

Modeling of a DOC SCR-F SCR Exhaust Line for Design Optimization Taking Into Account Performance Degradation Due to Hydrothermal Aging

Simon Dosda and David Berthout
IFP Energies nouvelles, Institut Carnot IFPEN TE, France

Gilles Mauviot
Renault SA

Adeline Nogre
D2T

ABSTRACT

With the upcoming Euro 6c emission regulations, the performance of Diesel exhaust lines needs to be improved to meet NO_x and soot emission targets. A promising exhaust line architecture to reach these requirements is the association of a Diesel Oxidation Catalyst (DOC), a Selective Catalytic Reduction coated on a particulate filter (SCR-F) and a Selective Catalytic Reduction (SCR) catalyst. To develop this system, the car manufacturers have to face several challenges. One of the first is the design of the exhaust line volumes, which has a strong impact on the light-off temperatures of the catalysts and so on system performance. Then, urea injection has to be optimized with an adapted control system to maximize NO_x reduction while keeping low tailpipe ammonia emission. Moreover, performance degradation of catalysts due to harsh exhaust conditions during vehicle life time have to be detected by OBD system.

To meet these challenges, system simulation approaches allow to create virtual exhaust lines and perform, in a quite short time and with reduced cost, a large number of tests over a wide range of operating conditions. In this work, an exhaust line composed of a DOC, a close coupled SCR-F and an underfloor SCR has been modeled using both synthetic gas bench and engine bench tests data. From this reference, hydrothermal aging impact on the SCR catalysts has been studied on synthetic gas bench using several aging conditions. Using these experimental data combined with results from previous work on catalyst aging modeling, a hydrothermal aging model has been developed to represent the evolution of the SCR catalyst performance. This model has then been validated on data from a complete exhaust line having aged SCR catalysts. To finish, different changes in the exhaust line geometry have been investigated by simulation, moving the under-floor SCR to a close coupled position with volume variations. The impact of these modifications on the exhaust line performance has then been confirmed using test bench results.

This methodology has proven to be very efficient in modeling a complete Diesel Euro 6c after-treatment exhaust line, allowing to easily take into account the impact of exhaust line geometry or catalyst aging in order to optimize this complex system.

CITATION: Dosda, S., Berthout, D., Mauviot, G., and Nogre, A., "Modeling of a DOC SCR-F SCR Exhaust Line for Design Optimization Taking Into Account Performance Degradation Due to Hydrothermal Aging," *SAE Int. J. Fuels Lubr.* 9(3):2016, doi:10.4271/2016-01-2281.

INTRODUCTION

In the frame of the upcoming Euro 6c regulations for Diesel engines, both NO_x and PM emissions have to reach a very low level in a wide range of operating conditions. The new WLTC driving cycle and the regulation of Real Driving Emissions, including conformity factors, require developing a highly effective aftertreatment system while fulfilling CO₂ fleet emissions targets [1] [2]. To meet this objective, one envisaged exhaust layout solution is the association of a Diesel oxidation catalyst (DOC) or a NO_x storage catalyst (NSC) with the combination of a close-coupled ammonia selective catalytic reduction coated on a particulate filter (SCR-F) and an additional SCR [3] [4] [5]. In recent years, SCR-F also called SDPF or SCRoF by different

manufacturers received considerable attention due to the combination of two functionalities, NO_x conversion and filtration of soot, in one system. The benefits of this component is to take advantage of the high temperatures due to its close-coupled position to provide higher NO_x conversion [3] [6]. However some drawbacks as the competitive NO₂ consumption by SCR and soot, the effects of soot loading on NO_x conversion and the regeneration of the filter, need to be considered in the development of a SCR-F and in its exhaust line integration. NH₃-SCR and soot interactions and their influence over the performance have been addressed in the literature [7] [8] but this work is still in progress.

Advanced control laws are also needed for the management of the distribution between NH_3 stored in the SCR-F and NH_3 slip used for the passive SCR device while the emission of tailpipe NH_3 slip shall remain below the regulation limit in any driving condition. Thermal durability [9] and aging evolution prediction are also a major concern for on board diagnostic (OBD) systems. Consequently, the complexity of the exhaust line and its management require specific simulation tools to design and optimize the catalysts behavior. Different approaches have been developed to model SCR-F systems [7] [10] [11], but classical approaches are often Cpu time consuming making it impossible to use for control command development and validation.

In this paper, a simulation platform of an exhaust line comprising a DOC, a SCR-F and an additional SCR has been built for control and design applications keeping a physical approach. A compromise between robustness, accuracy and Cpu time had to be found to be able to take into account geometry variations and hydrothermal aging.

