A PHYSICALLY-BASED, CONTROL-ORIENTED DIESEL OXIDATION CATALYST (DOC) MODEL FOR THE APPLICATIONS OF NO/NO2 RATIO ESTIMATION USING A NOX SENSOR

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ABSTRACT
NO and NO2 are generally considered together as NOx in engine emissions. Since NO2/NOx ratio is small in diesel engine exhaust gas, very often, existence of NO2 is ignored in studies/applications. However, current diesel aftertreatment systems generally include diesel oxidation catalysts (DOCs) at upstream of other catalysts such as diesel particulate filter (DPF) and selective catalytic reduction (SCR). DOC can significantly increase the NO2 fraction in the exhaust NOx. Because NO2 and NO have completely different reaction characters within catalysts, e.g. NO2 can assist DPF regeneration while NO cannot, and SCR De-NOx rate can be increased with higher NO2/NOx ratio (no more than 0.5), considerations of NO2 in aftertreatment systems are becoming necessary. Nevertheless, current onboard NOx sensors cannot differentiate NO and NO2 from NOx. This induces an interest in the method of estimating the concentrations of NO and NO2 in the exhaust gas by available measurements. In this paper, a physically-based, DOC control-oriented model which considers the NO and NO2 related dynamics and an engine exhaust NO/NO2 prediction method were proposed for the purposes of NO/NO2 ratio estimation in diesel engine aftertreatment systems, and the developed model was validated with experimental data.

INTRODUCTION
With increasing fuel prices, diesel engines have regained significant amount of attention in vehicle industry globally in recent years. However, problems of high NOx and particulate matter (PM) emissions have not been completely solved yet. Advanced aftertreatment systems such as diesel particulate filter (DPF) and selective catalytic reduction (SCR) systems can reduce more than 90% of particulate matter and NOx emissions. But the current and near future emission regulations worldwide strictly restrict the emissions to be decreased to even lower levels, which make diesel engine emission controls remain as great challenges in the field [1][2][3][4][5][6].

Among the current technologies, DPF and SCR are two of the most promising systems for particulate matter and NOx emissions reductions. DPFs, some are categorized as catalytic particulate filter (CPF) because of their catalytic regeneration capabilities, are capable of trapping a significant amount of PM from diesel engine emissions [7]. Particles captured by the DPF are oxidized by means of thermal assisted and NO2 assisted reactions (CPF) as listed in Eq. (1) and Eq. (2), and then be removed from DPF as CO and CO2 [7]. These processes are generally known as DPF regenerations.

\[ C + \left(1 - f_{\text{thermal}}\right)O_2 \rightarrow f_{\text{thermal}}CO + \left(1 - f_{\text{thermal}}\right)CO_2, \]  
\[ C + (2 - f_{\text{NO2}})NO_2 \rightarrow f_{\text{NO2}}CO + \left(1 - f_{\text{NO2}}\right)CO_2 + (2 - f_{\text{NO2}})NO, \]
where \( f_{\text{thermal}} \) and \( f_{\text{NO2}} \) are reaction fractions of CO/CO2 of the above processes.

SCR, on the other hand, typically utilizes ammonia (NH3) as the reductant to convert NOx emissions to N2 by the following catalytic reactions [8].

\[ 4NH_3 + 4NO + O_2 \rightarrow 4N_2 + 6H_2O, \]  
\[ 4NH_3 + 3NO_2 \rightarrow 7/2N_2 + 6H_2O, \]  
\[ 4NH_3 + 2NO + 2NO_2 \rightarrow 4N_2 + 6H_2O. \]  
It has been shown that the reaction of Eq. (5) is much faster than the standard reaction of Eq. (3) by one order of magnitude, and is even faster than the slow reaction of Eq. (4) [9].
Based on the above chemical reactions, it can be seen that NO2 plays an important role inside DPFs and SCRs. The presence of NO2 can speed up the DPF regenerations, and higher NO2/NOx ratio (no more than 50%) can also increase the SCR NOx conversion efficiency. But such features are no applicable to NO. Because of these differences, to improve the performances of DPF and SCR, a DOC is usually placed at the upstream of DPF and SCR to convert some of the NO in exhaust gas to NO2 by catalytic reactions, which leads to a higher NO2/NOx ratio comparing to that of the engine-out exhaust gas.

