UNDERSTANDING THE STRUCTURE AND PROPERTY OF DIESEL SOOT PARTICLES: CONTRIBUTIONS OF HREM AND EELS

D.S. SU

Department of Inorganic Chemistry, Fritz-Haber-Institut der Max-Planck Gesellschaft, Faradayweg 4-6, D-14195 Berlin, Germany

Reducing the exhaust emission of automotives is one of the most important measure in environmental protection. The abundant undesirable compounds found in exhaust gases are CO, HC, NO_x, SiO_x and particulates or soot. Soot is a by-product of incomplete fuel combustions. Due to the increasing concern of the environmental threat of soot particulates and their adverse effects on human organism, US and European legislations have introduced particulate standards for diesel engines and have tightened the standards over the years. Nowadays, engine manufactories enhance the effort in developing new low emission engine to fulfil the new particulate standard that will be applied in 2005.

The rapid development in engineering requires an updating investigation of soot particulates emitted by new low emission engines. Recently, we studied the morphology and microstructure of soot of an optimised low-emission EURO-IV test diesel engine, equipped with a double step controlled charging and an external controlled cooled exhaust gas recirculation. A Philips TEM/STEM CM 200 FEG equipped with a field-emission gun is used to study the morphology and microstructure of the soot. The accelerating voltage is set to 200 kV.

High-resolution image in Fig. 1 presents the image contrast of a spherical primary particle agglomerated with other primary particles. A core-shell structure can be identified: the core is of about 5 nm in size and exhibits an ill-defined structure. The shell consists of layers of carbon in radial symmetry. Primary particles with such an onion like core-shell structure is in consistence with other observations and can be interpreted by the existing theoretical models of soot nucleation: the polycyclic aromatic hydrocarbons (PAH) condense to clusters that coagulate to cores. The subsequently adsorbed PAH-clusters dehydrate and undergo atomic re-arrangement forming graphene layers at high temperatures.

Surprisingly, our observation reveals that the core-shelled primary particles are not in majority of soot emitted from the low-emission diesel engine. The high-resolution image in Fig. 2 shows one part of an agglomerated particle. The particle shows neither defined structure nor regular shape. The basic structure is the defect-rich and bended graphene layer, formed from PAHs. Circle like contrast along the periphery of the particle is found, which can be identified as from fullerene molecule and fullerenoid cluster [1]. This is the evidence that fullerenes and fullerenoid clusters can be formed in the combustion process of fuel in the optimised low emission diesel engine. The investigation reveals that, besides PAH, fullerene and fullerenoid clusters are also the

source of soot formation. Since this kind of soot and PAH-soot covered by fullerene and fullerenoid can exhibit different oxidative behaviours with O_2 or NO_2, the finding may have consequences on the automotive catalysts and on the treatment of exhaust after combustion.

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Reference
[1] D.S. Su, J.-O Müller, D. Rothe, E. Jacob, R. Schlögl, to be published

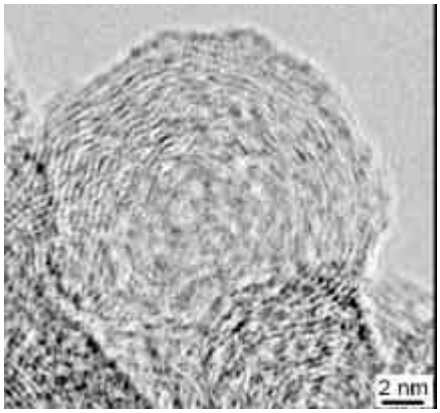

Fig. 1 (left) Core-shell structured soot particulate found in a low emission diesel engine.

Fig. 2(bottom) Soot agglomerate with undefined structure. Contrasts due to fullerenes and fullerenoids on the periphery are arrowed.