Numerical Simulation of NOx Reduction in a SCR System

A S Blinov, N S Malastowski and L L Myagkov
Bauman Moscow State Technical University, 5, 2-ya Baumanskaya str., Moscow, 105005, Russia
E-mail: as_blinov@bmstu.ru

Abstract. The stringent limits of emission standards require advanced emission control technologies to be used in modern on and off highway diesel engines. They include both in-cylinder and aftertreatment measures where the latter now have become almost mandatory. Selective catalytic reduction aftertreatment systems are widely used for nitrogen oxide (NOx) conversion in exhaust gases into harmless N2. To reduce time and costs, at the design stage of SCR systems numerical modelling is applied. Mathematical models and methods providing high prediction accuracy with an acceptable level of computational efforts are required. In this work an approach for complete simulation of SCR systems based on the coupling of commercial CFD software with developed multichannel 1D catalyst model is presented. The first one is used to carefully describe processes occurring upstream in the catalytic converter, particularly, during urea water solution injection and flow mixing. As a result, the distributions of flow parameters at catalyst inlet are derived. They are subsequently imported as boundary conditions into a developed multichannel catalyst model that allows one to take them into account when calculating NOx conversion efficiency. Based on the proposed approach a SCR system was simulated. The effect of non-uniform distributions of NH3 concentration and the gas flow velocity at the catalyst inlet on its performance was investigated. It has been shown that they have a great impact on NOx conversion and should be taken into account during the catalyst modelling.

1. Introduction
To comply with tight ecological standards diesel engines, it is necessary to incorporate different emission control technologies [1 - 7]. Selective catalytic reduction systems are a widely used technique for NOx reduction in exhaust gases [8, 9]. They convert NOx to N2 and H2O through catalytic chemical reactions with NH3 (6). The latter is supplied to a SCR system in the form of urea-water-solution (UWS) injected in the exhaust pipe before the catalytic converter. Due to high exhaust gas temperature the UWS evaporates (1) and decomposes to NH3 and CO2 through chemical reactions of thermolysis (2) and hydrolysis (3) [10]:

\[ CO(NH_2)_2 \text{ (solution)} \rightarrow CO(NH_2)_2 + 6.9H_2O; \]  
\[ CO(NH_2)_2 \rightarrow NH_3 + HNCO; \]  
\[ HNCO + H_2O \rightarrow NH_3 + CO_2. \]

Many physical and chemical processes occur in SCR systems: the exhaust gas movement, the UWS injection and its interaction with the flow and the walls of exhaust pipes, the heat-transfer processes, the UWS evaporation and thermal decomposition, chemical reactions taking place in the gas flow, on the
catalyst surfaces, etc. [11]. They should be properly optimized to provide high NO\textsubscript{X} conversion efficiency for meeting emission standards requirements [12].

Numerical modeling has become an effective tool to solve such kind of optimization tasks [13]. At different design stages, models of various complexity are applied; that provides reduction of time and computational efforts [11].

At early design stage the simplified mathematical models are used [14]. They allow one in a short time to evaluate different system layouts and to find out the most promising ones. These kind of models are often implemented in thermodynamic engine simulation software, e.g. AVL BOOST, GT-POWER, ExACT, GADSYN etc. They allow one to perform the coupled modeling and optimization of an engine and aftertreatment systems. The results of numerical simulations of preliminary size dimensions and the layout of a SCR system, the catalyst characteristics, the UWS injection strategy that provide the required conversion efficiency are defined.

These simplified models can't properly describe gas flow dynamics in exhaust pipes, the UWS injection and decomposition processes, the mixing of gas flow chemical compounds that determine distributions of flow parameters at catalyst inlet and directly affect the conversion efficiency [15]. For this purpose, CFD models and related software, e.g. ANSYS FLUENT, CONVERGE, AVL FIRE, STAR-CCM+ etc., are used [16]. On the other hand, the application of such complex models for complete simulation of SCR systems requires high computational efforts. This is due to different timescales for UWS spray dynamics (ms) and catalyst transients (min) [17].

