Optical investigations to the influence of engine operating parameters on physical and chemical properties of soot particles

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In recent years, scientists revealed the fact that emitted soot particles change with different engine types and different engine generations. This is not only true for a mass reduction according to emission regulations and for a reduction of the emitted particle number due to progress in combustion and exhaust gas aftertreatment devices, but also for the physical and chemical properties like BET surface area [1], reactivity [2-4] or primary particle size [5] of engine-out soot particles. It has been shown by many research groups that these properties have an impact on health [6] as well as an influence on the reactive behavior in catalytic aftertreatment devices [4, 7]. Besides the influence of engine design and fuel properties, many of these changes in engine-out soot particle properties are influenced by the development in engine combustion. For example in modern diesel engines the injection pressure has been raised up to over 2000 bar and the use of emission gas recirculation (EGR) is a common tool to control combustion and emissions. Therefore the main focus of our work is on the influence of changes in engine operating parameters on combustion and hence on the physical and chemical properties of emitted soot particles. For the parameter study, measurements with an optically accessible single cylinder engine and a modern production Audi V6 TDI engine on a dynamometer were conducted. The results from both engines were compared to each other to better evaluate the global reliability of the trends.

Parameter study at an engine dynamometer (Audi 3.0l V6 TDI engine)
Different operating points (Table 1) with changes in either rail pressure, Lambda or torque were investigated with an SMPS, thermogravimetry and HR-TEM imaging. The samples for the measurement devices were taken directly out of the tailpipe. For the SMPS and the thermogravimetry a rotating disc diluter and a thermodenuder were used to condition the sample flow prior to the measurements.

<table>
<thead>
<tr>
<th>Operating point</th>
<th>OP TDI1</th>
<th>OP TDI2</th>
<th>OP TDI3</th>
<th>OP TDI4</th>
<th>OP TDI5</th>
<th>OP TDI6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Torque Nm</td>
<td>70</td>
<td>70</td>
<td>210</td>
<td>210</td>
<td>240</td>
<td>240</td>
</tr>
<tr>
<td>Injection pressure bar</td>
<td>600</td>
<td>750</td>
<td>950</td>
<td>1150</td>
<td>1000</td>
<td>1200</td>
</tr>
<tr>
<td>Lambda</td>
<td>3.0</td>
<td>3.0</td>
<td>1.8</td>
<td>1.8</td>
<td>1.5</td>
<td>1.5</td>
</tr>
</tbody>
</table>

For all operating points the SMPS measurements revealed a reduction in the mobility diameter of the emitted particles with increasing rail pressure. The same trend was seen for the mean diameter of the primary particles that were measured out of many HR-TEM images of soot agglomerates. An exception was the step from OP TDI 2 to 3 with almost no change in diameter. This was most probably due to a strong increase in engine load with an enrichment of the in-cylinder mixture.

For all operating points samples were taken onto quartz glass filters for thermogravimetric measurements. The samples were heated up to 600 °C with 5 K/min in a nitrogen atmosphere in order to evaporate all volatiles on the sampled soot. After cooling down to 300
°C the samples were heated up to 700 °C in synthetic air to oxidize the soot. The results clearly revealed a reduced temperature for the main oxidation of the soot samples with smaller primary particles. From OP TDI 1 to 6 the temperature for the maximum sample mass loss rate during oxidation differed for almost 50 K. Two possible explanations for this behavior are an increased surface to mass ratio for smaller particles or a change in particle morphology.

Parameter study at an optically accessible single cylinder DI diesel engine
To further investigate the reason for changes in soot particle formation a parameter study with the analysis of the complete engine sequence of events was carried out (Table 2). For the in-cylinder measurements Mie scattering, laser-induced exciplex fluorescence and spectroscopy were applied. For engine-out measurements, soot particles were counted and sized with a SMPS system (with rotating disc diluter and thermodenuder) and the primary particle size was determined by HR-TEM imaging. So far, only some of the operating points have been evaluated and are presented in this study.

