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Competence Center Energy and Mobility

NEADS

Next Generation Exhaust Aftertreatment for Diesel Propulsion Systems

Scope of project

New SCR catalyst materials are investigated in order to achieve high reactivity and conversion also at low exhaust gas temperatures. In addition a ceramic foam based substrate is under development in order to replace the conventional diesel oxidation catalyst improving the performance and lifetime of the subsequent aftertreatment system (particulate filter and or SCR system).
The project is organised in 3 sub-projects: Sub-project I develops zeolites based catalytic materials for passenger cars (as well as medium and heavy duty vehicles). Sub-project II is aiming at the development of the micro reactor, while sub-project III investigates emission formation and reduction paths from the combustion through the aftertreatment systems.
The sub-projects in turn make use of the tools and analytics as well as knowledge developed and acquired in 3 tasks «new instrumentation for particle characterisation», «numerical simulation» and «atmospheric interactions».

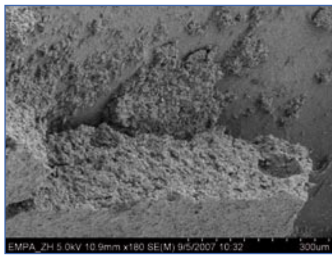
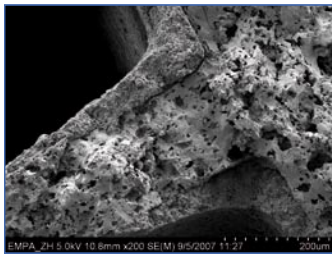


Fig. 1: SEM images of the ceramic foam based (above) and the extruded (below) catalyst.



Goals and activities of sub-projects

I: New generation of zeolite SCR catalysts

The main focus of the project is directed to the producing of stabilized iron exchanged zeolites with respect to hydrothermal aging. In order to do so we try to stabilize the zeolites by phosphating or exchanging with secondary metal ions. The measured SCR activity after hydrothermal aging is used as indicator for the stability. Promising new SCR-active metal-exchanged zeolites have been identified.

II: Ceramic foams as exhaust microreactor

Development of ceramic foam substrates will focus on several topics:

- Optimization of surface area, conversion properties, flow resistance and flow homogenization properties through specific and improved design as well as production of ceramic foams
- Further comparison of the conversion properties of ceramic foams and extruded monoliths on the available Compressed Natural Gas (CNG) engine with λ-sweeps. Several ceramic foam densities, as well as specific wash coat amounts in comparisons will be considered
- Demonstration of advantages of the ceramic foams as diesel oxidation catalysts upstream of a diesel particulate filter. Quantifications of the particulate filter loading with Computer Tomography
- Application of surface analysis techniques for analyzing soot and ash deposits on the particulate filter
- Prototyping and vehicle tests for Compressed Natural Gas (CNG) and Diesel applications

The conversion efficiency of ceramic foam catalysts in regard to unburnt HCs and NOx is higher in comparison to a 400cpsi extruded monolith catalyst of the same overall volume at low and moderate exhaust mass flow rates. At higher exhaust mass flow rates, on the other hand, the conversion efficiency of the extruded monolith catalyst is higher. The higher backpressure induced by the ceramic foams did not seem to affect the fuel consumption of the engine.

The conversion rates of the ceramic foam catalysts at deliberately lowered exhaust temperatures are often more than twice as high as the corresponding ones of the extruded monoliths. This is of importance since improved energetic

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management in modern engines aiming in improving fuel consumption results in decreasing exhaust temperatures.

III: Combustion interface

A fast phenomenological model, suitable for online calculations, is being developed to calculate the engine out soot emissions from common-rail diesel engines, using only engine parameters available from the engine control unit as inputs. Furthermore, the model should be capable of predicting the soot emissions during both steady state and transient engine operation. Such a model can provide detailed information as to the soot loading rates to which an aftertreatment will be exposed, and can be used to optimize regeneration strategies.

The steady state test rig is close to completion, though operation under transient conditions will be accomplished in the near future.

After parameterizing the model using evolutionary algorithms, the model was capable of reproducing the qualitative and quantitative soot emissions trends of a production diesel engine (DaimlerChrysler OM611) for operation with a standard diesel fuel under steady state conditions.

Goals and activities of tasks

A: New instrumentation for particle characterisation

Microspectroscopy

The necessary tools to establish X-ray transmission microspectroscopy for imaging phase separated nanostructured organic material will be developed with the aim to obtain a microscopic picture of soot particle properties at the nanoscale. The aim of the first year was to construct a final version of the environmental cell at the POLLUX microscope at SLS, to demonstrate its capabilities with a well established model system of atmospherically relevant particles and to obtain a range of C-edge and O-edge NEXAFS spectra from reference materials.

We constructed a gas cell that can be mounted at the POLLUX microscope at SLS to allow in-situ controlling of gas composition, pressure and temperature during analysis. As a proof of the capabilities of the new cell, we imaged 500nm diameter polystyrene spheres within the cell under vacuum and 1atm He, and obtained C K-edge spectra consistent with the literature.