However, most studies assume NOx is completely NO, which is mostly due to the fact that current onboard NOx sensors can only measure the total NOx concentration and the majority of engine exhaust gas NOx is still NO [6][15][16][17][18]. This causes the problem that the NO2 related dynamics is hard to be considered in aftertreatment control designs. The issue is especially important for SCR control where urea injection rate should be precisely controlled to avoid undesired tailpipe NOx emissions and ammonia slip. To achieve this objective, knowledge of precise SCR dynamics is necessary, and several studies have pointed out the importance of separating NO and NO2 in SCR modeling and control [10][11][12][13][14]. Since these two species cannot be measured separately by current onboard NOx sensors, NO and NO2 concentrations need to be estimated by an observer and thus control-oriented models of the upstream catalysts are necessary. Some models which covered these dynamics are available in the literature. However, they are mostly numerical models and are difficult to be used for real-time control purposes [19][20][21][22][23][24].

The objective of this study is to develop a control-oriented model which can be used to estimate the concentrations of NO and NO2 in the aftertreatment systems from available measurements.

Due to the page limit, this paper only presents the studies of engine and DOC modeling. The DPF modeling study is presented in [26] and the NO and NO2 observer is presented in [27], all by the authors. These three papers together provide a complete study of NO and NO2 dynamics and estimations in engine-out-DOC-DPF aftertreatment system. The estimation results can be used for SCR control designs.

The rest of this paper is organized as follow. The setup of the engine and aftertreatment systems is explained at first. After that, the proposed DOC model, the engine-out NO and NO2 prediction method, and the corresponding assumptions are explained. Comparisons and analyses of experimental data and model validation results are discussed at the end and followed by conclusions.

**EXPERIMENTAL SETUP AND TEST PROCEDURE**

**Hardware**
The test bench includes: a V8 medium-duty diesel engine equipped with variable geometry turbo, dual-loop EGR, DOC and DPF which are stock aftertreatment devices of the engine, and two SCRs at downstream of the DOC/DPF, as can be seen in Figure 1 and Figure 2. The low pressure loop EGR was disabled and the SCR catalysts were bypassed during the tests in this study.

For the measurement systems, ETAS-INCA was used to log the ECU related signals, including engine speed, temperatures, MAF, fuel injections, and etc. Horiba MEXA 7500 was used to measure NO, NOx, and O2 concentrations at engine outlet, between DOC and DPF, and after DPF. The Horiba system was calibrated with sample gases and assumed to be accurate. Two Siemens VDO NOx sensors were used to measure the O2 and NOx concentrations upstream and downstream of the DOC/DPF. A differential pressure sensor was used between DOC and DPF to estimate the DPF loading. Also temperatures of critical points inside the engine, before DOC, between DOC and DPF, and after DPF were monitored by thermocouples. Besides, AVL 733S fuel balance and temperature control module were used to provide accurate fuel consumption measurement and consistent fuel supply temperature.

**FIGURE 1. ENGINE AFTERTREATMENT TEST SETUP**

**FIGURE 2. SCHEMATIC PRESENTATION OF THE TEST SETUP**
Test Procedures
The test procedure is shown in Table 1. Tests were started at engine warm-up conditions, where the engine coolant temperature reached 90 °C and DOC and DPF temperatures were higher than 160 °C. The engine was controlled to one of the desired operating points listed in Table 1 until the emission concentration measurements reached steady-states, and then the engine was stepped to the next point. The engine torque was measured by the torque sensor inside the dynamometer. But notice that the torque values listed in Table 1 are reference values, actual torques may slightly vary with experiments.

