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Modelling of selective catalytic reduction systems using discrete-time Linear Parameter Varying models

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Abstract: In this work, a Linear Parameter Varying (LPV) model of the Selective Catalytic Reduction (SCR) system, used for NO_x reduction placed as after-treatment systems in diesel engines is developed. The LPV model structure is formed utilising the physical properties of the system yielding only 7 unknown parameters for a third order model structure of the SCR, which is a significant reduction in number of parameters in comparison with other nonlinear models used in this paper. The states of the model however, do not possess any physical interpretation. The LPV model structure is validated using real measured data from cell tests at Scania AB with promising results. The proposed model is compared to previous global nonlinear models of the system, i.e. a nonlinear state space model and a Hammerstein-Wiener model of the system.

Keywords: Linear parameter varying systems, non-linear systems, selective catalytic reduction systems.

1. INTRODUCTION

Our modern society is highly dependent on heavy-duty vehicles. Transportation of good and human beings is increasing rapidly as one of the effects of globalisation and population development. Although these vehicles are necessary for keeping the living standards high, being powered by diesel engines, they emit a substantial amount of harmful emissions. Nitrogen oxide (NO_x) is a significant part of these. NO_x which consists of a combination of NO and NO_2, causes damage on the environment and is hazardous for human beings and wildlife. To be able to remove these emissions, after-treatment systems are widely used. Engine tuning was used to meet Euro I to Euro III legislations, Rasheed et al. (2013). With the stringent legislations introduced in Euro IV and V, new methods for NO_x reduction was necessary. One of the commonly used after-treatment systems today is the Selective Catalytic Reduction (SCR) system, where ammonia is used as a reduction agent to selectively reduce the NO_x. These systems are shown to be an economic way of reducing the NO_x.

With the Euro VI emission laws, The European Parliament and the Council (2009), even better performance of the SCR systems are required. In the future, the limitation on NH_3 will also be a new challenge to deal with. For this, good control of the SCR system is necessary. Good control design requires good models for the system. The sensors used today are cross-sensitive to both NO_x and NH_3, giving inaccurate output, Ericson (2009). By using reliable models, this problem can be mitigated. It is known, due to the chemical reactions involved that the dynamics of the system are highly nonlinear, see e.g. Ericson (2009); Schär et al. (2006).

Modelling of this nonlinear system can be divided into two branches, the physical/chemical modelling, where numerous authors have contributed in the development of detailed and complex models for the SCR using the complete chemical reactions. E.g. in Sjövall et al. (2010) a transient kinetic model for SCR of NO_x with ammonia over an Fe-zeolite catalyst for a wide range of temperatures (i.e. 150°C to 650°C) is presented. In Ericson (2009), a complete model of the SCR based on the temperature and ammonia propagation inside the catalyst is presented. The same idea is developed in the work of Schär et al. (2006); Feng (2010). Recently, Rasheed et al. (2013) presented a model based feed forward controller using the model presented in Schär et al. (2006) There are also models of the SCR where the dynamics of the system are simplified, one of these works are presented Brandt et al. (2000), where a model of a three-way catalytic converter is developed. The second branch includes black-box modelling,Tayamon et al. (2011); Zambrano et al. (2011); Tayamon and Zambrano (2013). Several identification approaches using the nonlinear black-box identification techniques has been used, for instance nonlinear state space models, Tayamon et al. (2011), where feedback linearisation techniques have been applied for control of the system.

LPV modelling approach, where the nonlinear system is described as a linear system with time-varying parameters in the system matrices, is a technique of reducing the nonlinearities in a nonlinear plant. LPV models can be seen as a more general form of linear time-varying systems, where the parameters are time varying in the system description. In recent years, LPV modelling approach has gained a major attention for identification and control. An overview of the available LPV models can be

