#### SOOT PARTICLE DEPOSITION AND OXIDATION IN NOVEL CATALYST STURCTURES

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#### INTRODUCTION

Since conventional soot particle filter systems exhibit a significant increase in exhaust gas back pressure and are easily clogged by engine oil ashes, current research activities are focused on the development of filterless soot particle deposition structures [1]. First experiments confirmed the high potential of newly developed structures for efficient particle deposition. Detailed studies of the involved processes including thermophoresis are currently under way and are used to optimize the design and operating conditions. The reaction mechanisms and kinetics of soot particle oxidation by nitrogen oxides are also investigated for different deposition structures and types of soot under a wide range of experimental conditions relevant for modern diesel engine exhaust systems.

#### METHODS

For the investigation of the soot deposition mechanisms a novel model gas test bench was developed allowing the characterization of soot particulate deposition in specially designed flat bed reactor systems. With a flown through cross sectional area of 10 x 6 mm these reactors provide a high flexibility concerning the type of deposition structure, their length (74.5 to 290 mm), temperature conditions (25 to  $450^{\circ}$ C), model gas composition and flow velocity under laboratory scale conditions. Simulated hourly gas space velocities range between 10,000 and 200,000 h<sup>-1</sup>. The model soot aerosol was produced by spark discharge (PALAS GfG 1000). For the measurement of the size distribution of the soot aerosol before and after the deposition device a SMPS system was applied consisting of an electrostatic classifier (TSI EC 3071) and a condensation particle counter (TSI CPC 3025 A). Complex heat control devices were adopted to ensure isothermal sampling conditions at all sampling points to cancel out sampling losses mainly caused by thermophoresis. The soot deposition studies are followed by oxidation experiments of the soot

deposited on the structures in the flat bed reactors with mainly  $NO_2$  in typical diesel engine exhaust gas conditions varying space velocity, temperatures and gas composition. Therefore it is possible to describe the oxidation kinetics of soot particles deposited under realistic flow conditions as they occur in novel particle deposition systems. The experiments with spark discharge soot are complemented by experiments with real diesel soot particles sampled at engine test benches.

#### RESULTS

Different kinds of stainless steel deposition structures were characterized concerning their deposition behaviour. Diffusion, impaction, interception and turbulent deposition are found to be the dominating deposition mechanisms. According to the type of structure the deposition efficiency is dependent on the soot loading (Figure 1) and therefore increases with time. Particle deposition was found to be significantly higher for structures which were homogeneously coated with micrometer sized steel spheres. Experiments at a heavy duty vehicle (HDV) engine test bench shows particle mass reduction of about 70% applying these structures in a silencer housing. With this exhaust gas aftertreatment system the particulate emissions were significantly below the EURO IV emission limit value of 20 mg/kWh in the ESC and 30 mg/kWh in the ETC test cycle. The maximum pressure drop was 101 mbar during the ESC test cycle which is far below the values reported from monolithic filter systems. Further engine test bench showed no blow off effects and did not indicate any clogging behaviour as it was observed in tests with conventional diesel particulate filters.



**Figure 1**: Particle deposition efficiency  $\varepsilon$  as a function of  $d_p$  in a novel catalyst structure.

Oxidation experiments with real HDV soot deposited on the novel catalyst structures show a significant influence of the oxidation rate on the deposition structure, temperature and gas composition [2]. The differential rates of carbon mass consumption at  $350^{\circ}$ C were around 5 times as high as the ones at  $300^{\circ}$ C. The rates increased slightly with NO<sub>2</sub> concentration. Significant carbon consumption was already observed at concentrations as low as 65 ppm NO<sub>2</sub>. Model calculations based on the kinetic rates derived from the experiments showed that soot oxidation under realistic exhaust gas conditions is sufficient for a continuous removal of diesel particulate matter deposited in the novel catalyst structures.

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#### REFERENCES

- Jacob E., D'Alfonso N., Döring A., Reisch S., Rothe D., Brück R., Treiber P., PM-KAT: Nichtblockierende Lösung zur Minderung von Dieselruß für EuroIV-Nutzfahrzeugmotoren, H.P. Lenz (Hrsg.) 23.
   Internationales Wiener Motorensymposium, 25.-26. April 2002, Band 2<u>: Fortschritt-Berichte VDI Reihe 12</u> <u>Nr. 490</u> Düsseldorf: VDI-Verlag 2002, S. 196-216.
- Jacob E., Rothe D., Schlögl R., Su D.S., Müller J.-O., Niessner R., Messerer A., Adelhelm C. Pöschl U.,
  Müllen K., Simpson C., Tomovic Z. Dieselruß: Mikrostruktur und Oxidationskinetik, H. P. Lenz (Hrsg.) 24.
  Internationales Wiener Motorensymposium, 15. 16. Mai 2003, Band 2: *Fortschritt-Berichte VDI Reihe 12* <u>Nr. 539</u> Düsseldorf: VDI-Verlag 2003, S. 19 45.

# Soot Particle Deposition and Oxidation in Novel Catalyst Structures

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#### Motivation

- Experimental Setup
- Particle Deposition
- Soot Oxidation
- Summary & Conclusion

### **TIM** PM-KAT<sup>®</sup> - A Novel Particle Removal System

MAN





Aims: 1) Efficient PM removal

2) Low pressure drop

#### 3) No clogging

(Jacob et al. Wiener Motoren Symposium 2002 Rothe et al. Haus der Technik 2003)



**<u>Requirement:</u>** Low-emission engine (PM < 50 mg/kWh)





### **TIM** Experimental Setup - Model Catalytic System





### Flat Bed Reactor (FBR)



- Easy to equip with deposition structures
- PID temperature control
- Macroscopic investigation of deposition pattern
- Microscopic investigation (SEM)



# **TIM** Soot Particle Deposition Investigations

#### **Operating conditions**

|                                      | Min.                  | Max.                  |
|--------------------------------------|-----------------------|-----------------------|
| Length [mm]                          | 74.5                  | 298                   |
| GSV [h <sup>-1</sup> ]               | 23,500                | 451,000               |
| v <sub>c</sub> [m s <sup>-1</sup> ]  | 1.4                   | 18.8                  |
| T [°C]                               | 25                    | 400                   |
| c <sub>p</sub> [cm <sup>-3</sup> ]   | ~ 5 x 10 <sup>6</sup> | ~ 3 x 10 <sup>7</sup> |
| c <sub>m</sub> [mg m <sup>-3</sup> ] | ~ 1.2                 | ~ 20.0                |

Particle size spectra



#### $\rightarrow$ Screening of soot particle deposition efficiency $\epsilon$



### **BS Structure**









- Generation of turbulent eddies
- Deposition of soot particles via
  - Diffusion
  - Impaction
  - Interception
  - Turbulence







- Increased surface roughness & soot storage space
- > Enhanced deposition efficiency (interception & impaction)



# **TIM** Microsphere-BS Deposition Efficiency MANGOLD



- High initial ε
- Acceptable pressure loss ( $\leq$  80 mbar)
- μm-sized agglomerates





## **HDV Engine Test Bench Results**





TLM



#### Motivation

- Experimental Setup
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### **Differential Rate Coefficient**



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## **Concentration & Temperature Dependency**





- 1. Novel deposition structures (microsphere coating)
- **2. High deposition efficiencies** (up to 80%)
- 3. Low pressure drop (< 100 mbar)
- 4. Consistent laboratory & engine test bench results
- 5. Efficient soot oxidation under ESC/ETC conditions (continuous regeneration feasible)
- → Economic & reliable solution for low emission HDV-engines (EURO IV requirements)