Due to the limitations that only one Horiba emission measurement channel is available and the NO and NOx cannot be measured simultaneously, four tests with the same engine operating conditions were implemented in order to have the NO/NO2 measurements at each sampling point as shown in Table 2. The Horiba sampling line was connected to different measuring points in each test for NO or NOx measurements, details of the tests are listed in Table 2.

The purposes of test 1 are to validate the NOx sensor measurements installed upstream and downstream of the DOC/DPF by the Horiba system and to validate one of the assumptions that the NOx concentration does not change through the DOC/DPF. Based on this assumption and under the condition that the NOx sensors are accurate, the NOx concentrations inside the DOC and DPF can be known by the NOx sensor reading. Assuming NOx consists of only NO and NO2 [25], NO concentrations of engine exhaust, after DOC, and after DP were measured by test 2 to test 4, and the corresponding NO2 concentrations can be calculated by the differences between NOx sensor and Horiba readings.

TABLE 1. EXPERIMENT TEST PROCEDURE

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TABLE 2. HORIBA MEASUREMENT POINTS AND SPECIES

<table>
<thead>
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NO/NO2 MODELING

DOC NO and NO2 Dynamic Model
Possible chemical reactions which can happen inside DOC are summarized below [7][24].

\[
\begin{align*}
CO + \frac{1}{2}O_2 & \rightarrow CO_2, \\
H_2 + \frac{1}{2}O_2 & \leftrightarrow H_2O, \\
C_3H_6 + \frac{9}{2}O_2 & \rightarrow 3H_2O + 3CO_2, \\
NO + \frac{1}{2}O_2 & \leftrightarrow NO_2, \\
H_2O + CO & \leftrightarrow H_2 + CO_2, \\
NO + CO & \leftrightarrow CO_2 + \frac{1}{2}N_2, \\
NO + H_2 & \rightarrow H_2O + \frac{1}{2}N_2, \\
9NO + C_3H_6 & \rightarrow 3CO_2 + \frac{9}{2}N_2 + 3H_2O, \\
C + \left(1 - \frac{f_{the}}{2}\right)O_2 & \rightarrow f_{thermal}CO + (1 - f_{the})CO_2, \\
C + (2 - f_{NO2})NO_2 & \rightarrow f_{NO2}CO + (1 - f_{NO2})CO_2 + (2 - f_{NO2})NO.
\end{align*}
\]

Since only NO and NO2 are considered in this study, the reactions of Eqs. (9)(11)(12)(13)(15) are taken into account in this study. Besides, because oxygen concentration is relatively high, it can be assumed unchanged after DOC. Also it is supposed that the reactions of Eq. (11)(12)(13) are slow and can be ignored. This assumption can be verified by the similar NOx concentrations measured at upstream and downstream of the DOC as shown in Figure 5. Reaction of Eq. (15) is also assumed to be slow comparing to Eq. (9), and it is ignored in the DOC modeling. This reaction, on the other hand, is considered in the DPF model. DPF has much higher soot (carbon, C) concentration and the majority of this NO2 assisted carbon oxidation process happens inside a DPF. Based on the above assumptions, the NO/NO2 dynamics inside DOC is able to be explained by Eq. (9). The reaction rates can be modeled by the modified Arrhenius equation [28].

\[
R_{oxi} = k_{oxi}T_{DOC}^{\alpha_{oxi}}\cdot e^{-\frac{E_{oxi}}{RT_{DOC}}} \cdot C_{NO,DOC} \cdot C_{O_2,DOC},
\]

\[
R_{deoxi} = k_{deoxi}T_{DOC}^{\alpha_{deoxi}}\cdot e^{-\frac{E_{deoxi}}{RT_{DOC}}} \cdot C_{NO_2,DOC},
\]

where \( R_{oxi} \) is the reaction rate of Eq. (9) in the forward direction (oxidation), \( R_{deoxi} \) is the reaction rate of Eq. (9) in the reverse direction (deoxidation), \( \alpha_{oxi}, \alpha_{deoxi}, \beta_{oxi}, \beta_{deoxi}, \kappa_{oxi}, k_{deoxi}, E_{oxi} \), and \( R \) are positive constants, \( C_{NO}, C_{NO_2}, \) and \( C_{O_2} \) are mole concentrations of NO, NO2, and O2, and \( T_{DOC} \) is temperature.
FIGURE 3. CSTR MODEL OF DOC

