New Strategies for Particulate Emission Reduction of HD Vehicles

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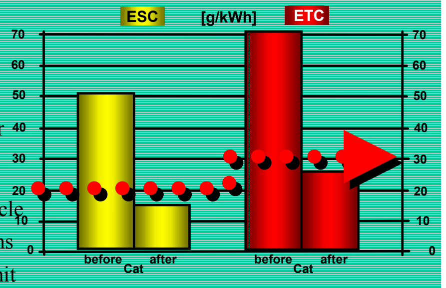
PM-Kat® - DPM Removal Approach

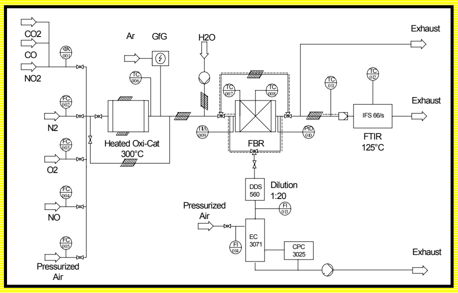
- Continuous deposition and volatilisation of soot particles by oxidation with NO_2/O_2 in filterless catalyst structures
- Low pressure drop, < 80 mbar
- No danger of clogging
- Reduction of heavy duty vehicle (HDV) diesel engine emissions below EURO IV emission limit values



- Oxidation reactions:

$$C_{(s)} + 2NO_{2(g)} \rightarrow CO_{2(g)} + 2NO_{(g)}$$
$$C_{(s)} + NO_{2(g)} \rightarrow CO_{(g)} + NO_{(g)}$$
$$C_{(s)} + \frac{1}{2}O_{2(g)} \rightarrow CO_{(g)}$$
$$C_{(s)} + O_{2(g)} \rightarrow CO_{2(g)}$$

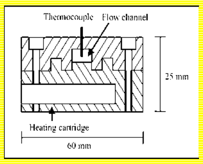
Model Catalytic Converter System

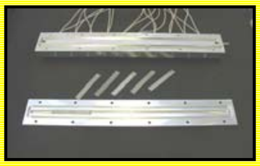


- Simultaneous investigation of soot deposition and oxidation
- Application of different model soots (spark discharge, hexabenzocoronene) and real diesel soot (HDV, LDV)
- Particle number concentration up to 3 x 10^7 cm^{-3}
- Particle size distribution measurements with Scanning Mobility Particle Sizer (SMPS)
- Particle mass concentration measurements with Photoacoustic Soot Sensor (PASS)
- Soot mass concentration up to 2.5 mg m^{-3}
- Multicomponent gas analysis with FTIR, Bruker IFS 66/s (LOD_{CO} = 0.5 ppm, LOD_{CO_2} = 0.15 ppm)

Flat Bed Reactor (FBR)

- A = 10 x 6 mm²
- L = 300 mm
- High flexibility
- 25 to 600°C, PID controlled
- GSV between 10,000 and 300,000 h^{-1}
- measured Δp in agreement with full size catalyst





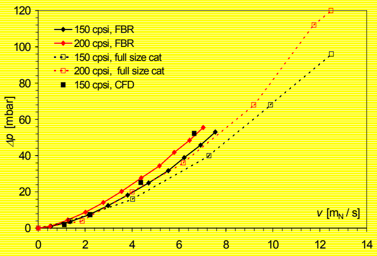


Figure 1. Pressure drop Δp measured in the model catalytic system (FBR) and in a full size catalyst for two different cell densities. Comparison with Δp determined from CFD simulations.

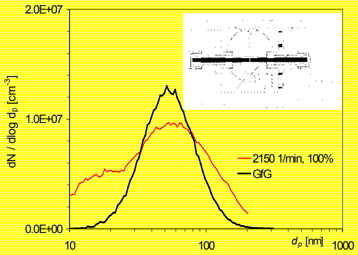


Figure 2. Particle size distribution for an HDVengine (ESC point 10) and a comparable spark discharge soot model aerosol (GfG 1000, Palas GmbH, Karlsruhe, Germany).

Deposition Structures

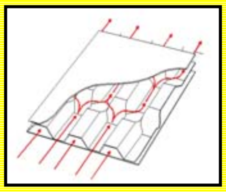


Figure 3. Schematic drawing of the PM-KAT® sandwich structure consisting of alternatingly arranged flat and corrugated stainless steel foils which provides favourable flow patterns and high resistance to thermal and mechanical stresses.

Carbon Mass Balance

$$\frac{dm_C}{dt} = \dot{V}\,T(d_p)\,c_{m,C} - m_C k_{diff} - m_C k_{reentr}$$

- (Size resolved) deposition efficiency → SMPS and PASS
- Soot oxidation kinetics → FTIR
- Reentrainment → PASS

Goal: Phenomenological model to describe the soot particle deposition, oxidation and reentrainment processes occuring in the catalyst structure.

Acknowledgements

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Particle Deposition Behaviour

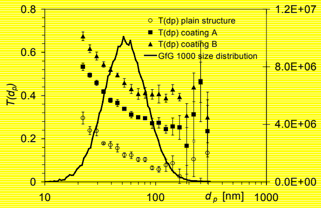

Figure 4. Size resolved deposition efficiency $T(d_p)$ for the plain catalyst structure and two different coatings.

