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Design and experimental validation of a physics-based oxygen storage — thermal model for three way catalyst including aging

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A B S T R A C T

In this paper, a physics-based, oxygen storage-thermal model for a three way catalyst (TWC) is developed and experimentally validated. This model is then extended to account for aging impacts on the TWC. In order to identify the model parameters, a series of *ad hoc* experiments were designed to test the device over various engine operating conditions. Four TWCs of different ages were tested to investigate the effects of TWC aging on the oxygen storage dynamics. Results show that aging can be lumped within a single model parameter, referred to as *oxygen storage capacity*. Sensitivity analysis shows only negligible dependence of oxygen storage capacity on catalyst operating temperature. The comprehensive model is validated over real driving conditions for different catalyst ages. The developed model has the potential to enhance the design of optimization-control techniques for fuel consumption benefits and on-board diagnostics health measurement robustness.

1. Introduction

Tightening emissions standards and the 54.5 mpg fleet average fuel efficiency target on production vehicles by 2025 have spurred great interest in pursuing advanced control and optimization strategies to improve engine and aftertreatment systems performance (EPA, 2011). In order to meet new emissions regulation targets, the engine and the exhaust gas aftertreatment system are becoming increasingly complex through the introduction of technologies such as exhaust gas recirculation, particulate filters, etc.

For such complex systems, the *traditional* calibration approach based on open loop experimental maps has become impractical. The use of mathematical models to predict system and component behavior has become the preferred method to obtain increased performance, speed-up the control development process and reduce the calibration effort, Zhu, Wang, Sun, and Chen (2015). In modern Direct Injection (DI) engines, a Gasoline Particulate Filter (GPF) is added downstream of the TWC to limit particulate emissions. In this configuration, the TWC behavior influences the oxygen content and exhaust gas temperature flowing into the particulate filter, wielding influence over GPF soot regeneration events (Nicolin, Rose, Kunath, & Boger, 2015). Therefore, accurate prediction of TWC outlet oxygen concentration and temperature are crucial for estimation of GPF soot loading and regeneration.

Catalytic converters decrease toxic exhaust gas emissions by catalyzing a redox (oxidation or reduction) reaction. In particular, the TWC is used to simultaneously reduce nitrogen oxides (NOx), while and oxidize hydrocarbons (HC) and carbon monoxide (CO) are oxidized. Modern catalytic converters are capable of conversion efficiency (at steady state) approaching 100% when the normalized air–fuel ratio is controlled near stoichiometry, see Fig. 1.

The normalized air–fuel ratio is defined as:

$$\lambda = \frac{(A/F)_{actual}}{(A/F)_{stoich}} \tag{1}$$

where $(A/F)_{actual}$ is the actual air to fuel ratio and $(A/F)_{stoich}$ is the stoichiometric air–fuel ratio. With this formulation, when $\lambda < 1$, there is more fuel compared to the stoichiometric condition and the exhaust gas is said to be *rich*. When $\lambda > 1$ there is more air relative to the stoichiometric condition and the exhaust gas is referred to as *lean*.

Tailpipe emissions are highly affected by transient variations of the pre-catalyst air–fuel ratio that occur during real driving conditions. In order to compensate for transient λ deviations from stoichiometry, cerium is added to the TWC system and it functions as an oxygen buffer thanks to its ability to store and release oxygen, Kim (1982).

TWC performance is also dependent on catalyst temperature: the chemical reactions occurring in the catalyst guarantee a satisfactory pollutant conversion efficiency only above a certain threshold, usually around 300 °C (Fig. 2).

Catalyst temperature and oxygen storage level cannot be directly measured with commercial sensors. A cost effective option to monitor these quantities is through the use of properly designed modeling tools.

A wealth of literature has been published on after-treatment modeling for both diesel and gasoline engines (e.g. Chen and Wang, 2014,

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Nomenclature

TWC	Three Way Catalyst
PSO	Particle Swarm Optimization
PDE	Partial Differential Equation
t	Time [s]
λ	Normalized air–fuel ratio
T_{cat}	TWC solid phase temperature [K]
T_g	TWC gas phase temperature [K]
T_{amb}	External environment temperature [K]
T_{exh}	Exhaust gas temperature [K]
T_{lo}	Catalyst light off temperature [K]
$\dot{m}_{exh}$	Exhaust gas mass flow rate [kg s^{-1}]
ρ_g	Exhaust gas density [kg m^{-3}]
ρ_s	TWC solid phase density [kg m^{-3}]
c_{p_g}	Specific heat of the exhaust gas [J kg^{-1} K^{-1}]
c_s	TWC solid phase specific heat [J kg^{-1} K^{-1}]
λ_g	Exhaust gas conductivity [W m^{-1} K^{-1}]
λ_s	TWC solid phase conductivity [W m^{-1} K^{-1}]
h	Convective heat transfer coefficient [W m^{-2} K^{-1}]
h_{out}	Convective heat transfer coefficient with the environment [W m^{-2} K^{-1}]
ϵ	TWC open cross sectional area [0 – 1]
A_{cs}	TWC cross sectional area [m^2]
A_{geo}	TWC specific geometric area [m^{-1}]
A_{out}	TWC external surface [m^2]
V_{cat}	TWC volume [m^3]
D_{cat}	TWC diameter [m]
z	Axial dimension [m]
K_{reac}	Proportional constant of $\dot{Q}_{reac}$ [J kg^{-1} m^{-3}]
ΔH_i	Reaction enthalpy difference [J mol^{-1}]
i	Reaction index
j	Computational cell index
$\dot{Q}_{reac}$	Heat produced by reactions [W m^{-3}]
η	TWC efficiency [0 – 1]
R_i	Reaction rate for the ith reaction [mol m^{-3} s^{-1}]
k_i^f	Forward reaction rate ($i = 1, 2$)
k_i^b	Backward reaction rate ($i = 1, 2$)
K_i	Chemical equilibrium constant
ΔG_k	Gibbs free energy variation [J]
OSC	Total Oxygen Storage Capacity [mol m^{-3}]
u	Space velocity [m s^{-1}]
k_{rad}	Radial mass transfer coefficient [m s^{-1}]
c_0	Total concentration in the gas [mol m^{-3}]
M_{exh}	Average molar mass of composition [kg mol^{-1}]
E_i	Activation energy [J mol^{-1}]
A_i	Pre-exponential factor
ϕ	Normalized oxygen storage level [0 – 1]
Ce	Cerium chemical element symbol
$[Y]$	Concentration of species Y [mol m^{-3}]
$[Y]_g$	Concentration of species Y in the gas [mol m^{-3}]
$[Y]_{wc}$	Concentration of species Y in the washcoat [mol m^{-3}]