REFERENCE EXHAUST LINE

Experimental Setup

The reference exhaust line is composed of a 1.6L Diesel Oxidation Catalyst (DOC), a 2.8L close coupled Selective Catalytic Reduction coated on a particulate filter (SCR-F) and a 1.4L under-floor Selective Catalytic Reduction catalyst (SCR). The DOC is provided with a HC storage function and a NO oxidation function designed to achieve an optimal NO_2/NO_x ratio upstream of the SCR-F. Both the SCR-F and the SCR are Cu-Zeolite type. A schematic representation of the aftertreatment system is presented in Figure 1.

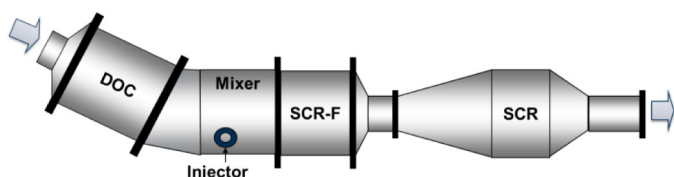


Figure 1. Schematic diagram of the reference exhaust line, composed of a DOC, a SCR-F and an under-floor SCR

Pollutants are measured at each end of the exhaust line catalysts. Pollutants upstream and downstream the DOC are measured using two 7 gas analyzers, while pollutants downstream SCR-F and SCR catalysts are measured using respectively a FTIR and a FTUV analyzer, allowing to measure the ammonia slip downstream both SCR catalysts.

Simulation Platform Setup

System simulation provides effective tools to model a complete exhaust line system while keeping a reasonable simulation time. Expectations for this kind of simulator is to provide a good estimation of pollutant concentrations along the exhaust line while keeping a simulation time below real time.

The simulation platform was composed of a physical exhaust line model set up using the IFP-Exhaust library of LMS Imagine.Lab AmesimTM software [12], coupled with a control platform in Matlab/Simulink®.

The exhaust line simulation platform is shown on Figure 2.

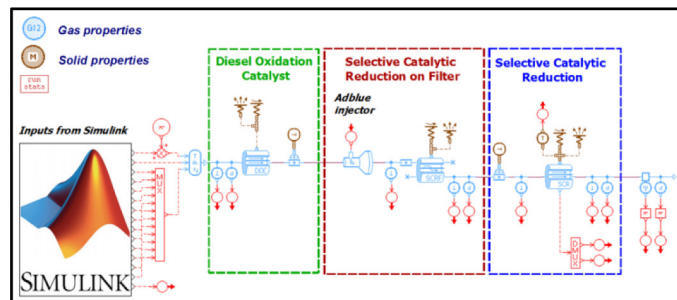


Figure 2. Simulation platform of the aftertreatment system in LMS Amesim

For each catalyst, calibration of the kinetic parameters has been done using synthetic gas bench (SGB) tests and is presented in the following sections.

Catalysts Calibration on Synthetic Gas Bench Tests

Diesel Oxidation Catalyst

To develop the model of the DOC, some driven parameters have been considered. Because of its position before the other catalysts, we focused on the accuracy of the NO_2/NO_x ratio, and the catalyst temperature mainly controlled by the exothermicity of HC oxidation and consequently on the HC adsorption/desorption function.

The model is considered as a 0D reactor [12], mass balance is considered for the different species using equation 1:

$$\frac{dm_j}{dt} = x_{in,j} \cdot dm_{in} - x_{out,j} \cdot dm_{out} + M_j \cdot \sum_i (v_{i,j} \cdot R_i) \quad (1)$$

Where:

- m_j is the mass of the species j ,
- $x_{in,j}$ and $x_{out,j}$ are the inlet and outlet mass fractions of the species j [-],
- dm_{in} and dm_{out} are respectively the inlet and outlet mass flow rates,
- M_j is the molar mass of the species j ,
- $v_{i,j}$ is the stoichiometric coefficient of the species j for the reaction i ,
- R_i is the rate of the reaction i .

To compute the gas and monolith temperatures, energy balances for the gas phase (2) and solid phase (3) are computed as follows:

$$\begin{aligned} \rho_g \cdot V_{open} \cdot Cp_g \cdot \frac{\partial T_g}{\partial t} &= dh_{in} - dh_{out} + h_g \cdot S_{geom} \cdot (T_s - T_g) \\ &+ \sum_k (dm_{store_k} \cdot h_k) \end{aligned} \quad (2)$$