To reduce the time and computational efforts, the simulation can be divided into two sequential steps: modeling processes before and in the catalyst. For the first one, CFD models are used; for the second one – different catalyst models [18]. In this study, such integrated approach based on the coupling of ANSYS Fluent with the developed multichannel 1D catalyst model is presented.

2. Materials and methods

A schematic representation of the developed integrated approach for complete simulation of SCR systems is shown in Figure 1.

![Figure 1. Integrated approach for complete simulation of a SCR system.](image)

During the 1-st step of simulation, the gas flow dynamics throughout the SCR system, the UWS injection, evaporation and decomposition are modeled in ANSYS Fluent. The purpose of the simulation is to obtain data of flow parameters distributions at the catalyst inlet: temperature, velocity, chemical composition, etc.

The gas flow is described by Reynolds-averaged Navier-Stokes (RANS) equations coupled with the Realizable k-ε turbulence model and Enhanced Wall Treatment model [19]. For the catalyst, the brick porous medium approximation is adopted. The permeability resistance of the flow is governed by Darcy’s law where linear and inertial loss terms are determined based on experimental data [20]. The
UWS injection is modelled using the Lagrangian Discrete Phase Model [19]. The primary spray breakup is not simulated but is specified by Rosin-Rammler droplet size distribution and droplet speeds derived from experimental data [21]. The secondary droplet breakup is calculated using the Taylor Analogy Breakup model [22]. The evaporation of UWS droplets is described by the convection-diffusion controlled model for multicomponent particles [23]. Two-way coupling of mass, momentum and energy is considered between the gas flow and droplets. The decomposition process of evaporated urea is modelled through single step volumetric chemical reactions of thermolysis and hydrolysis (2, 3). The formation of intermediate reaction by-products is neglected. The reaction rates are calculated according to the law of mass action. The kinetic parameters and thermal effect of the reactions are taken from the literature [23].

In the next simulation step, physicochemical processes occurring in a SCR catalyst are simulated using the developed mathematical model implemented in MATLAB. It is based on the multichannel approach whereby a 1D single channel model is applied for each of them. It allows one to take into account gas flow parameters nonuniformity at the catalyst inlet. A detailed description of the applied single channel catalyst model can be found in the previous author’s work [24]. It simulates four dominant catalytic surface reactions: ammonia adsorption (4), desorption (5) and oxidation (6), NOx reduction (7):

\[ \text{NH}_3(g) \rightarrow \text{NH}_3(s); \quad (4) \]
\[ \text{NH}_3(s) \rightarrow \text{NH}_3(g); \quad (5) \]
\[ 4\text{NH}_3(s) + 3\text{O}_2(g) \rightarrow 6\text{H}_2\text{O}(g) + 2\text{N}_2(g); \quad (6) \]
\[ 4\text{NH}_3(s) + 4\text{NO}(g) + \text{O}_2(g) \rightarrow 6\text{H}_2\text{O}(g) + 4\text{N}_2(g). \quad (7) \]

Reaction rates are calculated according to equations (8 - 11). Kinetic parameters of chemical reactions (pre-exponential constants and activation energies) are defined based on the experimental data:

\[ R_{\text{ads}} = k_{\text{ads}}C_{\text{NH}_3}(1 - \theta); \quad (8) \]
\[ R_{\text{des}} = k_{\text{des}}\exp\left(-\frac{E_{\text{des}}(1 - \theta)}{RT_w}\right)\theta; \quad (9) \]
\[ R_{\text{oxid}} = k_{\text{oxid}}\exp\left(-\frac{E_{\text{oxid}}}{RT_w}\right)\theta; \quad (10) \]
\[ R_{\text{red}} = k_{\text{red}}\exp\left(-\frac{E_{\text{red}}}{RT_w}\right)C_{\text{NO}}\theta; \quad (11) \]
\[ \theta = \frac{C_{\text{NH}_3}(s)}{\Omega}. \quad (12) \]

where \(k_i, E_{a,i}\) – pre-exponential constant and activation energy of the i-th reaction, respectively; \(R\) – universal gas constant; \(T_w\) – catalyst wall temperature; \(\theta\) - NH\(_3\) fraction loading onto the catalyst surface; \(\Omega\) – number of reaction-sites per volume of washcoat.

