(g) + 6\text{H}_2\text{O}(g)$.
- No adsorption of reaction products occurs.
- Homogenous, incompressible flow of ideal gas.
- The influence of washcoat diffusion is negligible.

Kinetic gas theory is used to describe adsorption. Desorption, SCR reaction, and the ammonia oxidation are modelled with Arrhenius functions. Applying mass balance laws, a system of coupled PDEs results. For easier computer implementation and later use in model-based control systems or estimators, the PDEs are approximated by ODEs through partitioning the converter into n idealised cells along its flow axis. Temperature distribution and gas concentrations are assumed to be homogenous in each cell. The catalyst in each cell is assumed to be a perfect heat exchanger, and the exhaust gas leaving the cell has the same temperature as the catalyst. The thermal part of the cell model takes into account the energy released or consumed by the reactions, the conductive heat flux, and the energy loss by radiation. Defining

$$\begin{aligned}
a_0 &= \frac{R_{s,EG}}{p_{Amb}}, \quad a_1 = \frac{n_{Cell}}{\varepsilon V_{Cat}}, \quad a_2 = k_{UD} e^{\left(\frac{-E_{a,UD}}{Rx_5}\right)}, \\
a_3 &= c_S \alpha_{P_{Rob}} \sqrt{\frac{Rx_5}{2\pi M_{NH_3}}}, \quad a_4 = c_S k_{Des} e^{\left(\frac{-E_{a,Des}}{Rx_5}\right)}, \\
a_5 &= c_S R k_{SCR} e^{\left(\frac{-E_{a,SCR}}{Rx_5}\right)}, \quad a_6 = c_S k_{Ox} e^{\left(\frac{-E_{a,Ox}}{Rx_5}\right)}, \\
a_7 &= \frac{n_{Cell} c_{p,EG}}{c_{p,Cat} m_{Cat}}, \quad a_9 = \frac{\varepsilon_{Rad,SCR} \sigma_{SB} A_{Rad,SCR}}{c_{p,Cat} m_{Cat}}, \\
a_{12} &= \frac{\varepsilon V_{Cat} \Delta h_{UD}}{c_{p,Cat} m_{Cat}}, \quad a_{13} = \frac{\varepsilon V_{Cat} \Delta h_{Ads}}{c_{p,Cat} m_{Cat}}, \\
a_{15} &= \frac{\varepsilon V_{Cat} \Delta h_{SCR}}{c_{p,Cat} m_{Cat}}, \quad a_{16} = \frac{\varepsilon V_{Cat} \Delta h_{Ox}}{c_{p,Cat} m_{Cat}},
\end{aligned}$$

and the state, input, and output vector to be

$$\begin{aligned}
\underline{x} &= [x_1 \ x_2 \ x_3 \ x_4 \ x_5]^T = [c_U \ c_{NH_3} \ c_{NO_x} \ \Theta_{NH_3} \ T]^T \\
\underline{u} &= [u_1 \ u_2 \ u_3 \ u_4 \ u_5]^T = [\dot{n}_U^* \ \dot{n}_{NH_3}^* \ \dot{n}_{NO_x}^* \ \dot{m}_{EG}^* \ T]_{in}^T \\
\underline{y} &= [y_1 \ y_2 \ y_3 \ y_4 \ y_5]^T = [\dot{n}_U^* \ \dot{n}_{NH_3}^* \ \dot{n}_{NO_x}^* \ \dot{m}_{EG}^* \ T]_{out}^T
\end{aligned}$$

yields the following equations for each cell:

$$\begin{aligned}
\dot{x}_1 &= a_1(u_1 - a_0 u_4 x_5 x_1) - a_2 x_1 \\
\dot{x}_2 &= \left(\begin{array}{l} a_1(u_2 - a_0 u_4 x_5 x_2) + a_4 x_4 + \dots \\ \dots + 2a_2 x_1 - a_3 x_2(1 - x_4) \end{array} \right) \\
\dot{x}_3 &= a_1(u_3 - a_0 u_4 x_5 x_3) - a_5 x_3 x_4 x_5 \\
c_S \dot{x}_4 &= a_3 x_2(1 - x_4) - (a_4 + a_6)x_4 - a_5 x_3 x_4 x_5 \\
\dot{x}_5 &= \left(\begin{array}{l} a_7 u_4(u_5 - x_5) - a_{12} a_2 x_1 - a_{16} a_6 x_4 + \dots \\ \dots - a_{13} [a_3 x_2(1 - x_4) - a_4 x_4] + \dots \\ \dots - a_{15} a_5 x_3 x_4 x_5 - a_9(x_5^4 - T_{Amb}^4) \end{array} \right) \\
y_1 &= a_0 u_4 x_1 x_5, \quad y_2 = a_0 u_4 x_2 x_5, \quad y_3 = a_0 u_4 x_3 x_5, \\
y_4 &= u_4, \quad y_5 = x_5.
\end{aligned} \tag{3}$$

2.3 Model of the Catalytic Converter System

As stated in the beginning of section 2, the model for the whole catalytic converter system is built with one oxidation cell and n identical SCR cells. The input vectors of cells 1 to n are the output vectors of the preceding cells:

$$u_{i+1} = y_i, \quad i = 1, 2, 3, \dots, n. \tag{5}$$

The oxidation cell is given the index 0. The order of the complete model is $2+5n$.

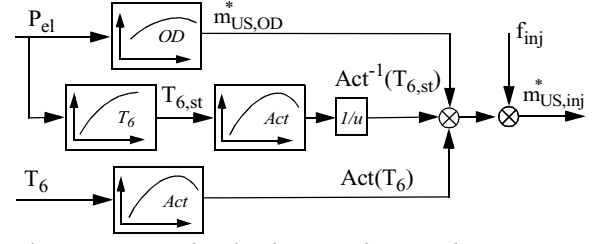


Fig. 2. Strategy for the dosage of urea solution.

3. EXPERIMENTAL SETUP

Measurements were conducted on a test bench consisting of a Liebherr D926 engine (6.6 liter, 4 cylinder), a generator, and a data logger for engine output signals. The engine is turbocharged and incorporates an inter-cooler. The maximal power is limited to 110 kW, the engine speed is set at 1500 rpm. The space velocity in the SCR catalyst is in the range of 28 to $47 \cdot 10^3/h$ (calculation with volume flow at norm conditions).

The concentrations of NO_x and NO were measured before and after the catalytic converter system, with a slow and a fast analyser, respectively. For the dynamic tests the fast NO and the slow NO_x signals were transformed into one hybrid signal that combines the dynamics of the fast analyser with the static gain of the slow instrument. The concentration of NH_3 was measured downstream of the catalytic converter system. The dynamics of the NH_3 analyser was determined, a compensator designed, and the signal then reconstructed off-line.

The strategy used to determine the desired amount of urea solution to be injected during the experiments is depicted in Fig. 2. It is static and consist of four blocks containing characteristic curves. The first and most important block calculates the optimal amount of urea solution to be injected $m_{US,OD}^*(P_{el})$, the second block determines the temperature downstream of the catalytic system at stationary conditions $T_{6,st}(P_{el})$, both as a function of engine power. The third and fourth blocks provide the catalytic activity, $Act(T_{6,st})$ and $Act(T_6)$, as a function of the temperature downstream of the catalytic converter at stationary conditions or measured at this point, respectively. The catalytic activity is defined as follows:

$$Act = \left(\frac{\dot{n}_{NO_x,4}^* - \dot{n}_{NO_x,6}^*}{\dot{n}_{NO_x,4}^*} \right)_{st}. \tag{6}$$

Note that the output of the third block is inverted. Additionally the product of $m_{US,OD}^*$, $Act^{-1}(T_{6,st})$ and $Act(T_6)$ is multiplied by the factor f_{inj} to determine the amount of urea solution to be injected $m_{US,inj}^*$.

At stationary conditions there are no thermal transients, and the factors $Act^{-1}(T_{6,st})$ and $Act(T_6)$ cancel each other. When, additionally, f_{inj} is set 1,

$\dot{m}_{US,inj}^*$ is equal to $\dot{m}_{US,OD}^*$, leading to a maximum reduction of NO_x and an NH_3 slip of exactly 10 ppm. Therefore the signal $\dot{m}_{US,OD}^*$ is called optimal dosage (OD).