To avoid partial differential equations in the model and to keep the model with a reasonable complexity for control design applications, the DOC is assumed to be a continuous stirred tank reactor (CSTR) as shown in Figure 3. Based on the mass conservation law, the dynamics of NO concentration inside DOC can be modeled by the following equation.

\[
V_{DOC} \frac{dC_{NO,DOC}}{dt} = F_{in}C_{NO,in} - F_{out}C_{NO,DOC} - R_{oxi} + R_{deoxi},
\]  

(18)

where \( V_{DOC} \) is the DOC volume, \( T_{in} \) is the engine exhaust temperature, \( T_{DOC} \) is the DOC temperature, \( F_{in} \) is the engine exhaust gas volume flow rate, \( F_{out} \) is the DOC outlet gas volume flow rate, \( C_{x,in} \) are the engine exhaust mole concentrations of species \( x \), and \( C_{x,DOC} \) are the mole concentrations of species \( x \) inside DOC and are assumed to be the same with the DOC outlet concentration based on the CSTR assumption. Assuming the mass flow rates of DOC inlet and outlet are the same at steady-state and there is no backpressure caused by DOC, Eq. (18) can be rearranged as:

\[
\dot{C}_{NO,DOC} = \frac{F_{in}}{V_{DOC}} \left( C_{NO,in} - C_{NO,DOC} \right) - \frac{R_{oxi}}{V_{DOC}} + \frac{R_{deoxi}}{V_{DOC}}.
\]  

(19)

In the above DOC model, the DOC inlet gas temperature can be directly measured by the thermocouple upstream of the DOC, the DOC temperature is assumed to be the same as the DOC outlet gas temperature, which can also be directly measured by the thermocouple downstream of the DOC, and the engine exhaust gas volume flow rate can be estimated by the engine intake mass air flow rate and fuel injection rates [29]. Besides, the DOC oxygen concentration in Eq. (16) is assumed to be the same with the engine exhaust oxygen concentration measured by the upstream \( NO_x \) sensor since diesel exhaust is generally very lean and the amount of oxygen consumed by DOC can be ignored.

For the \( NO_2 \) dynamics, since \( NO_x \) composition is assumed to be NO and \( NO_2 \) and \( NO_x \) concentrations can be directly measured by sensors, the \( NO_2 \) concentration can be simply explained by the subtraction of NO from \( NO_x \) measurement:

\[
C_{NO_2} = C_{NO_x} - C_{NO}.
\]  

(20)

and the \( C_{NO_2,DOC} \) in Eq. (17) can also be obtained by above equation. of DOC, \( C_{NO,in} \), needs to be known, i.e. the engine-out exhaust NO concentration is needed. Since this value cannot be directly measured by onboard sensors, a model which can predict engine-out \( NO/NO_2 \) concentration from available sensors is required. The formation of NO during engine combustion has been extensively studied in [30][31][32][33][34]. It is understood that the formation includes many chemical reactions and can be affected by numerous environment factors, including some variables which are hard to be measured by onboard sensors. Such features make the development of engine-out NO and \( NO_2 \) control-oriented model challenging. Nevertheless, an interesting relation between engine-out exhaust \( NO_2/NO_2 \) ratio and total \( NO_x \) concentration was observed in this study.