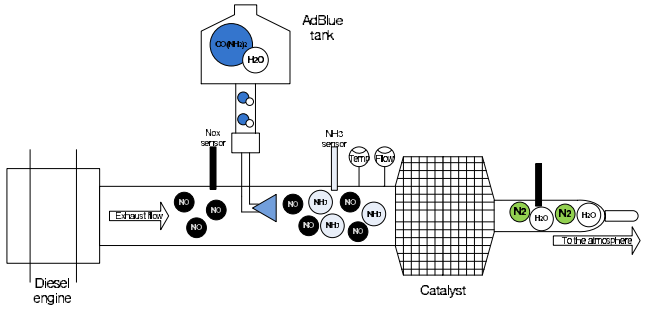

Fig. 1. SCR system

found in Tóth (2010). The theory for LPV identification methods is well-developed. Mohammadpour and Scherer (2012) presents an overview of the theory of the technique together with many application. One of the main reason for this attention is the ability to apply well-known linear gain scheduling controllers for these systems, Apkaria and Adams (1998). But also other well-known linear control techniques re-defined for the LPV system such as LQG, Wu and Packard (1995) can be implemented. Besselmann et al. (2012) presents an MPC approach for LPV systems.

The main contribution of this paper is to present a global input-output LPV model on state-space form of the SCR system. The paper is organised as follows, a brief description of the SCR system and the reactions together with the system setup and the collected data are given in Section 2. The proposed model structure together with the comparison models are presented in Section 3. The identification procedure is described in Section 4. The results are discussed in Section 5 and finally, some concluding remarks are given in Section 6.

2. THE SELECTIVE CATALYTIC REDUCTION SYSTEM

2.1 Chemistry of the SCR system

An overview of the SCR system is depicted in Fig. 1. The SCR system consists of three main stages. Firstly, urea, working as a reduction agent, is injected into the hot exhaust gas flow at the input of the catalyst where the water is evaporated. Urea is contained in a harmless aqueous solution commercially named AdBlue, which consist of 32.5% of urea. The reacting substance reducing the actual NO_x is the ammonia, therefore urea needs to be thermally converted to ammonia first.

The second stage is when the ammonia is partially adsorbed on the surface of the catalyst. At this stage the dominant reactions occur in the catalyst, i.e. the ammonia reacts with the NO_x emitted by the engine.

- Adsorption and desorption of ammonia on the catalyst
$$NH_3(\text{gas}) \leftrightarrow NH_3(\text{solid}) \tag{1}$$

NO_x is composed primarily of NO and a lesser amounts of NO_2. The resulting compounds of the ideal reaction is nitrogen gas (N_2) and water (H_2O).

The two main reactions of the SCR are for the conversion of NO is

- Standard SCR reaction
$$4NH_3 + 4NO + O_2 \rightarrow 4N_2 + 6H_2O \tag{2}$$
- Fast SCR reaction
$$4NH_3 + 2NO + 2NO_2 \rightarrow 4N_2 + 6H_2O \tag{3}$$

In this case, (3) is kinetically more favourable than (2).

Further, the NO_2 reaction is much slower than (2) and (3) and therefore negligible.

The final and third stage of the SCR is the oxidisation of the ammonia where the reaction is given by

- Adsorption and desorption of ammonia on the catalyst
$$4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O \tag{4}$$

The resulting chemical/physical model of these reactions consist of the following ODE:s where the governing equations related to the adsorption and desorption of the ammonia in the catalyst are represented by the rate of change of the ammonia coverage, θ_{NH_3}, which is the amount of stored ammonia inside the catalyst and is defined by the following reaction rates

$$\begin{aligned}\frac{\dot{\theta}_{NH_3}}{c_S} &= r_{\text{Adsorption}} - r_{\text{Desorption}} - r_{\text{SCR}} - r_{\text{Oxidisation}} \\ &= r_1 - r_2 - r_3 - r_4,\end{aligned} \tag{5}$$

where c_S is the number of reaction sites per volume of catalyst wash-coat. The reaction rates in (5) are given by

$$\begin{aligned} r_1 &= k_1 c_{NH_3}\left(1-\theta_{NH_3}\right), \\ r_2 &= k_2 e^{\left(-\frac{E_{a,2}\left(1-\theta_{NH_3}\right)}{RT}\right)}\theta_{NH_3}, \\ r_3 &= k_3 e^{\left(-\frac{E_{a,3}}{RT}\right)} c_{NO_x}\theta_{NH_3}, \\ r_4 &= k_4 e^{\left(-\frac{E_{a,4}}{RT}\right)}\theta_{NH_3}, \end{aligned}$$

where, $k_i,\ i = 1,\ldots,4$ is the preexponential factor of the Arrhenius type expression and $E_{a,i},\ i = 2,\ldots,4$ is the activation energy. R is the molar gas constant and T is the surface temperature and c_j is the concentration of the compound j.