Species equations of NH\(_3\) and NO in the flowing gas and NH\(_3\) adsorbed on the catalyst surface are written as:

\[ \frac{\partial C_{\text{NH}_3}}{\partial z} = \frac{s}{A_gu}(\text{R}_{\text{ads}} - \text{R}_{\text{des}}); \quad (13) \]
\[ \frac{\partial C_{\text{NO}}}{\partial z} = \frac{s}{A_gu}(\text{R}_{\text{red}}); \quad (14) \]
\[ \frac{\partial \theta}{\partial t} = (\text{R}_{\text{ads}} - \text{R}_{\text{des}} - \text{R}_{\text{red}} - \text{R}_{\text{oxid}}). \quad (15) \]

where \(s\) - number of reaction-sites per channel length; \(z\) - coordinate along the channel centerline.

Gas and catalyst wall temperatures are calculated based on the energy equations:
\[
\frac{\partial T_g}{\partial z} = -\frac{h P}{\rho_g A_g C_p g u} (T_g - T_w); \\
\frac{\partial T_w}{\partial t} = -\frac{h P}{\rho_w A_w C_p w} (T_w - T_g)
\]

where \( T_g \) – gas temperature; \( h \) – convective heat transfer coefficient; \( P \) – cross section perimeter; \( \rho \) – density, \( A \) – cross section area; \( C_p \) – specific heat capacity; \( t \) – time.

Boundary conditions at each channel entrance are determined based on CFD results imported from ANSYS Fluent in the way shown in Figure 2. The procedure consists of the following steps: the import of gas flow parameters, the distribution at the catalyst inlet, the interpolation of imported data at channels corners, the averaging of values at channel corners.

![Figure 2](image)

**Figure 2.** Estimation of boundary conditions at channels inlet: a – import of CFD results, b – interpolation and the averaging procedure, c – obtained boundary conditions.

The upwind difference scheme is used to approximate space derivatives of the equations (13, 14, 16). This leads to a system of time-dependent ordinary differential equations that are solved using standard MatLab ode23s algorithm [25].

As a result of numerical simulations, the concentrations of NO\(_X\) along the catalyst channels and at the catalyst outlet are derived. They are further used to calculate the NO\(_X\) conversion efficiency.

3. Results and discussion

Using the described integrated approach, a simple SCR system was investigated. It represents an insulated exhaust pipe equipped with a SCR catalyst block and UWS injector according to Figure 3.

![Figure 3](image)

**Figure 3.** Computational domain.

At the exhaust pipe inlet, the hot air with an average velocity of 13.2 m/s and a temperature of 261 °C was supplied. It contained 205 ppm of NO, 18% of O\(_2\), 3% of H\(_2\)O [26].
The SCR catalyst block represented the cylinder honeycomb-type substrate with flow-through square channels. It had an outer diameter of 278 mm, cell density of 300 cpsi and a wall thickness of 0.2 mm. The substrate was made of synthetic cordierite with the following properties: density of 2100 kg/m$^3$, specific heat capacity of 1000 J/(kg·K), thermal conductivity of 2 W/(m·K). The catalyst material was Cu-zeolite. Kinetic parameters of catalytic reaction rates (8 - 11) used in the simulations were taken from the experimental work [27] and are summarized in Table 1.

### Table 1. Kinetic parameters of catalytic reaction rates [27].

| Reaction               | Pre-exponential constant, $k_i$ | Activation energy, $E_{a,i}$ [J/mol] |
|------------------------|---------------------------------|--------------------------------------|
| Ammonia adsorption     | 0.25                            | 0                                    |
| Ammonia desorption     | $1.4 \times 10^6$               | $1.01 \times 10^5$                   |
| Ammonia oxidation      | $2.2 \times 10^8$               | $1.4 \times 10^4$                    |
| NO reduction           | $1.5 \times 10^8$               | $7.9 \times 10^4$                    |

In ANSYS Fluent, the catalyst substrate was defined as a porous medium in terms of porosity equal to 0.7, inertial and viscous resistance coefficients were 20.414 m$^{-1}$, $3.846 \times 10^7$ m$^{-2}$ in the direction of flow and 20414 m$^{-1}$, $3.846 \times 10^{10}$ m$^{-2}$ in the directions perpendicular to the flow. The resistance coefficients were calculated based on experimental data [20] to reproduce actual pressure losses across the monolith.