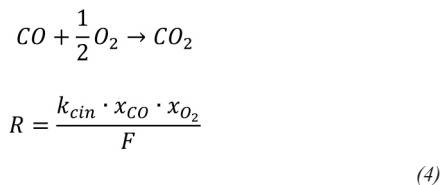
$$\begin{aligned} Cp_s \cdot \rho_{app} \cdot V_{cata} \cdot \frac{\partial T_s}{\partial t} &= dh_{ext} - h_g \cdot S_{geom} \cdot (T_s - T_g) \\ &- \sum_i (\Delta H_i \cdot R_i) - \sum_k (dm_{store_k} \cdot h_k) \end{aligned} \quad (3)$$

Where:

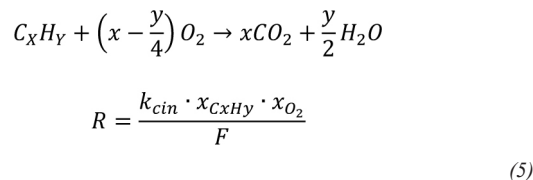
- V_{open} is the open volume of the monolith,
- V_{cata} is the total reactor volume,
- ρ_g and Cp_g are the density and the specific heat of the gas mixture,
- ρ_{app} is the catalyst density,
- Cp_s is the catalyst heat capacity,
- dh_{in} and dh_{out} are the enthalpy flow rate at inlet and outlet ports,
- dh_{ext} is the external heat flow rate,
- h_g is the convection heat transfer coefficient,
- S_{geom} is the geometric surface,
- T_s and T_g are respectively the monolith and the gas temperatures,
- $dm_{store,k}$ is the storage mass flow rate of the species k,
- h_k is the specific enthalpy of the gas k,
- R_i is the rate of reaction i,
- ΔH_i is the heat of the reaction i.

The reactions considered and associated reaction rates are the following:

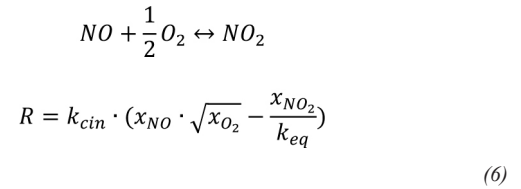
CO oxidation:



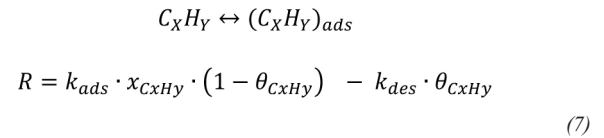
HC oxidation:



NO/NO₂ equilibrium:



HC storage:



Where:

- k_i is the rate constant of the reaction i,
- x_j is the molar fraction of the species j,
- θ_k is the fraction of adsorption site occupied for the species k,
- F is a resistance term taking into account the inhibition effects of CO, HC and NO.

In the reaction rates, the Arrhenius equation is used for the rate constants, k :

$$k_i = A_i e^{-\frac{E_i}{R \cdot T_{mono}}} \quad (8)$$

Where:

- A_i is the pre-exponential factor of the reaction i,
- E_i is the activation energy of the reaction i,
- R is the perfect gas constant,
- T_{mono} is the monolith temperature.

The parameters A_i and E_i need to be set to the experimental results. The calibration of these parameters has been processed by following the standard tuning synoptic presented in [Figure 3](#).

The sample used for SGB tests has a volume of 0.2 L. Upstream gas composition used for the light off tests is detailed in [Table 1](#). Upstream temperature rise from 80°C to 400°C with a slope of 20°C/min, while the space velocity is kept at a value of 55 000 h⁻¹. It is noted that space velocity is ratio of volumetric inlet flow rate divided by the entire catalyst volume. Results obtained on one of these tests are shown in [Figure 4](#).

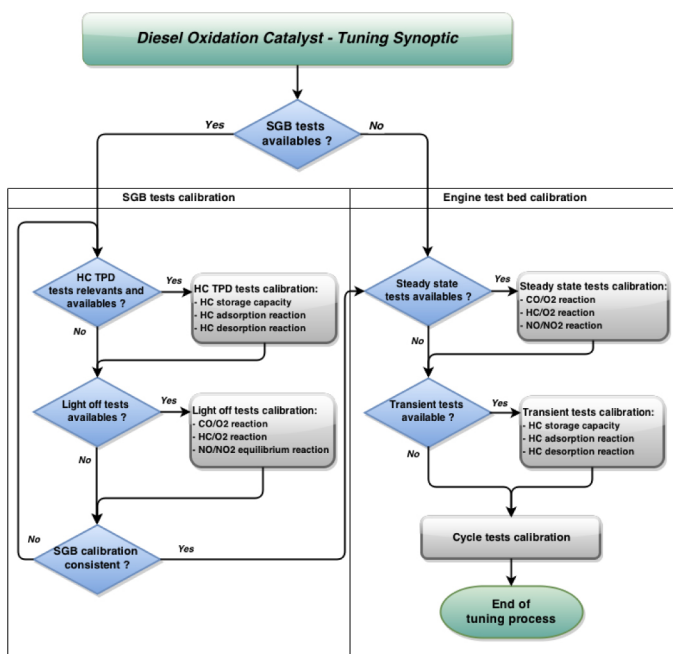


Figure 3. Diesel Oxidation Catalyst Tuning Synoptic

Table 1. SGB light off tests gas composition for the DOC

Species	Molar concentration	Unit
CO	1500	ppm
HC	430	ppmC
NO	100	ppm
CO ₂	4	%
H ₂ O	4	%
O ₂	14	%