The output NO_x is then given by

$$c_{NO_{x,\text{out}}} = \frac{a_1 c_{NO_{x,\text{in}}}}{a_2 F_{\text{tot}} T + RTR_3}, \tag{6}$$

where a_1 and a_2 are constants and F_{tot} is the exhaust gas flow.

2.2 Data

Two types of data are used for this work. The first one is collected using a high fidelity simulator based on the physical model described in Ericson (2009), the simulator uses the input load and speed to the engine generate the inputs to the SCR according to the chemical properties of a given SCR system. One of the main advantages for using the simulator is the ability to analyse the input signals to the SCR such that the data covers all operating points and frequencies of interest. For validation of the model, the World Harmonised Transient Cycle (WHTC), Steven (2001) a transient test with a length of 1800 s which specifies engine speed and load values is used. The first section of the WHTC data includes a high transient part,

describing urban driving conditions. At the end there is a slightly more stationary part, representing driving on the highway. This cycle is used for validation of the emission legislations set by the EU and the USA, Steven (2001).

The second data type is a WHTC data set collected at Scania AB. This data is used to validate the model structure, note that the real measured data is also used for identification the unknown parameters separately. The data is collected in cell environment and therefore, the output NO_x is measured using high accuracy NO_x sensors, reducing the NO_x and ammonia cross-sensitivity problem.

3. MODEL STRUCTURES

In this section, the LPV model structure is presented in detail. A brief overview of the comparing models are also given.

3.1 Discrete-time Linear Parameter Varying systems

A nonlinear plant model may be described as a linear parameter varying, LPV, system which is a subclass of nonlinear systems. LPV models are models with linear structures, where the system description contains parameters that vary w.r.t time t.

A discrete-time LPV system on state space form can be described as

$$\begin{aligned} x(t+1) &= A(\rho(t))x(t) + B(\rho(t))u(t) \\ y(t) &= C(\rho(t))x(t) + D(\rho(t))u(t), \end{aligned} \tag{7}$$

where $x(t) \in \mathbb{R}^n$, $u(t) \in \mathbb{R}^{n_u}$, $y(t) \in \mathbb{R}^{n_y}$ and $\rho(t) \in \mathbb{R}^{n_\rho}$ denote the state, the input, the output and the scheduling signal vector, i.e. the varying parameter vector of the system. $\rho(t)$ is assumed to be priori unknown, but measurable online. $A(\rho(t)), B(\rho(t)), C(\rho(t)), D(\rho(t))$ are analytic matrix valued functions.

This model setup can further be written as

$$\begin{pmatrix} x(t+1) \\ y(t) \end{pmatrix} = \begin{pmatrix} A(\rho(t)) & B_1(\rho(t)) & B_2(\rho(t)) \\ C_2(\rho(t)) & D_1(\rho(t)) & D_2(\rho(t)) \end{pmatrix} \begin{pmatrix} x(t) \\ u(t) \\ w(t) \end{pmatrix}, \tag{8}$$

where $u(t)$ is the controllable input signal and $w(t)$ is the exogenous input signal, that is measurable at time t but not controllable.

3.2 The proposed LPV model

The model structures used in this paper, is mainly based on the knowledge of the physical model of the system as described in Schär et al. (2006). However, there are no physical interpretation of the model structure and the states.