Table 2: Parameter study at the single cylinder engine

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Engine speed rpm</td>
<td>600 / 800 / 1000</td>
</tr>
<tr>
<td>Injection pressure bar</td>
<td>800 / 1000 / 1300</td>
</tr>
<tr>
<td>Boost pressure, abs. bar</td>
<td>1.05 / 1.25 / 1.45</td>
</tr>
<tr>
<td>Injection timing °CA</td>
<td>-10 / -6 / -3</td>
</tr>
<tr>
<td>EGR rate %</td>
<td>0 / 25 / 50</td>
</tr>
</tbody>
</table>

For both, a rise in rail pressure and a rise in in-cylinder boost pressure (air mass) the SMPS and HR-TEM measurements have shown a decrease in particle mobility diameter and primary particle size. For a closer look on the combustion process which strongly influences to the particle formation, the operating points with a variation in rail pressure were investigated with integral and spatially resolved spectroscopy.

At first, a time resolved spectroscopy with the integrated in-cylinder combustion luminosity was carried out. The overall soot luminosity of the operating points with higher injection pressures was always lower. For a better evaluation of this data, the spatially resolved combustion luminosity at two different wavelength was observed through the piston bowl window with an intensified CCD camera. For an evaluation on soot, the soot signal around 550 ± 20 nm was recorded through an interference filter. As a second species, the chemiluminescence of the OH-radical was also recorded through the piston bowl window. The OH* signal was filtered by an interference filter at 308 ± 10 nm. For the operating points with higher injection pressures the intensity of the OH* signal increased and appeared earlier with regard to the injection time. The soot signal in contrast was lower for higher injection pressures. In a comparison between soot and OH* signal the appearance of OH* was in advance of the soot signal especially for the high injection operating point. With these results it is obvious that the ratio of OH* to soot signal increased strongly for higher injection pressures. This supports the assumption that the soot oxidizing OH* leads to a decreased soot formation rate and a decreased concentration of primary particles, thus causing smaller primary particles and lower soot agglomerate concentrations [8]. With the different results from two different approaches, it was shown that engine-out soot properties are depending on engine operating conditions and hence on combustion. Current studies are therefore focusing on expanding the variations of engine operating parameters
and additional techniques like Electron Energy Loss Spectroscopy and BET surface measurements of soot samples and in-cylinder mixture formation studies.


Optical Investigations of the Influence of Engine Operating Parameters on Physical and Chemical Properties of Soot Particles

MOTIVATION

Soot particle emissions change with engine generations and engine operating conditions with regard to:

- chemical and physical properties
- biological (toxicological) and chemical reactivity
- influence on human health and environment
- exhaust aftertreatment of PM emissions

Objective:

Determine correlation between engine operating conditions and soot particle properties

PARAMETER STUDIES

Operating Points (OP) for an Audi 3.0L V6 TDI engine on a dynamometer testbench

Fixed:

- Engine geometry / Engine speed

Variations:

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<td>750</td>
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</tr>
</tbody>
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A test matrix with 240 operating points was carried out in an optically accessed single-cylinder engine

Fixed:

- Engine geometry / Diesel fuel mass (13 mg) / Intake air temperature

Variations:

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MEASUREMENT TECHNIQUES

- Engine Sequence of Events
  - Injection
  - Mie scattering
  - Mixture formation
  - λ-mapping
  - Combustion
  - Spectroscopy, thermodynamics
  - Engine-out emissions
  - SMPS, microscopy, thermogravimetry, BET

RESULTS

Spectroscopy and engine-out results for the optically accessed single-cylinder engine

Effect of injection pressure on engine-out soot particle distribution

Effect of boost pressure on engine-out soot particle distribution

Results for engine-out emissions of the Audi 3.0L V6 TDI engine

High Resolution Transmission Electron Microscopy (HR-TEM) imaging to determine the mean size of primary particles

The size distribution of primary particles is determined by measuring multiple soot agglomerate images from samples of each operating point

Thermogravimetry of engine-out soot samples

Soot samples on a quartz glass filter were dried in N₂ atmosphere up to 873K prior to the temperature programmed oxidation in synthetic air.

Summary

All temporary evaluated measurements demonstrate:

- smaller primary particles for higher injection pressure
- lower oxidation temperatures for soot samples with smaller primary particles
- different temporal and spatial formation of characteristic combustion-specific species
- intense OH* appearance reduces soot formation due to oxidation reactions

Current and future work

Additional studies will be carried out to evaluate the

- influence of mixture formation on soot formation
- effect of transient engine conditions on soot properties
- soot property impact to biological and catalytic systems

In-cylinder fuel distribution