At non-stationary conditions this strategy with four blocks containing characteristic curves helps to reduce transient NO_x and NH_3 emissions.

4. PARAMETRISATION OF THE MODEL

A number of parameters used in the oxidation cell and in the SCR cells are known a priori. However, a total of 17 parameters can only be estimated. They include the static gains, $(\alpha_{Cond} A_{Cond})$, two products $(\epsilon_{Rad,i} A_{Rad,i})$, all activation energies $E_{a,i}$ and pre-exponential factors k_i , the sticking probability α_{Prob} , the reaction energy of the SCR Δh_{SCR} , and four time constants, namely c_S and three instances of $(c_{p,i} m_i)$. These parameters were subject to an optimization.

In order to keep the computational efforts manageable, the number of SCR cells has to be kept as small as possible. On the other hand, a precise model requires a certain minimum of cells. Once $n = 3$ was found to be a good compromise, the model was parametrised using a series of three SCR cells.

The optimization of the parameters was done using both static and a choice of dynamic measurements. The overall optimization criterion was:

$$J_{tot} = J_{st} + \sum_i (w_{dyn,i} J_{dyn,i}). \quad (7)$$

For the optimization with the static measurements, the simulation was run with the series of SCR cells only. A total of 20 measurements at stationary conditions were taken: Power levels of 25, 50, 75 and 100 kW were considered and the following values of f_{inj} were selected: 0.5, 0.75, 1, 1.25 and 1.5. Measurements at $f_{inj} = 1$ were given the highest weight $w_{i,j}$, the lowest was given to $f_{inj} = 0.5$ and 1.5. Differences in temperature and molar flow of urea, NO_x , and NH_3 , all downstream of the SCR catalyst, contributed to the optimization criterion J_{st} .

During measurements on the test bench a complete urea decomposition was observed. In order to ensure that the model would work the same way the weighting factors were chosen $w_1 \gg w_2$.

$$J_{st} = \sum_{i,j} \left(w_{i,j} \left[\begin{array}{c} \Delta T_6^2 + w_1 (\Delta n_{U,6}^*)^2 + \dots \\ \dots + w_2 [(\Delta n_{NO_x,6}^*)^2 + (\Delta n_{NH_3,6}^*)^2] \end{array} \right] \right). \quad (8)$$

The dynamic measurements chosen for parameter optimization had to ensure that the model has the correct time constants. Simulations were done with the entire

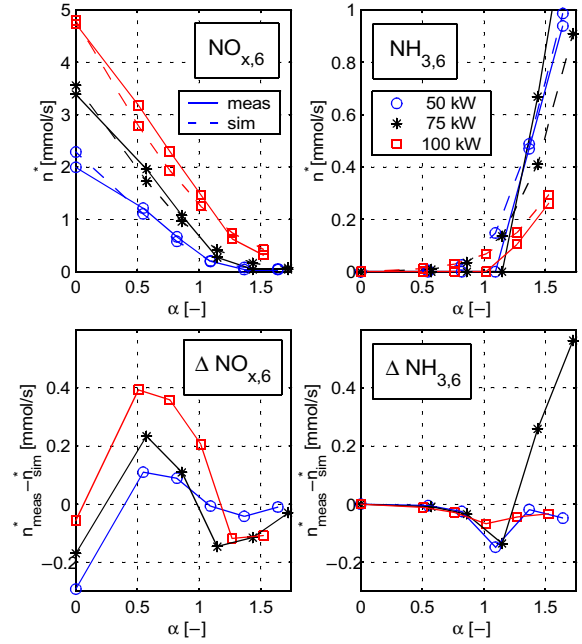


Fig. 3. Static behavior: Measurement results vs. the optimized model.

model of the plant. The differences in temperature up- and downstream of the SCR catalyst and the differences in NO_x and NH_3 downstream of the SCR catalyst contributed to the criterion J_{dyn} :

$$J_{dyn,i} = \frac{1}{(t_b - t_a)} \int_{t_a}^{t_b} \left[\Delta T_5^2 + w_2 (\Delta n_{NO_x,6}^*)^2 + \dots \right. \\ \left. \dots + \Delta T_6^2 + w_2 (\Delta n_{NH_3,6}^*)^2 \right] dt. \quad (9)$$

Fig. 3 shows the static gains at the 20 measured points after the parameter optimization. In all four plots, the feed ratio α is used as the abscissa:

$$\alpha = \frac{2 \dot{n}_{U,inj}^*}{\dot{n}_{NO_x,4}^*}. \quad (10)$$

The differences in molar flow of NO_x and NH_3 should be zero for all feed ratios.