Several steady-state measurements of engine-out exhaust \( NO_2 \) and \( NO_x \) concentrations were plotted in Figure 4. The 10 sets of data in Figure 4 represent 10 different engine speeds ranging from 750 RPM to 2900 RPM, and each speed comes with various engine loadings ranging from 0 Nm to 550 Nm. It can be found that the \( NO_2/NO_x \) ratio decreased consistently and exponentially with the engine-out exhaust \( NO_x \) concentration disregard of engine operating conditions. This empirical relation can be modeled by the following equation.

\[
C_{NO_2,eng} = C_{NO_x}k_1e^{-k_2C_{NO_x}},
\]  

(21)

where \( C_{NO_2,eng} \) and \( C_{NO_x} \) are the engine-out exhaust \( NO_2 \) and \( NO_x \) concentrations, and \( k_1 \) and \( k_2 \) are positive constants. Since \( NO_x \) concentration can be directly measured, engine-out NO and \( NO_2 \) concentrations can be predicted by this equation.

This empirical relation, even though cannot be explicitly explained, is illustrated by the following physical relations. The majority of \( NO_2 \) formation, based on the current understanding, is right after the engine combustion by the following reaction [35]:

\[
NO + H_2O \rightarrow NO_2 + OH,
\]  

(22)

where \( H_2O \) is generated during combustion and the formation rate mostly depends on the oxygen concentration [31]. Consequently, \( NO_x \) generated during combustion is mostly NO [32][33][34]. Since air fuel mixture is leaner at lower engine load, more \( H_2O \) is formed because of the higher oxygen concentration, and thus higher \( NO_2 \) fraction is seen at low engine load regions, i.e. when \( NO_x \) emission is low. On the other hand, at higher load, combustion mixture is richer and, because the combustion temperature is higher, oxidation of \( N_2 \) to NO is favored such that more NO is generated and less oxygen is available for the reaction of Eq. (22), which leads to less \( H_2O \) formation and thus a smaller fraction of \( NO_2 \).
Parameter Identification

Parameters of the DOC model in Eqs. (16)(17)(19) and the engine-out model in Eq. (21) were identified by minimizing the steady-state error between model predicted and Horiba measured NO concentrations at the engine operating conditions listed in Table 1. Genetic algorithm (GA) was used to solve the optimization problems. GA is a stochastic search based optimization method which is noted by the ability of solving problems with high nonlinearities and non-convex properties. Detail of GA is not discussed in this paper and can be found in [36][37], and some practical applications can be found in [38][39]. Comparison of identified engine-out NO and NO2 dynamic model and experimental data is shown in Figure 4. Results of the DOC model are shown in the experiment results section.

EXPERIMENTAL RESULTS AND ANALYSES

Figure 5 shows the Horiba and NOx sensor measurements from test 1 with speed ranges from 750 RPM to 2400 RPM. Each pulse in the figure corresponds to an engine speed in Table 1, increasing from 750 RPM to 2400 RPM. The engine load at each speed was increased from the lowest torque, e.g. 54 Nm in 750 RPM, to the highest torque, e.g. 271 Nm in 750 RPM. Torque was increased once the emission concentrations reached steady-states. Engine speeds were reduced to idle after the highest torques and steady-state were reached at each speed. Such test procedure and data presentation are applied to all the tests and the figures presented in the rest of this paper.

Two remarks can be made based on Figure 5. At first, since the two NOx sensors have been pre-calibrated to have the same readings under the same NOx concentration and the upstream NOx sensor and Horiba readings have close readings, assuming Horiba reading is accurate, it can be concluded that the two NOx sensor readings are trustful. Notice that the maximum NOx sensor reading is 1650 PPM, the slightly higher Horiba reading at the peak value during 750 RPM was due to the limitation of NOx sensor range. Secondly, while the NOx sensor readings upstream and downstream of DOC/DPF are very close, the assumption that the total NOx concentration does not change throughout the DOC/DPF is thus verified, i.e. reactions of Eq. (11)(12)(13) can be ignored. Besides, an interesting feature of the NOx sensors observed is the effect of soot concentrations. It can be seen that the upstream NOx sensor reading are very consistent to the other two readings at initial, but it showed increased offset after been used for a short period of time. It is believed that the NOx sensor can degrade very fast with the presence of high soot concentration, i.e. upstream of DPF. Figure 6 shows another set of test results from speed 750 RPM to 2000 RPM, where two different NOx sensors were used, and these sensors have been measuring the same points for a long period of time. It can be clearly seen that the upstream NOx sensor showed significant error. Based on this observation, only the NOx sensor downstream of DPF was used as the NOx emission measurement.