At the pipe centerline, the urea water solution (32.5% urea by weight) was injected along the gas flow. The spray is described as a solid cone pattern with an injection cone angle of 40°. The initial droplet size distribution was specified using the Rosin–Rammler function with $n = 2$, $d = 20$ taken from the experimental work [28]. The initial droplet velocity was 10 m/s. The UWS mass flow rate was 45 mg/s. It was calculated based on the assumption of equal molar concentrations of NH$_3$ and NO with complete urea decomposition.

Results of the numerical simulation of gas flow dynamics throughout the SCR system and the UWS injection performed in ANSYS Fluent are shown in Figure 4.

**Figure 4.** Numerical simulation of a SCR system in ANSYS Fluent: a – velocity profile and UWS injection; b – distributions of velocity, NH$_3$ molar concentration and NH$_3$ to NO ratio at the catalyst inlet.

Figure 4a shows that not all of the injected UWS had evaporated and converted to ammonia before the catalyst inlet. The reason is the insufficient length of the mixing channel for the specified gas flow and injection conditions. As a result, the average concentration of produced NH$_3$ (0.0021 mol/m$^3$) at catalyst inlet was less than that of NO (0.0045 mol/m$^3$). The large diffuser cone angle and axial position of the injector led to flow maldistribution at catalyst inlet that was especially pronounced for gas velocity and NH$_3$ concentration (Fig. 4b). The concentration distributions of other species (NO, O$_2$, etc.) as well
as the gas temperature profile were nearly uniform.

The simulation of catalytic reactions was carried out using the developed mathematical model implemented in MATLAB. The obtained CFD results at the catalyst inlet were used as boundary conditions. The gas flow velocity and ammonia concentration profiles were imported to the model according to the described procedure shown in Figure 2. The other flow parameters were considered to be uniformly distributed and were set equal to their average values. As a result of numerical simulations, the change in the concentration of NO along the catalyst was derived (Fig. 5).

![Figure 5](image)

**Figure 5.** Contours of the NO concentration within the SCR catalyst.

Due to the maldistribution in the NH$_3$ concentration at the catalyst inlet (Fig. 4b), NOx conversion dominantly occurred in the central region of the converter (Fig. 5). The concentration of available NH$_3$ decreased in radial direction that led to reduction of NOx conversion efficiency to near zero values on the periphery of the block. The overall catalyst performance was analyzed by the average conversion of NOx along the catalyst length (Fig. 6). To assess the effects of the incomplete UWS conversion and flow maldistribution at the catalyst inlet on SCR performance, additional catalyst simulation with the uniform flow distribution and equimolar concentrations of NH$_3$ and NO was performed. The results were compared with the previous case (Fig. 6).

![Figure 6](image)

**Figure 6.** The change in the average NO concentration along the SCR catalyst under the actual and uniform flow distribution at the catalyst inlet.
The comparison of results showed a significant reduction in the NOx conversion efficiency under non-uniform conditions. The overall SCR performance for this case was 15.8% that almost 5 times less than for uniform conditions (75.9%). To increase the flow uniformity at the catalyst inlet, static mixers are commonly installed in real constructions of SCR systems [29]. They improve the flow mixing by creating a turbulent flow as well as the UWS evaporation breaking up the spray into smaller droplets.

Thus, processes occurring upstream of the catalyst have a great impact on its efficiency and should be carefully modeled. For this purpose the suggested integrated approach can be applied.