There are several inputs and outputs measured in the SCR, and many of them are attainable in the simulator. As it is seen in section 2.1, there are 4 main components affecting the dynamics of the SCR. The input NO_x and NH_3 are of course crucial for the NO_x reduction. One other component important for system behaviour is the temperature of the exhaust gas effecting the temperature of the solid material inside the catalyst. The chemical reactions described in section 2 are sensitive to the temperature of

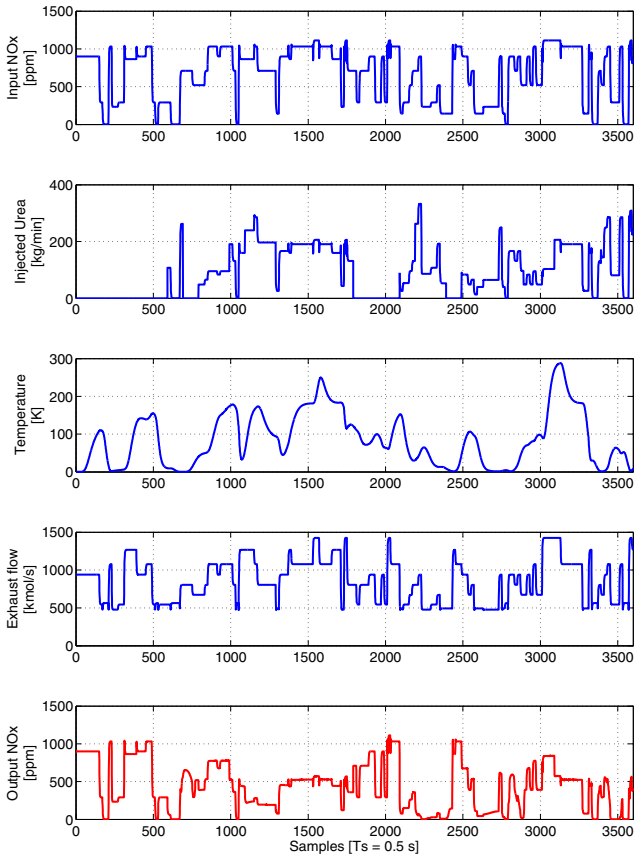

Fig. 2. The input signals and the output NO_x generated from the simulator environment.

the catalyst. The non-controllable inputs generated by the simulator to the SCR system are given by

$$\begin{aligned} v_1(t) &= c_{\mathrm{NO_{x,in}}}, \\ v_2(t) &= \rho_1(t) := F_{\mathrm{tot}}, \\ v_3(t) &= \rho_2(t) := T_{\mathrm{in}}, \end{aligned} \tag{9}$$

where, $c_{\mathrm{NO_{x,in}}}$ denotes the concentration of $\mathrm{NO_{x,in}}$ in [ppm], T_{in} in $[K]$ is the temperature of the exhaust gas prior to the SCR and F_{tot}, measured in [g/h] is the exhaust flow into the catalyst. The urea is the only controllable input to the system, therefore it is clear that $u(t)$ in (7) should be represented by the concentration of the NH_3,

$$u(t) = c_{\mathrm{NH_3}} \tag{10}$$

Fig. 2 illustrates the desired input and output of the SCR system generated using a transient step varying for the input load and speed to the simulator. This data is used to analyse the system inputs and their effect on the output NO_x. It can be noticed that the exhaust flow exhibits steady values and high frequency changes. The amount of injected urea is related to the NO_x input concentration and to the exhaust flow. In the simulator environment the input urea is connected to the input NO_x using a varying parameter, ammonia to NO_x ratio, ANR changing between $0-1$. By choosing a varying ANR, cross-correlation problems for input signals are omitted. Also, the temperature changes smoothly for a broad range of values, i.e. 470 K to 670 K approximately (200° C to 400° C approximately).

As we are dealing with chemical reactions, the temperature will effect the system exponentially as a function of the

reaction rate. This is taken into account by the following nonlinear transformation

$$v_3^*(t) = \rho_2^*(t) := e^{-a/T_{\text{in}}}, \tag{11}$$

where a is a constant value chosen as 1×10^4 as described in Tayamon et al. (2011).

