

Heavy Duty Vehicle Soot Oxidation and its Application in Regenerating Particulate Traps

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Reduction of soot discharged into the atmosphere by heavy-duty diesel engines is an important problem from an environmental viewpoint. World-wide legislation has tightened the particulate standards for diesel engines over the years. These have induced a rapid development of low emission heavy-duty diesel engines and equipment aiming at reducing soot. Such efforts lead to technologies for the treatment of soot particulate in the engine exhaust train. This work is focussed on this so called “*after-treatment*” of soot, which is based on the application of novel deposition structures in the exhaust line which are continuously regenerated by oxidation of the deposited soot. Results of a study on microstructure and oxidative behaviour of HD diesel engine soot are presented.

A Philips TEM/STEM CM 200 FEG transmission electron microscope equipped with a field-emitting gun is used to study the morphology and microstructure of the soot. The acceleration voltage is set to 200kV. For EELS measurements the Gatan-imaging-filter (GIF 100) is used to investigate the electronic structure.

The TEM-investigations reveal the inner structure of the carbonaceous materials. The diesel engine soot consists of primary particles (approx. 12 - 15 nm in diameter). The carbon is made from units of sp^2 hybridised carbon that form interlaced bundles of ribbons with eventual large areas of planar interconnection, so-called basic structural units (Fig. 1a). The primary units are bands of carbon with multiple continuous bending. The particles are built of bent graphenes and graphite ribbons indicating a fullerenoid structure. The EELS investigations reveal predominant graphite like bonding behaviour of the carbon (sp^2 hybridisation). The very strong bending of the graphenes lead to an increased partial s -character and to a fullerenoid electronic structure similar to onion like carbon (Fig. 1b).

To investigate the oxidation kinetics of real HDV diesel soot under well defined conditions, flat bed reactors equipped with novel soot deposition structures were exposed to an exhaust flow from an engine test bench and loaded with soot. In a laboratory scale model catalytic converter system they were exposed to a gas flow containing oxygen, nitrogen oxides, and water vapor at concentration levels relevant for real exhaust conditions.

The decrease of the soot carbon mass m_c was quantified by integration of the CO and CO₂ signals from Fourier-Transform Infrared Spectroscopy (FTIR) analysis. For an oxidation experiment of Euro III HDV soot with 8.8 Vol.% O₂, 3.0 Vol.% H₂O and 910 ppm NO_x on the deposition structures described above without catalytic coating, the ratio of m_c to its initial value $m_{c,0}$ is plotted against reaction time in Fig. 2a. The temperature of the FBR was increased from 300 to 350 and further on 400°C.

Fig. 2b displays the corresponding pseudo first order differential rate coefficient $k_{diff} = dm_c / (m_c dt)$ observed at 350°C for different NO₂ volume mixing ratios. In Fig. 3 the data from Fig. 2b and from analogous variations at 300°C are plotted against the NO₂ volume mixing ratio. In both cases k_{diff} varied only by about 15% over the range of 230 to 430 ppm of NO₂. Based on these results a continuous removal of DPM should be possible under the conditions of an ESC cycle. The reaction rates differ significantly from the ones reported from the

investigation of bulk samples, which confirms the need for the continuation of detailed reaction kinetic studies.

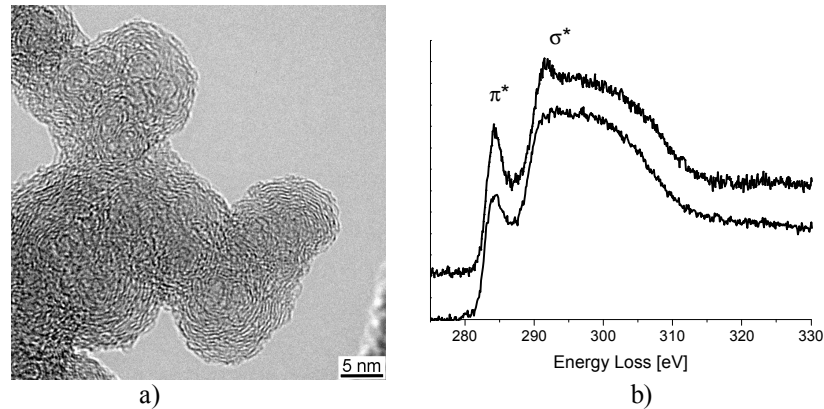


Fig. 1: a) HRTEM of HD diesel engine soot, b) EELS measurements

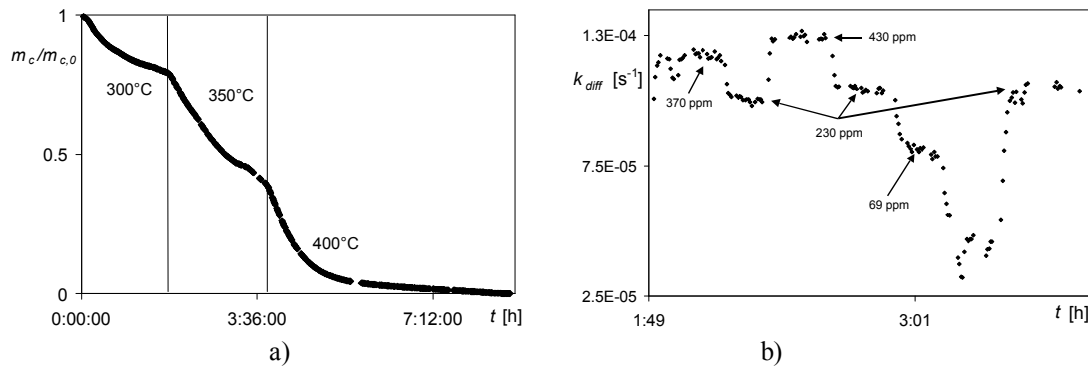


Fig.2: a) Relative carbon mass decrease $m_c / m_{c,0}$ during the oxidation of HDV diesel engine soot in the FBR system with 8.8 Vol.% O₂, 3.0 Vol.% H₂O and 910 ppm NO_x for 300, 350 and 400°C. b) Differential rate coefficient k_{diff} for the oxidation of HDV diesel engine soot in the FBR system with 8.8 Vol.% O₂, 3.0 Vol.% H₂O, and different NO₂ volume mixing ratios at 350°C.

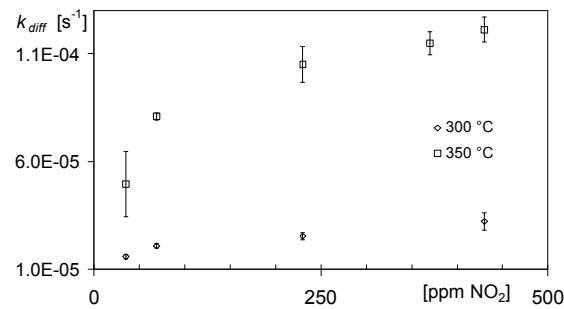


Fig. 3: Differential rate coefficient k_{diff} for the oxidation of HDV diesel engine soot in the FBR system as a function of NO₂ volume mixing ratio.

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