On the other hand, by comparing Figure 5 and Figure 6, it can be seen that even though the engine operating conditions and environmental factors were set to the same in each test, Figure 5 showed higher NOx emission during speeds of 750 RPM and 1000 RPM. It is because test 1 in Figure 5 started with a higher initial DPF loading and the engine introduced more oxygen (lower EGR) for DPF regeneration, which consequently generated higher NOx emissions. After the DPF load reached a lower value, it can be seen that the NOx emissions of the two tests are close, i.e. after 1200 RPM. Such phenomenon was also observed in other tests. More details are discussed in [26].
Figure 7 shows the measured steady-state engine-out exhaust NO and NO\textsubscript{x} concentrations by Horiba and NO\textsubscript{x} sensor in test 2, and the measured engine-out exhaust NO concentration is compared with the engine model in Eq. (20) and Eq. (21). It can be seen that the model predicted engine-out exhaust NO concentration is very close to the measured one by the Horiba gas analyzer. Some differences can be seen in the figure of 2900 RPM, but the maximum error is within the range of 10% which is acceptable.

Figure 8 shows the steady-state measurements of test 4, where the measured engine-out exhaust NO\textsubscript{x} concentration, estimated engine exhaust NO concentration, and measured NO concentration after DOC are shown. It can be clear seen that the NO concentrations after DOC were significantly reduced in most regions as expected, i.e. NO\textsubscript{2} were increased. However, at some low engine load points, i.e. lower exhaust temperature, the reverse reaction of Eq. (9) is faster than the forward reaction and more NO can be seen after DOC. It can be
explained by the facts that the reverse reaction favors lower temperatures as addressed in [40].

The steady-state data in Figure 8 from 750 RPM to 2400 RPM were used to identify the DOC model in Eqs. (16)(17)(19). Data of speeds 2600 RPM and 2900 RPM, on the other hand, were not used for parameter identification but used for model cross-validation. Figure 9 shows the simulation results of the identified model and the corresponding experimental data. As can be seen the estimated concentrations have high consistencies comparing to the measurements. The DOC NO model prediction root mean square error (RMSE) of the entire region is 24 ppm.

Figure 10 shows the comparisons of the simulated NO/NO$_2$ concentrations by the developed model with the experimental data which were not used in parameter identification. It can be seen that the model predicted results are very close to the actual values. The DOC NO model prediction root mean square error (RMSE) of the entire region is 29 ppm. Notice that NO$_2$ measurement is obtained by the subtraction of Horiba NO reading from NO$_x$ sensor reading. The negative NO$_2$ concentrations during idle speed are due to the more significant sensor errors when NO/NO$_2$ concentrations are lower.

CONCLUSIONS
A physically-based DOC dynamic model and an experimentally-based engine-out NO and NO$_2$ prediction method were proposed in this study for the objectives of diesel engine aftertreatment system NO and NO$_2$ concentration estimations. For the engine-out NO/ NO$_2$ estimation, a consistent relation between engine exhaust NO$_2$/NO$_x$ ratio and NO$_x$ emission concentration was observed. Based on this empirical relation, the engine exhaust NO concentration can be predicted by an onboard NO$_x$ sensor measurement. The DOC model was developed based on the physical insight into DOC characteristics. Possible chemical reactions were examined by experimental data. A control-oriented model with reasonable complexity and capable of capturing the main NO/NO$_2$ dynamics was presented. The model predictions showed high consistency comparing to the experimental data. This paper together with the correlative studies in our companion papers [26] and [27] provide an approach of estimating NO and NO$_2$ concentrations inside diesel engine aftertreatment systems, which is known of advantageous for the SCR urea-dosing control design.

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