Using the information above, a model on the form (7) where the varying parameters are the two non-controllable input signals, i.e. the input flow, v_2 and the modified temperature v_3^* and the product of these two, as seen in (6) can be used as varying parameters. Further, the effect of the input NO_x is included as a measurable but non-controllable input to the system.

$$\begin{aligned} x(t+1) &= A(\rho(t))x(t) + B_1(\rho(t))u(t) + B_2 v_1(t) \\ y(t) &= C(\rho(t))x(t) \end{aligned} \tag{12}$$

where

$$A(\rho(t)) = \begin{pmatrix} 0 & \rho_2^*(t) & 0 \\ 0 & 0 & \rho_2^*(t) \\ a_1\rho_1(t) & a_2\rho_2^*(t) & \rho_2^*(t) + a_3\rho_3(t) \end{pmatrix}$$

and

$$\begin{aligned} B_1(\rho(t)) &= \begin{pmatrix} 0 \\ 0 \\ b_1\rho_3(t) \end{pmatrix}, \quad B_2 = \begin{pmatrix} n_1 \\ 0 \\ n_2 \end{pmatrix} \\ C(\rho(t)) &= \begin{pmatrix} c_1 & \rho_1(t) & \rho_2^*(t) \end{pmatrix} \end{aligned} \tag{13}$$

where the time-varying parameter $\rho_1(t)$ is given by (9), $\rho_2(t)$ is given by (11) and $\rho_3(t)$ is defined by

$$\rho_3(t) = \rho_1(t)\rho_2^*(t).$$

and the unknown parameter vector θ

$$\theta = \begin{pmatrix} a_1 & a_2 & a_3 & b_1 & c_1 & n_1 & n_2 \end{pmatrix}. \tag{14}$$

The model structure described in (12) is based on the controllable canonical form, with all parameters unknown. The varying parameters as chosen in section 2, are added to each element step-wise to each non-zero element. The obtained results are compared to the previous result and the best output estimate is chosen. Each estimated parameter close to zero is set to zero.

3.3 Nonlinear state space model

The results obtained in this paper will be compared to a previous work by the author where a global nonlinear state space (NL-SS) model is used for identification of the SCR system. The nonlinear model is given on the form

$$\begin{aligned} \dot{x}_1(t) &= x_2(t) \\ &\vdots \\ \dot{x}_{n-1}(t) &= x_n(t) \\ \dot{x}_n(t) &= f(x(t), u(t), v(t), \theta), \\ y(t) &= x_1(t), \end{aligned} \tag{15}$$

where

$$v(t) = \begin{pmatrix} v_1(t) \\ v_2(t) \\ v_3^*(t) \end{pmatrix},$$

and $u(t)$ is the input urea as previously described, and θ is the unknown parameter vector to be estimated.

The identification procedure for this model is performed using a recursive prediction error method, Tayamon and Wigren (2010). For a detailed description of the identification algorithm and its application on the SCR system see e.g. Tayamon et al. (2011).

Remark: Note that the results obtained in the coming sections are not performed recursively. The final parameter vector is used for simulating the identification and validation estimated output.

3.4 Hammerstein-Wiener systems

Several Hammerstein-Wiener (HW) model structures were presented in a previous work by the author in Zambrano et al. (2011). The model structure with the best results obtained are used in this work, where the static nonlinearities on the input signals v_1, v_2, v_3 and u and are defined as piecewise linear (PWL) functions. The output nonlinearity is defined as a saturation on the output due to the physical limitations of the system were no negative NO_x can be produced.

The linear model of the HW-model is defined as a third order discrete-time state space model with the input

$$\bar{u}(t) = \begin{pmatrix} g_1(v_1(t)) \\ g_2(v_2(t)) \\ g_3(v_3(t)) \\ g_4(u(t)) \end{pmatrix},$$

and $g_i(.)$ is the PWL function for each input i.

Remark: The input temperature is not modified as in (11) in this model structure in contrary to the LPV and nonlinear state space model.

4. IDENTIFICATION

4.1 Data preprocessing

The order of magnitude of the input and output signals differ largely from each other. To obtain a data set which has good numerical properties, the signals are normalised using the standard deviation, σ and each signal's mean. The values used for preprocessing of the two data sets are shown in Table 1. Note that this preprocessing introduces the $-1.5 \leq \rho_i(t) \leq 16$ bound for $i = 1, 2, 3$.