At the same time we have used the standard sample holder setup to build up a data base of reference spectra at the C- and O- K edges for materials of relevance to this project, such as perylene, dihydroxybenzene, adipic acid, stearic acid, oleic acid and maleic acid.

W-ToF AMS

The high resolution time-of-flight aerosol mass spectrometer (W-ToF-AMS) is the only instrument on the market that allows the measurement of the elemental composition at a time resolution down to 1 second. This is important in the investigation of highly transient signals like the ones from a vehicle test bench. The higher mass resolution compared to a quadrupole aerosol mass spectrometer (Q-AMS) enables the determination of the chemical composition of the fragments and thus allows for a detailed description of the chemical processes occurring during atmospheric aging. This will be investigated in the PSI smog chamber.

New sensors

The focus of the activities is the investigation of new techniques for field measurement of particle emissions, which take into account the changed requirements of modern low emission engines. Conventional techniques as opacimeters can no longer be applied there. Methods based on electrical charging and optical techniques are investigated.

The «Diffusion Size Classifier DiSC», a device, charging particles by a unipolar diffusion charger (corona charger) and then classifying them by a diffusion separator is in a prototype stage.

First tests with the DiSC show a performance, which is adequate for monitoring engine emissions and ambient air particles. A commercial version of the instrument should be available 2008.

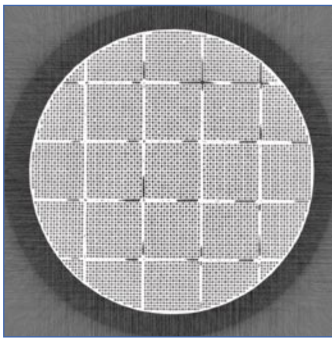

Fig. 2: Tomographic image of a cross section of a Diesel Particulate Filter (DPF) of an IVECO medium duty truck. Inflow channels with deposits can be clearly distinduished from outflow channels with almost no deposits.

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B: Numerical simulation

Task B focuses on modelling Diesel exhaust after-treatment by means of Selective Catalytic Reduction (SCR). Varying degrees of modelling complexities are reported in the literature, which depend on the specific physical processes considered. For the final project goal, i.e. prediction of the dynamic behaviour – with respect to upstream processes, catalyst conversion rates as well as device heat-up – of a full scale Diesel engine after-treatment system under transient operating conditions, a careful selection of the most suitable approaches is underway.

C: Atmospheric interactions

Chasing and dynamometer experiments

In order to assess the emissions of specific vehicles under real world conditions chasing experiments of specific vehicles with a mobile laboratory are planned. These chasing experiments will allow to determine emission measurements of vehicles in various driving conditions (e.g. different speeds, different loads) and to study fast transformation processes immediately after release and dilution.

The test bench and chasing results agree very well for the same conditions. Furthermore no nucleation mode (particle diameter $d < 30$ nm) was found which is consistent with other measurements when low sulphur fuel is used. In addition, these results demonstrate that dynamometric tests can simulate real world conditions very well in terms of physical parameters. The same will be tested for chemical properties.

Smog chamber studies

Experiments with the smog chamber will study interaction on a longer time scale. Secondary organic aerosol (SOA) from gaseous precursors emitted from various traffic types will be characterized and compared to the findings with SOA from biogenic precursors. Besides the overall yield, specific markers will be searched to distinguish between the SOA from the different sources. This will be used in an assessment of the importance of anthropogenic versus biogenic precursors.

Publications

- S. Brandenberger, O. Kröcher, A. Tissler, R. Althoff, Selective catalytic reduction of NOx by ammonia over metal exchanged zeolites, Catal. Rev., accepted with minor revisions.
- Dimopoulos P., Bach C., Vogt U. F., Herrmann K., Ceramic Foams as Catalyst Substrates: Pre-catalyst application homogenising the Exhaust Flow upstream of Aftertreatment Devices, SAE Paper 2007-24-0097.
- Winkler A., Hauert R., Dimopoulos P., Bach C., Aguirre M., Catalytic activity and aging phenomena of three-way catalysts in a compressed natural gas/gasoline powered passenger car, Applied Catalysis B, in review.
- Kirchen, P. and Boulouchos, K. Modellierung der rohen Russemissionen eines Dieselmotors. FVV Heft R 537, 2007.
- Kirchen, P. and Boulouchos, K. Russpartikel – Rohemissionsberechnung: Zwischenbericht. FVV Frühjahrstagung, 22.03.2007, Frankfurt, Germany.
- Kirchen, P. and Boulouchos, K. Measurement and Calculation of Engine-Out Soot Emissions. NEADS Workshop 13.4.2007, PSI, Villigen.
- Fierz, M., H. Burtscher, P. Steigmeier, M. Kasper, Field measurement of particle size and number concentration with the Diffusion Size Classifier (DiSC), SAE, 08PFL–484 (2007).
- Duplissy, J., M. Gysel, M.R. Alfarra, J. Dommen, A. Metzger, A.S.H. Prevot, E. Weingartner, A. Laaksonen, T. Raatikainen, N. Good, S.F. Turner, G. McFiggans, U. Baltensberger, The cloud forming potential of secondary organic aerosol under near atmospheric conditions, Gwophys. Res. Lett., in press, 2008.

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