Extended Abstract to the Presentation

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

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In recent years, many research groups 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 researchers 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 in-cylinder combustion process. 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. The objective is to visualize and analyze all in-cylinder processes that can possibly influence the soot formation and the soot oxidation. This data is than correlated with the results of the physical and chemical investigations of the sampled soot emissions. 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.
All Figures to the described results below are published in the printed slides to our talk that are available together with this extended abstract in the proceedings to the 15th ETH Conference on Combustion Generated Nanoparticles.

Parameter study at an engine dynamometer (Audi 3.0l V6 TDI engine)
The results from the production engine study were already presented on a poster of the 14th ETH-Conference [8] and in a paper of the 13th ETH-Conference [9]. Different operating points (Table 1) with variations in injection pressure, lambda and 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 1: Operating points for the Audi V6 TDI engine

Operating point		OP TDI1	OP TDI2	OP TDI3	OP TDI4	OP TDI5	OP TDI6
Torque	Nm	70	70	210	210	240	240
Injection pressure	MPa	60	75	95	115	100	120
Lambda (1/Φ)	--	3.0	3.0	1.8	1.8	1.5	1.5

For all operating points the SMPS measurements revealed a reduction in the mobility diameter of the emitted soot particles with increasing injection 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 particle 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 773 K with 5 K/min in a nitrogen atmosphere in order to evaporate all volatiles on the sampled soot. After cooling down to 573 K the samples were heated up to 973 K 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. BET surface measurements of soot samples and details from HR-TEM imaging will soon clarify this question.

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 high-speed Mie scattering and combustion visualization, 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.

Table 2: Parameter study at the single cylinder engine

Engine speed	rpm	600 / 800 / 1000
Injection pressure	MPa	80 / 100 / 130
Boost pressure, abs.	MPa	0.105 / 0.125 / 0.145
Injection timing	°CA	-10 / -6 / -3
EGR rate	%	0 / 25 / 50
Fueling rate	[mg/stroke]	17

For both, a rise in injection 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 injection pressure were investigated with integral, time resolved and spatially resolved spectroscopy.

At first, a time resolved spectroscopy with the integrated in-cylinder combustion luminosity was carried out. The overall soot luminosity for operating points with higher injection pressures and for the ones with higher boost pressure was always lower. The increase in boost pressure also induces a strong increase of the OH-radical (seen around 308 nm in the spectrum) during the time of combustion. For a better evaluation of this data, the spatially resolved combustion luminosity at two different wavelengths was observed through the piston bowl window with an intensified CCD camera. For an evaluation on soot, the soot signal around 490 ± 20 nm was recorded through an interference filter. As a second species, the chemiluminescence of the OH-radical was simultaneously recorded through the piston bowl window with an image doubler. The OH*-signal was filtered by an interference filter at 305 ± 10 nm. For the operating points with higher injection pressures and higher boost pressures the intensity of the OH*-signal has a stronger increase and appeares earlier with regard to the injection time. The soot signal in contrast is lower for higher injection pressures as well as for the higher boost pressures. In a comparison between soot- and OH*-signal the appearance of OH* is in advance of the soot signal for the investigated operating points. With these results it is obvious that the ratio of OH* to soot signal increases strongly for higher injection pressures and higher boost pressures. This is especially true for the first but short premixed combustion phase and the phase of soot oxidation after the main (diffusion) combustion phase. During the diffusion combustion phase a sharp drop in the OH*/soot-ratio is visible for all operating points.

To evaluate these results with regard to the emissions, SMPS measurements and HR-TEM imaging was performed. The SMPS measurements revealed the fact, that the mobility diameter of the soot particles, the overall soot particle mass and the overall soot particle number are reduced for an increase of the overall OH*/soot-ratio, hence with an increase of injection and boost pressure. The HR-TEM images also show a reduction of the average soot agglomerate concentration and size as well as a reduction of the primary particle size for increasing injection (and) or boost pressures.

This supports the assumption that the soot oxidizing OH* in an early combustion phase leads to a decreased soot formation rate and a decreased concentration of primary particles, thus causing smaller primary particles and lower soot agglomerate concentrations [10]. An increased concentration of OH* in a late combustion phase mainly supports the oxidation of present primary particles and soot agglomerates and reduces the engine-out soot concentration.

With the different results from two different approaches, it was shown that engine-out soot properties are strongly depending on engine operating conditions and hence on combustion. Currently the results of additional measurements like Mie scattering of the injection event, pressure indication of the combustion, laser induced exciplex fluorescence of the mixture formation, Electron Energy Loss Spectroscopy and BET surface measurements of soot samples are evaluated together with the presented results in order to get a complete assessment along the soot formation and soot oxidation processes that result in the existing difference of physical and chemical soot properties.

[1] Wittmaak, K.: In Search of the most relevant parameter for quantifying lung inflammatory response to nanoparticle exposure: Particle number, surface area, or what? Environmental Health Perspectives, 2007, 115(2): p. 187-194.

[2] Müller, J.-O., et al.: Diesel engine exhaust emission: oxidative behavior and microstructure of black smoke soot particulates. Environmental Science & Technology, 2006, 40: p. 1231-1236.