The main difficulty for the calibration of the DOC reactions is related to the unburned hydrocarbons, as HC used on SGB tests are a mixture of 4 species : C₃H₆ (28%), C₇H₈ (20%), C₁₀H₂₂ (45%) and CH₄ (7%). All these species have different kinetics for oxidation and storage reactions. For this study, no specific HC-TPD tests were performed, so calibration was entirely done on light off tests. These tests show that more than half of the total HC are stored at low temperatures. These HC are released at a temperature around 150°C, with a released pick around 190°C. It can also be noticed that a part of the HC remains unoxidized, even at temperature higher than 500°C, which is mainly due to the presence of methane. These phenomena are well represented by the simulator, considering that HC are modeled as only one global species. This simplified modeling has been chosen in order to avoid having model results depending on HC composition, which is not known on chassis bench and varies depending on the operating point of the engine.

CO and NO reactions calibrations only involve one reaction and are consequently simpler to calibrate, and more robust to test bed changes as they are less impacted by the exhaust gases composition. Main impact from exhaust gases composition for these reaction comes from inhibition effects, which are taken into account by the model.

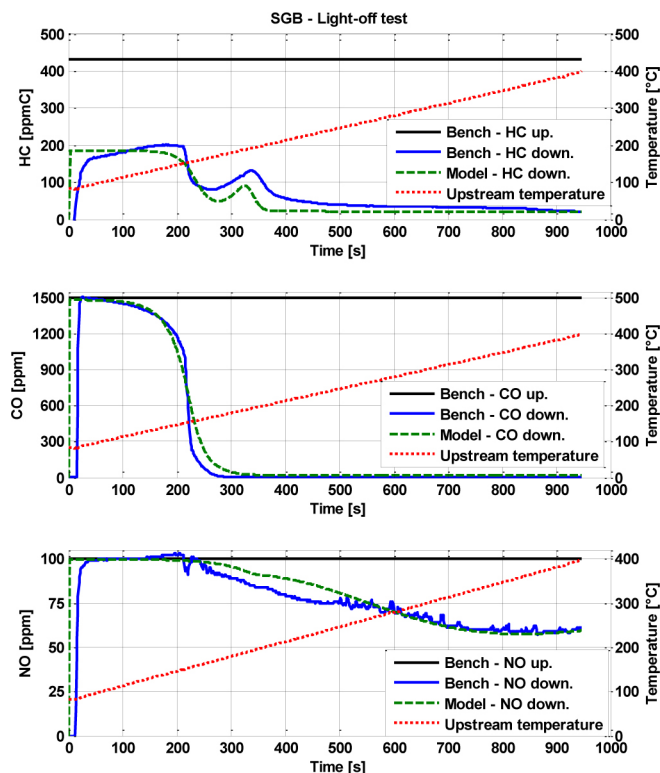


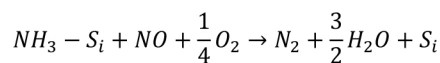
Figure 4. Simulation results (HC, CO and NO) on DOC SGB light-off test

Selective Catalytic Reduction Catalysts

The models of filter and monolith used for the NH₃-SCR need to be accurate regarding NH₃ slip and deNO_x efficiency with a reasonable Cpu time. They are inspired from our previous work on SCR [13, 14]. A two-site of adsorption for NH₃ model derived from Skarlis et al. [15] is used to take into account weakly and strongly bound NH₃. Standard, Fast and NO₂-SCR reactions are considered as well as oxidation reaction for NH₃. Concerning SCR-F, soot deposit and removal were not in the scope of this study, and therefore corresponding reactions have not been considered.

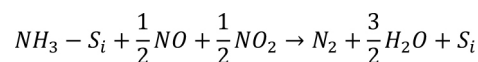
The reactions considered and the corresponding reaction rates are the following:

Standard SCR:



$$R = k_{cin} \cdot x_{NO} \cdot \theta_{NH_3-S_i} \quad (9)$$

Fast SCR:



$$R = k_{cin} \cdot x_{NO} \cdot x_{NO_2} \cdot \theta_{NH_3-S_i} \quad (10)$$