Remark: Note that the injected urea in the real data has the unit [g/min] causing the difference in magnitude in comparison for the simulated data.

4.2 LPV identification

The identification procedure, is done using the prediction error method. The model structure is defined as a nonlinear grey-box object using the system identification toolbox in MATLAB, Ljung (2000).

The initial parameter vector θ_0 was chosen ad hoc such that the eigenvalues of $A(\rho(t)), \forall\rho_i(t), -1.5 \leq \rho_i \leq 16, i = 1, 2, 3$ were inside the unit circle with the following θ_0

$$\theta_0 = \begin{pmatrix} -0.2 & -1 & -4 & 2 & -2 & 1 & 1 \end{pmatrix}.$$

The parameter estimation is bounded such that the parameters never violate the stability conditions for the system. This is easily done by forcing the eigenvalues to be always inside the unit circle.

Table 1. Values used for preprocessing.

	Variable	Output NO_x	Injected urea	Input NO_x	Modified temperature	Exhaust flow
Simulator	Mean	408.75	88.44	641.24	2.82×10^{-7}	838.03
	σ	315.36	470.48	357.13	2.53×10^{-7}	280.85
Measured	Mean	261.99	9.07	502.49	2.40×10^{-8}	620.78
	σ	235.52	11.48	408.08	3.06×10^{-8}	422.16

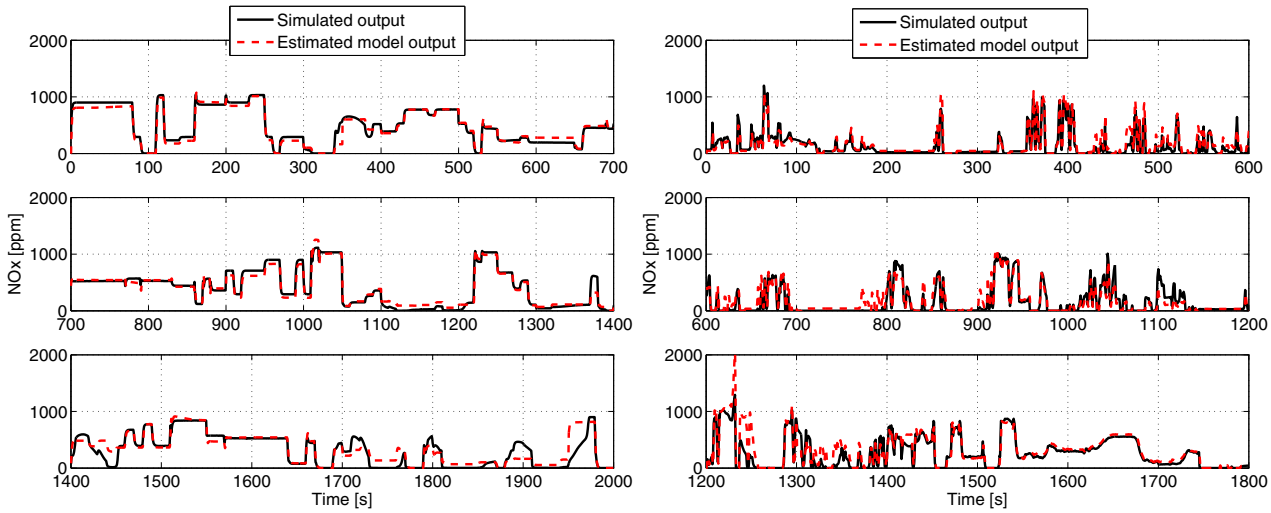

Fig. 3. Identification results, the left figure shows the identification data and the right the validation data with a model fit of 69.81 % and 58.32 % respectively. Note that the window is divided into three parts for improved viewablity of the results.