[3] Müller, J.-O., et al.: Morphology-controlled reactivity of carbonaceous materials towards oxidation. Catalysis Today, 2005, 102-103: p. 259-265.

[4] Jacob, E., et al.: Dieselruß: Mikrostruktur und Oxidationskinetik. VDI Fortschritts-Berichte, 2003, 539(2): p. 19-45.

[5] Oh, K.C. and H.D. Shin: The effect of oxygen and carbon dioxide concentration on soot formation in non-premixed flames. Fuel, 2006. 85: p. 615-624.

[6] Su, D.S., et al.: Cytotoxicity and Inflammatory Potential of Soot Particles of Low-Emission Diesel Engines. Environmental Science & Technology, 2008, 42(5): p. 1761-1765.

[7] Su, D.S., et al.: Fullerene-like soot from Euro IV diesel engine: consequences for catalytic automotive pollution control. Topics in Catalysis, 2004, 30/31: p. 241-245.

[8] Leidenberger, U. and D. Brüggemann: Optical investigations to the influence of engine operating parameters on physical and chemical properties of soot particles: 14th ETH Conference on Combustion Generated Nanoparticles, 2010, Zürich

[9] Leidenberger, U. et al.: Influence of Engine Operating Parameters on Physical and Chemical Properties of Emitted Soot Particles: 13th ETH Conference on Combustion Generated Nanoparticles, 2009, Zürich

[10] Angrill, O., et al.: Influence of exhaust gas recirculation on soot formation in diffusion flames. Proceedings of the Combustion Institute, 2000, 28(2): p. 2643-2649.

Bayreuth Engine Research Center

Deutsche Forschungsgemeinschaft

Lehrstuhl für Technische Thermodynamik und Transportprozesse
Prof. Dr.-Ing. D. Brüggemann

15th ETH-Conference on Combustion Generated Nanoparticles,
Zürich, 26th-29th June 2011

Ulrich Leidenberger, Sven Koch, Wolfgang Mühlbauer, Sebastian Lorenz and Dieter Brüggemann

Optical Investigations to the Influence of Diesel Engine Operating Parameters on Physical and Chemical Properties of Emitted Soot

Universität Bayreuth

Motivation

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Physical and chemical properties of soot particle emissions change with engine operating conditions [1-9]

- primary particle size
- oxidation behavior
- VOF / EC
- BET surface area
- morphology
- graphitization
- reactive groups
- toxic and inflammatory potential

→ **Influence on environment and human health**

→ **Consequences for catalytic aftertreatment**

[1] Su et al., 2008, [2] Su et al. 2004, [3] Jacob et al. 2003, [4] Ghzaoui et al. 2003, [5] Kennedy 2007, [6] Müller et al. 2005, [7] Wittmaack 2007, [8] Stratakis et al 2003, [9] Lapuerta et al. 2007

Motivation

Engine Sequence of Events

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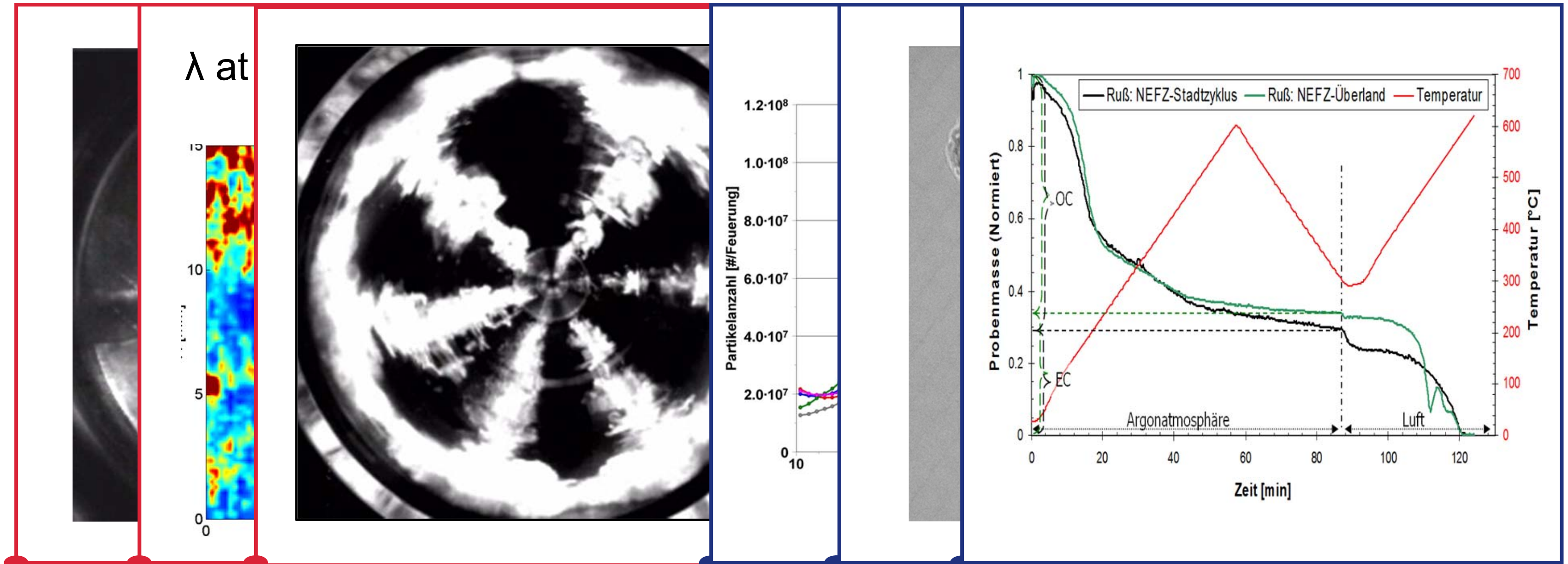
In-cylinder processes