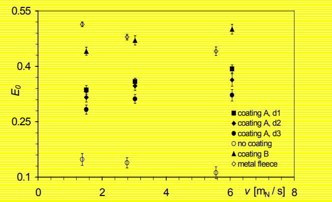

Figure 5. Number weighted mean deposition efficiency E_0 for the plain structure, 4 different coatings and an alternative approach based on a metal fleece – steel foil combination.

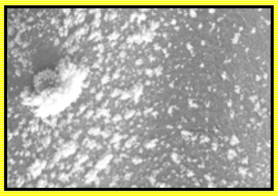

Figure 6. LDV-Soot accumulation at the microsphere side exerted to the exhaust flow.

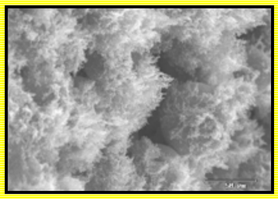

Figure 7. LDV-Soot deposition on the microsphere coated catalyst structure.

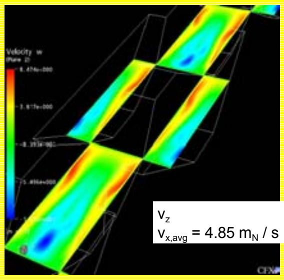

Figure 8. Vertical velocity component v_z determined by a 3D-CFD-simulation (CFX 5.6) of the catalyst structure.

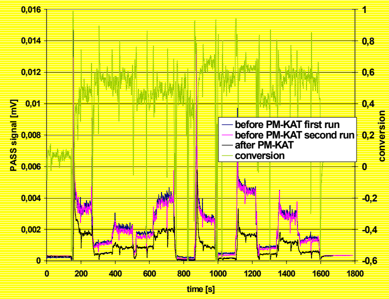

Figure 9. Soot concentration before and after the catalyst structure determined with the Photoacoustic Soot Sensor (PASS) during the HDV-ESC-cycle.

Oxidation Kinetics

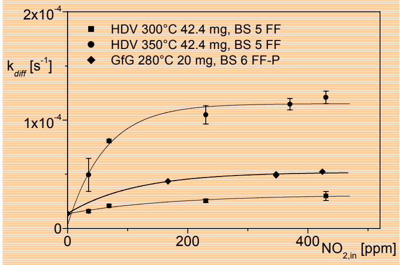

Figure 10. Differential rate coefficient k_{diff} for the oxidation of HDV diesel engine and GfG soot in the FBR system as a function of NO_2 volume mixing ratio.

Carbon Mass Balance

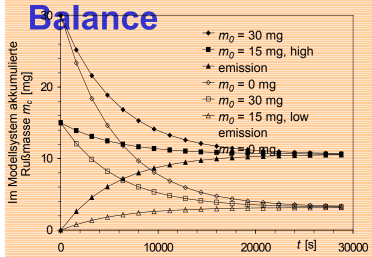

Figure 11. Accumulated soot mass in the FBR catalyst calculated with the phenomenological model based on the experimental results of this study. Simulation of 20 HDV-ESC cycles for 3 different initial mass loadings and two LDV emission modi.

Conclusions and Outlook

- Particle deposition efficiency between 45 and 85% for a wide range of realistic conditions in novel catalyst structures with microstructured coating
- Particle deposition driven by diffusion, interception and thermophoresis. Increased by the mixing characteristics of the catalyst structure.
- No significant reentrainment of soot deposits observed
- Differential rate coefficients for HDV soot oxidation between 3 x 10^{-5} s^{-1} (300°C) and 5 x 10^{-4} s^{-1} (400°C), beneficial influence of NO_2
- Good correlation between laboratory and engine test bench experiments
- Continuous soot deposition and oxidation appears to be feasible under ETC/ESC conditions (EURO IV)

References

C. van Gulijk, M. Makkee, J.A. Moulin, *Topics in Catalysis* 16 (2001), 285.

E. Jacob, N. D'Alfonso, A. Döring, S. Reisch, D. Rothe, R. Brück, P. Treiber, *23. Internationales Wiener Motorensymposium*, Band 2: Fortschritt-Berichte VDI Reihe 12 Nr. 490 Düsseldorf: VDI-Verlag 2002, 196.

E. Jacob, D. Rothe, R. Schlögl, D. S. Su, J.-O. Müller, R. Niessner, A. Messerer, C. Adelhelm, U. Poeschl, K. Müllen, C. Simpson, Z. Tomovic, *24. Internationales Wiener Motorensymposium*, Band 2: Fortschritt-Berichte VDI Reihe 12, Nr. 539 Düsseldorf: VDI-Verlag 2003, 19 - 45.

F. Jacquot, V. Logie, J.F. Brilhac, P. Gilot, *Carbon* 40 (2002), 335.

A. Messerer, H.-J. Schmid, C. Knab, U. Pöschl, R. Nießner, *Chem. Ing. Tech.* 76 (2004), 1092.

A. Messerer, D. Rothe, U. Pöschl, R. Niessner, *Topics in Catalysis* 30/31 (2004), 247.

A. Messerer, R. Niessner, U. Pöschl, *J. Aerosol Sci.* 34 (2003), 1009.