4. Conclusion
The integrated approach for the complete simulation of SCR systems based on the coupling of ANSYS Fluent with the in-house multichannel 1D catalyst model was developed. The simulations of the urea-SCR system using the suggested approach were performed. The results of CFD numerical simulations indicated the incomplete UWS conversion to NH3 as well as the presence of flow maldistribution in NH3 concentrations and gas velocity at the catalyst inlet. The effect of these factors on the performance of the SCR catalyst was analyzed by comparing NOX conversion efficiency with that at uniform flow distributions and equimolar concentrations of NH3 and NO at the catalyst inlet. For that purpose, the developed multichannel 1D catalyst model implemented in MATLAB was used. The comparison showed that results differed almost 5 times as compared to the lower value for non-uniform conditions. Thus, the suggested integrated approach has confirmed the ability to significantly improve the prediction accuracy with the acceptable level of computational efforts.

References
[1] Joshi A 2020 SAE Tech. Pap. (SAE International) 2479–2507
[2] Sharp C, Webb C C, Neely G, Carter M, Yoon S and Henry C 2017 SAE Int. J. Engines 10 1697–1712
[3] Zhao J, Grekhov L, Ma X and Denisov A 2020 Appl. Therm. Eng. 179 115699
[4] Zhao J, Grekhov L and Yue P 2020 Int. J. Automot. Technol. 21 649
[5] Kuznetsov A, Kharitonov S and Ryzhov V 2018 J. Eng. Gas Turbines Power 140(12)
[6] Kamaltdinov V G, Markov V A, Lysov I O, Zherdev A A and Furman V V 2019 Energies 12 2643
[7] Markov V A, Devyanin S N and Kamaltdinov V G 2020 Lect. Notes Mech. Eng. 845–853
[8] Koebel M, Elsener M and Kleemann M 2000 Catal. Today 59 335
[9] Johnson T V 2015 SAE Int. J. Engines 8 1152
[10] Birkhold F, Meingast U, Wassermann P and Deutschmann O 2007 Appl. Catal. B Environ. 70 119
[11] Falti R and Mutyal J 2012 SAE 2006 World Congr. Exhib. 40
[12] Bielaczyc P and Woodburn J 2019 Emiss. Control Sci. Technol. 5 86
[13] Echtel H, Schöffel S, Wenninger G, Fischer S, Lauer T and Möltner L 2010 MTZ Worldw. 71 54
[14] Chi J N and Dacosta H F M 2005 SAE Tech. Pap. (SAE International)
[15] Mauviot G, Berr F, Raux S, Perretti F, Malbec L M and Millet C N 2009 Oil Gas Sci. Technol. 64 285
[16] Praveena V and Martin L J 2018 SAE Tech. Pap. (SAE International)
[17] Nova I and Tronconi E 2014 Urea-SCR technology for deNOx after treatment of diesel exhausts vol 5 (New York: Springer)
[18] Wurzenberger J C and Wanker R 2005 SAE Tech. Pap. (SAE International)
[19] ANSYS Inc. 2013 ANSYS Fluent Theory Guide 15317
[20] Dammalapati S, Aghalayam P and Kaisare N 2019 Ind. Eng. Chem. Res. 58 20247
[21] Kim J Y, Ryu S H and Ha J S 2004 Proc. 2004 Fall Tech. Conf. ASME Intern. Combust. Engine Div. (American Society of Mechanical Engineers) 165–170
[22] O’Rourke P J and Amsden A A 1987 SAE Tech. Pap. (SAE International)
[23] Shirodkar V S 2016 *Front. Heat Mass Transf.* 7
[24] Blinov A, Malastowski N and Myagkov L 2019 *E3S Web Conf.* 06013
[25] Shampine L F and Reichelt M W 1997 *SIAM J. Sci. Comput.* 18 1
[26] Nadareishvili G G 2020 *Int. J. Emerg. Trends Eng. Res.* 8 119
[27] Na H, Reed D, Annaswamy A, Laing P M and Kolmanovsky I 2011 *SAE 2011 World Congr. Exhib.*
[28] Spiteri A, Eggenschwiler P D, Liao Y, Wigley G, Michalow-Mauke K A, Elsener M, Kröcher O and Boulouchos K 2015 *Fuel* 161 269
[29] Mehdi G, Zhou S, Zhu Y, Shah A H and Chand K 2019 *Processes* 7