Depcik and Assanis, 2005, Guzzella and Onder, 2009, Katare, Patterson, and Laing, 2007 and Lepreux, Creff, and Petit, 2012). Despite the differences in the adopted technologies, the common modeling challenge lies in considering the complex mass transport, thermal and chemical dynamics typical of aftertreatment systems to predict, in a computationally efficient way, the macroscopic phenomena of interest.

Two modeling approaches are predominant in literature: physics-based modeling and empirical modeling. Physics-based models for TWC are developed in Auckenthaler (2005), Depcik and Assanis (2005), Montenegro and Onorati (2009), Oh and Cavendish (1982) and in Shamim,

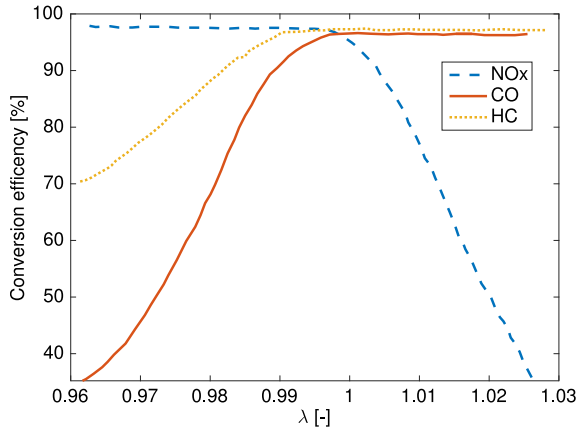

Fig. 1. Dependence of the TWC steady state conversion efficiency on the normalized air–fuel ratio λ.
Source: Figure reproduced from DTEC (2011).

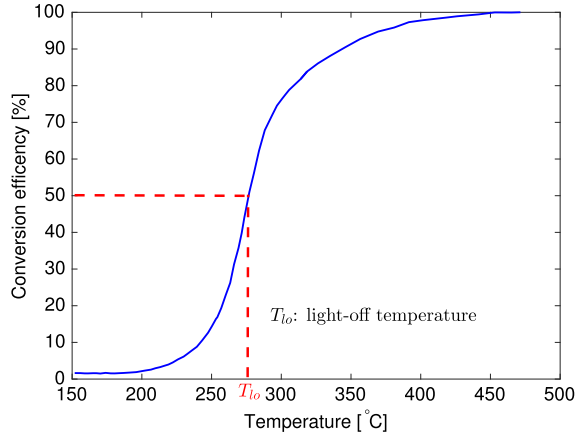

Fig. 2. The effect of temperature on TWC conversion efficiency, Bresch-Pietri, Leroy, and Petit (2013). The "light-off temperature" of the converter is defined as the temperature at which the reduction efficiency reaches 50%, Brandt, Wang, and Grizzle (2000).

Shen, Sengupta, Son, and Adamczyk (2002). In those works, the TWC operation is described using energy and mass balance equations coupled with a complex kinetics model to predict the conversion of undesired engine emission species within the catalyst. Estimation of the kinetic parameters can make identification of these models quite challenging. In addition, physics-based models are computationally complex, as they utilize non-linear coupled Partial Differential Equations (PDEs). The real time application of such models for temperature estimation and oxygen storage level control is highly limited.

In Kumar et al. (2012) and Möller, Votsmeier, Onder, Guzzella, and Gieshoff (2009), physic-based models are considered but with a reduced number of chemical species. Even though the computational burden of these models is reduced, their real-time application is still unfeasible since the chemical species concentrations at the catalyst inlet are required inputs and no gas analyzer is available for today's production vehicles. In Bickel, Odendall, Eigenberger, and Nieken (2017), a 1-D model with a simplified kinetic reaction is developed to study the oxygen storage capability of fresh catalysts under very different precious metal loadings. The resulting model was characterized using only five parameters. In Kiwitz, Onder, and Guzzella (2012), a simple physics-based oxygen storage model with a reduced number of species is presented, where the species concentrations are estimated from the wide-range lambda sensor measurements upstream the catalyst.