Properties of Diesel Soot Particles to be Catalytically Reacted with NO_2

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Abstract

Soot particles of submicron size in exhausts of diesel engines are, according to the actual knowledge, toxicologically dubious so that in the European Community the highest limit for the particle emission in the legal norm for engine-exhaust will be drastically decreased in the next few years. This has initiated a large number of efforts to optimise the combustion processes [1]. A system to reduce soot particles with a temporarily particle deposition on a filterless catalyst system with continuous regeneration is developed. The aim is a soot free diesel engine without particle filter to reach the EURO IV/V limit values [2]. The design of such systems requires a detailed knowledge of the carbon to be reacted with NO_2.

The main interests of the present work are focused on the morphology, microstructure, size-distribution and electronic state of the particle agglomerates [3] as well as the surface functional groups [4-6]. Results are obtained from conventional and high-resolution transmission electron microscopy (TEM, HRTEM), electron energy loss spectroscopy (EELS), energy dispersive X-ray spectroscopy (EDXS), X-ray photoelectron spectroscopy (XPS), and infrared spectroscopy in diffuse reflection (DRIFTS).

Different samples are collected directly from the exhaust gas of EURO IV/V test diesel engine of MAN Nutzfahrzeuge AG using a special particle collector, allowing to reproduce the important parameters for the collection procedure. Soot particles are collected at different parts of the engine-catalyst-system. Other test-cycles vary the engine parameters as load and revolutions per minute.

The morphology of the soot is characterised by means of TEM. The characterisation of the electronic structure is performed with EELS and the analysis of elements in the soot with EDXS.

The spherical soot-particles have a diameter of 20-30 nm, but not all of the primary particles exhibit spherical form as expected in the case of furnace soot. The high-resolution images show that the particles consist of small graphite-like crystallites, oriented around the particle centre. Agglomerates contain primary particles and arrays of amorphous carbon.

The EEL near edge fine structure of the measured carbon-K-edge reveals differences in the electronic structure of the particulates due to the bonding of carbon atoms (sp^2 and sp^3). The carbon edges also show features from parts of amorphous and graphite like regions. EDXS-measurements show signals of sulphur, iron, calcium, phosphorus and zinc originating from engine oil ash.

To reveal surface properties of the soot particles investigation methods like XPS and DRIFTS are applied.

XPS show contributions of oxygen functional groups in the C1s and O1s spectra. Ethers, lactone, carbonyl and carboxyl groups can be derived. In addition to this method, infrared spectroscopy in diffuse reflection show vibrational bands which can also be assigned to oxygen functional groups on the surface of the soot particles.

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