Modeling and Simulation of NO_x Abatement with Storage/Reduction Catalysts for Lean Burn and Diesel Engines

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Introduction

The unsolved problem of exhaust gas aftertreatment for diesel and lean burn engines still lead to enormous emissions of nitrogen oxides (NO_x). One of the most promising approaches is the NO_x Storage and Reduction Catalyst (NSR) which utilizes the NO_x storage on barium sites to form nitrates during the lean phase and their reduction to nitrogen in a rich atmosphere [1]. Detailed models, which are based on physical and chemical processes on the molecular level, are indispensable to exploit the full potential of this technique.

Experimental Work

The experimental work, which is accomplished at the Institute for Chemical Process Engineering at the University of Stuttgart, is based on well-defined model catalysts of monolithic structure and of varying complexity. Platinum and rhodium are chosen as noble metal components. The washcoat consists of γ-Al_2O_3 and for the more complex systems ceria and barium for the uptake of oxygen and nitrogen oxides were added. The investigations of the kinetics are carried out under isothermal conditions in a flat bed reactor [2] using a realistic model exhaust gas. Furthermore, lateral withdrawals allow the measurement of gas concentration profiles along the length of the catalyst. The experimental system is equipped with a fast responding mass spectrometer for the measurement of short lean/rich cycles.

Numerical Model

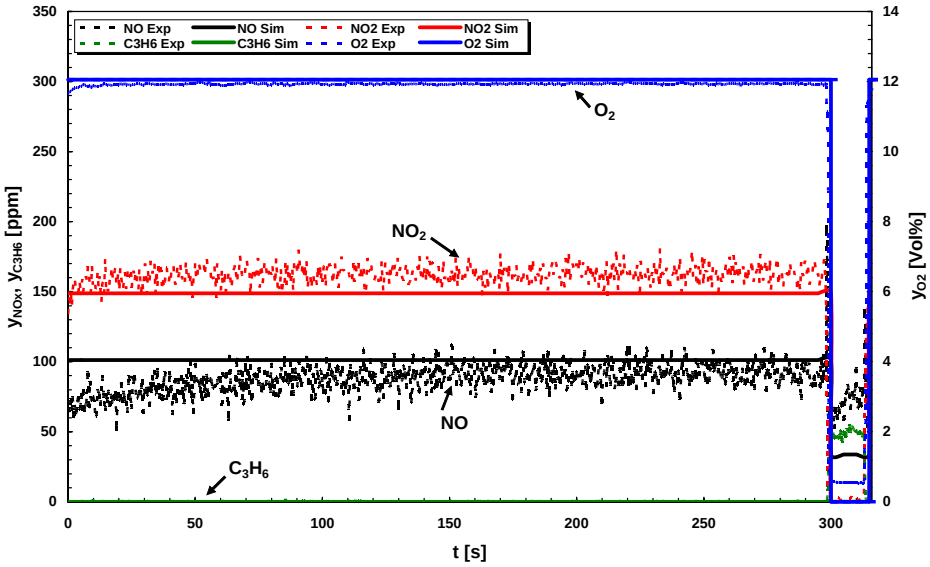
The numerical simulations are carried out using the software package DETCHEM [3, 4], which uses detailed reaction mechanisms for the conversion on the noble metals and the storage and reduction processes on the barium particles. DETCHEM is a FORTRAN based package that is designed to couple chemistry models with Computational Fluid Dynamics (CFD) programs. It applies hierarchically arranged detailed models from an atomic scale up to reactor scale. The core is a library for the description of species properties based on atomistic models and for reactions among gas-phase and surface species based on elementary step reaction mechanisms. Upon this, the two-dimensional flow field in a single channel is modeled using the boundary-layer assumption. Radial transport models include

composition dependent diffusion coefficients in the gas phase and an effectiveness factor approach for the washcoat. Inlet conditions and NO_x storage capacities of the single channel simulations vary in time.

The mechanism for the simulation of the oxidation and reduction processes is based on an elementary step reaction mechanism for platinum catalysts [4]. In this work, some reactions were modified and the mechanism was extended by reactions on support and storage media.

Results

Simulation results will be presented for $Pt/\gamma\text{-}Al_2O_3$, $Pt/Ba/\gamma\text{-}Al_2O_3$ and $Pt/Ce/Ba/\gamma\text{-}Al_2O_3$ catalysts at periodic lean/rich cycles ($0.9<\lambda<2.4$). The developed mechanism consists of more than 80 elementary step reactions for 35 chemical species, including reactions for oxygen storage as well as NO_x uptake.



Comparison of simulation and experimental results for a lean/rich cycle (300 sec/15 sec) of a $Pt/\gamma\text{-}Al_2O_3$ catalyst at 350°C

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Literature

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