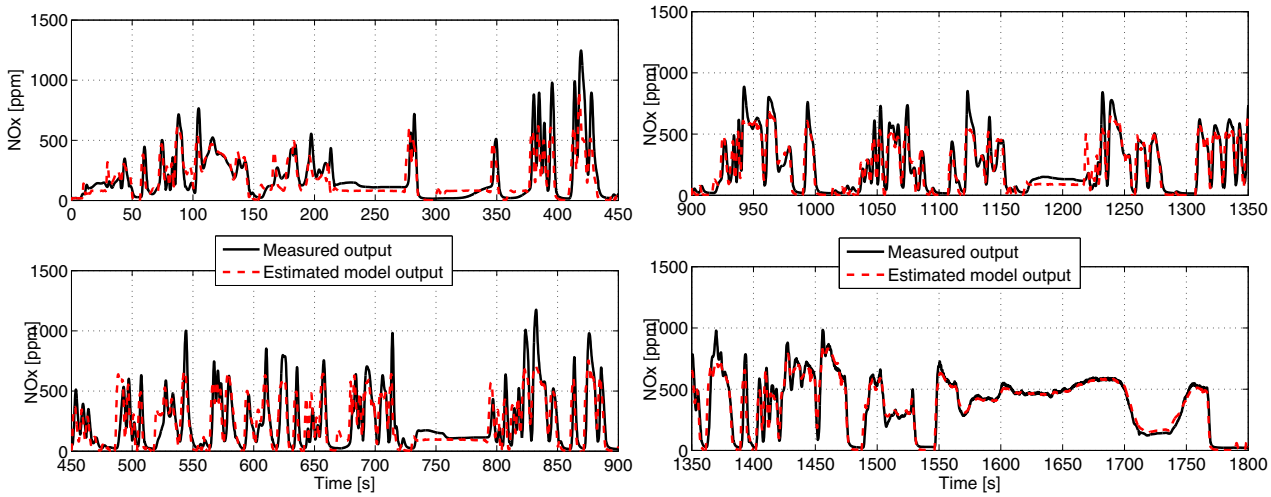

Fig. 4. Identification results, the left figure shows the identification data and the right the validation data. The model fit is for identification data 51.05% and 62.21% for the validation data. Note that the window is divided into three parts for improved viewablity of the results.

5. RESULTS

5.1 Identification results

For a quantitative comparison of the proposed model with the previous models, the model fit (FIT) and the mean square error (MSE) value is used for performance criteria.

$$\text{FIT} = \left(1 - \frac{||y_{\text{plant}} - y||_2}{||y_{\text{plant}} - \bar{y}_{\text{plant}}||_2}\right) \cdot 100, \tag{16}$$

$$\text{MSE} = \frac{1}{N}\sum_{t=1}^{N} \left(y(t) - y_{\text{plant}}(t)\right)^2. \tag{17}$$

where y is the estimated output from the model and y_{plant} is the output NO_x from the simulator/measured data. $\bar{y}_{\text{plant}}$ is the mean value of y_{plant} and N is the total number of samples. Further $\|.\|_2$ denotes the spectral norm.

The identification and validation data for from the simulator are depicted in Fig. 3. The figure illustrates that the model is capable of capture the system dynamics quite well for the given data. For a complete overview of the results

Table 2. Results

Model	Identification		Validation	
	FIT	MSE	FIT	MSE
NL-SS	75.88%	0.0299	58.67%	0.0454
HW	73.06%	0.0341	50.35%	0.0511
LPV	69.84%	0.0391	59.32%	0.0432

and comparison between the models see Table 2, where it can be seen that the proposed model works quite well. The LPV model performs almost as good at the nonlinear state space model. However, the LPV model is estimated using only 7 parameters in contrary to the nonlinear state space with 17 unknown parameters. The identification and validation results for the measured cell data are illustrated in Fig. 4.

One interesting observation of the obtained results are that there is no need to introduce a nonlinear saturation function on the output NO_x using the LPV model.

6. CONCLUSIONS

A black-box LPV model for the SCR system was presented in this work. The identified model represent the NO_x behaviour for transient operating conditions quite well. Of all the candidate models studied, this model provide a significant better reproduction of the experimental data over the whole analysed period in means of model fit and MSE and the total number of parameters identified. The simplicity of the model and low number of parameters identified is one of the main advantages of this model structure in comparison with other nonlinear models.

Future work includes development of suitable controllers and comparison with the controllers designed for the other nonlinear models described in this paper.

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