- The difference in NO_x at every engine power level shows a curve reaching its maximum at a point below optimal dosage and its minimum at a point above optimal dosage. The reason for this is probably the modelling of one SCR reaction only. For engine power levels of 50, 75, and 100 kW ΔNO_x does not exceed 10 percent of the raw emission levels of NO_x , except at 50 kW and a feed ratio of zero.
- The measured ammonia slip shows a sharp rise at $\alpha \approx 1, 1$. The chosen model structure seems to be unable to match this sharp rise, provoking the typical shape in the difference in ammonia slip.

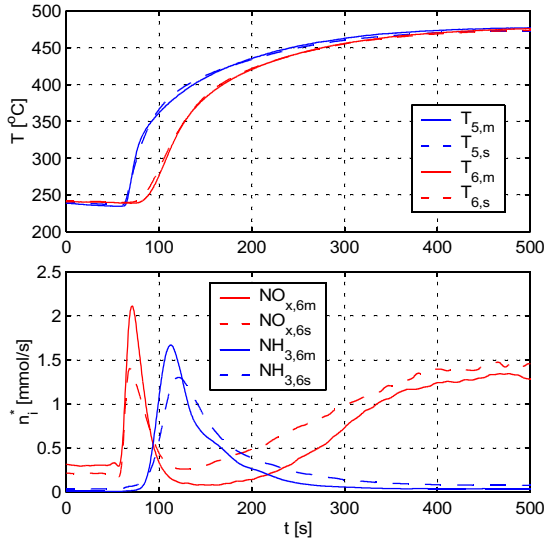


Fig. 4. Dynamic parameter optimization: The engine power is risen from 25 to 110 kW.

Fig. 4 shows an experiment where the engine power is raised stepwise from 25 to 110 kW.

- The thermal part of the model is quite representative of the real conditions.
- The chemical part shows discrepancies in static gains. The time constants are correct, however: Both the peaks and the slow transients in NO_x and NH_3 are represented correctly.

5. MODEL VERIFICATION

In order to verify the model for the catalytic system described in this paper, the catalytic system was investigated first by looking at the states of the model during stationary conditions. Second, the measurement results are compared to those obtained from the simulation with the model.

5.1 SCR along the Catalytic System

Fig. 5 shows the devolution of the SCR along the catalytic converter for 75 kW of engine power and for five different levels of urea dosage. The indices of the model cells are used as abscissae.

- Urea is decomposed almost completely in the first SCR cell. No urea is escaping from the catalytic converter system.
- The concentration of NH_3 is higher for higher dosage levels. It is very small for low dosage levels, since NH_3 adsorbs immediately. The concentration along the catalytic converter is always decaying.
- Starting at the same concentration, the decay in the concentration of NO_x occurs faster with higher dosage levels. Most of the SCR occurs in cell 1. The higher the dosage level, the smaller is the gain in NO_x reduction when an additional 25% of the optimal dosage (OD) is injected.

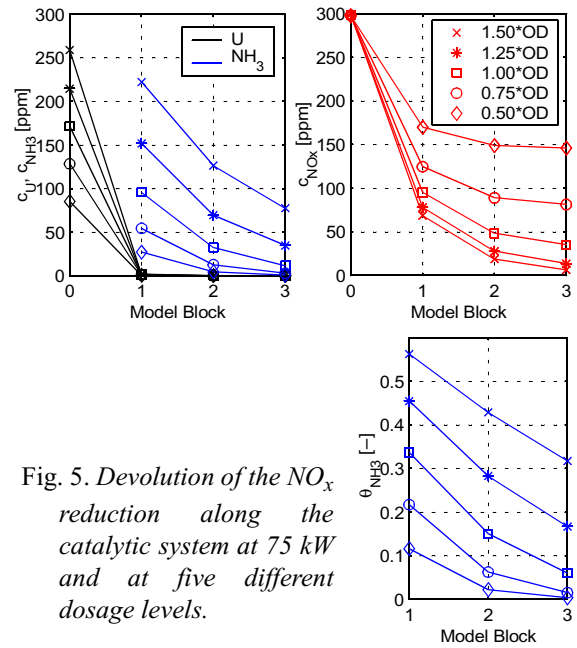


Fig. 5. Devolution of the NO_x reduction along the catalytic system at 75 kW and at five different dosage levels.

- The maximum coverage of the catalytic surface with ammonia is found in SCR cell 1. The shapes of the surface coverage curves are similar to the concentration curves of ammonia.
- With the choice $f_{inj} = 1$ the concentration of urea in the oxidation cell is about 170 ppm. After decomposition in SCR cell 1, most of the NH_3 adsorbs and is used in the SCR. Neither the concentration of NH_3 nor that of NO_x exceed 100 ppm. In SCR cells 2 and 3 the SCR of NO_x is completed. Finally, 40 ppm of NO_x and about 10 ppm of NH_3 exit SCR cell 3. Mass balance calculations show that about 70 ppm of NH_3 are directly oxidized.

5.2 Comparison of Measurements and Simulation

The comparison of measurements and simulation of dynamic experiments confirm the results stated above. The following observations are worth mentioning as well:

- The temperatures differences ΔT_5 and ΔT_6 vary in the range of $\pm 10^\circ C$ and $\pm 15^\circ C$, respectively.
- During simulations with optimal dosage, NO_x emission levels and NH_3 slip are too high. The difference ΔNO_x rarely exceeds 10 percent of the raw emission levels of NO_x (except at very low engine power). The simulated ammonia slip represents high measured peaks quite well, small ones often are overestimated.

6. CONCLUSIONS

The mean-value model of an SCR system for a mobile heavy-duty application includes just the five chemical reactions urea decomposition, ammonia adsorption and desorption on the SCR catalyst, oxidation of ad-

sorbed NH_3 , and the SCR itself, converting gaseous NO_x and adsorbed NH_3 .

The parameter optimization results in a model that is valid in a wide range of power and dosage levels. All time constants are modelled correctly. The thermal part is quite representative of the real conditions. The chemical part provides a good prediction for NO_x emissions and an acceptable consistency of simulated and measured NH_3 slip.

The model is thus validated to be suitable for the investigation of the processes occurring in the catalyst.

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NOMENCLATURE

Parameters:

A	area
Act	catalytic activity
c	concentration
c_p	heat capacity
c_s	concentration of active surface atoms per gas volume
E_a	activation energy
R_s	specific gas constant for exhaust gas
J	criterion for the optimization
k	pre-exponential factor
M	molar mass
m	mass
m^*	mass flow
n_{cell}	number of SCR cells
n^*	molar flow
p_{amb}	pressure of ambient air
P_{el}	engine power
r_V	latent energy
S	area of one mole of active surface atoms
T	temperature
u	input vector
V_{Cat}	volume of the SCR catalytic converter
V^*	volume flow
w	weighting factor of the parameter optimization
x	vector of state variables
y	output vector
α	feed ratio

α_{Prob}	sticking probability
Δh	reaction enthalpy
ε	fraction of gas volume in the SCR converter
Θ_{NH_3}	surface coverage by ammonia
σ_{SB}	radiation constant

Indices:

Ads	adsorption
BP	boiling point
Cat	catalyst
Cond	conduction
CO_2	carbon dioxide
Des	desorption
dyn	dynamic
H_2O	water, liquid (l), and gaseous (g)
EG	exhaust gas
inj	injected
m	measured
NO	nitric oxide
NO_2	nitrogen dioxide
NO_x	nitrogen oxide: $\text{NO} + \text{NO}_2$
OC	oxidation catalyst
OCW	wall of the oxidation catalyst
OD	optimal dosage
Ox	oxidation
Rad	radiation
SCR	selective catalytic reduction
s	simulated
st	static
U	urea, $(\text{NH}_2)_2\text{CO}$
UD	urea decomposition
4	measurement upstream of oxidation catalyst
5	measurement upstream of SCR catalyst
6	measurement downstream of SCR catalyst

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