Engine-out emissions



- HS-spectroscopy
- Laserinduced exciplex fluoresc.
- Mie-scattering and HS-imaging
- TGA/BET-analysis
- HR-TEM-imaging
- SMPS-measurement

Soot Study Part I
Single Cylinder Engine
Variation of Operating Parameters

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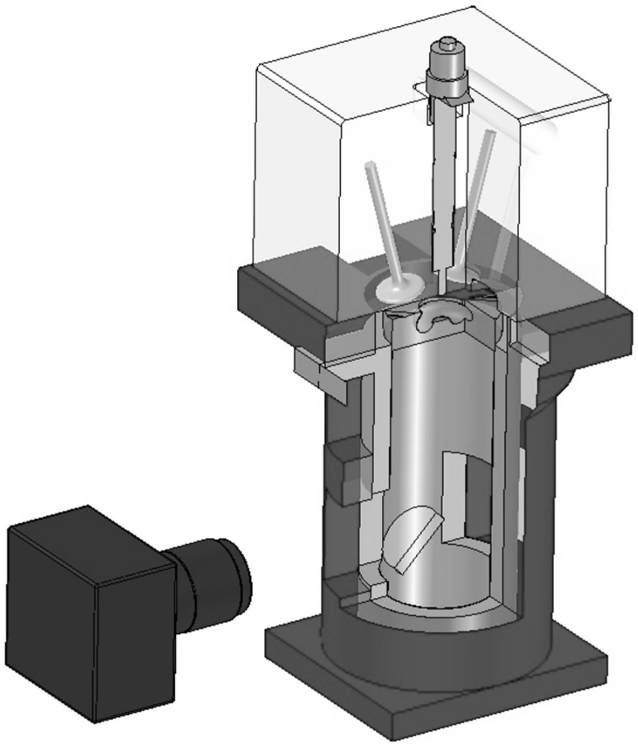
- Optical investigation of the in-cylinder processes
- Simultaneous sampling of the engine-out soot emissions



speed in rpm	600; 800; 1000
injection pressure in MPa	80; 100; 130
injection timing in °CA BTDC	-10; -6; -3
boost pressure in MPa	0.105; 0.125; 0.145
EGR-rate in %	0; 25; 50
fueling rate in mg/stroke	17

Soot Study Part II
Full Size Engine
EURO4-TDI on a Test Rig

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Tailpipe Soot Sampling

- Direct soot-sampling for HR-TEM measurements
- Sampling on Tissuequartz-filters and SMPS measurements
- (Electro-)Filter sampling for soot analysis

Parameters of the Operating Points (OP)

- Steady-state operating conditions
- Variation of injection pressure, torque and lambda

	OP1	OP2	OP3	OP4	OP5	OP6
torque in Nm	70	70	210	210	240	240
injection pressure in MPa	60	75	95	115	100	120
lambda (1/ϕ)	3	3	1.8	1.8	1.5	1.5

HR-TEM-Imaging

Size of Primary Particles

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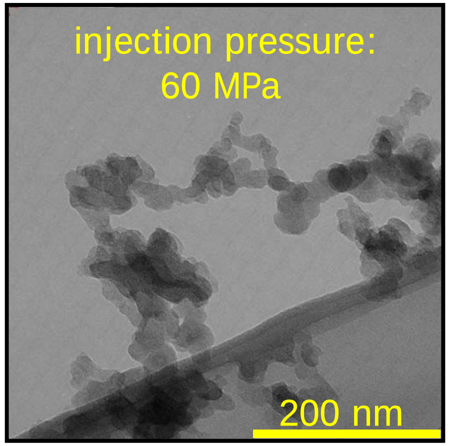

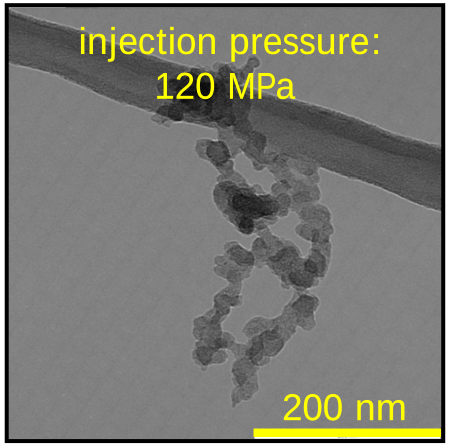

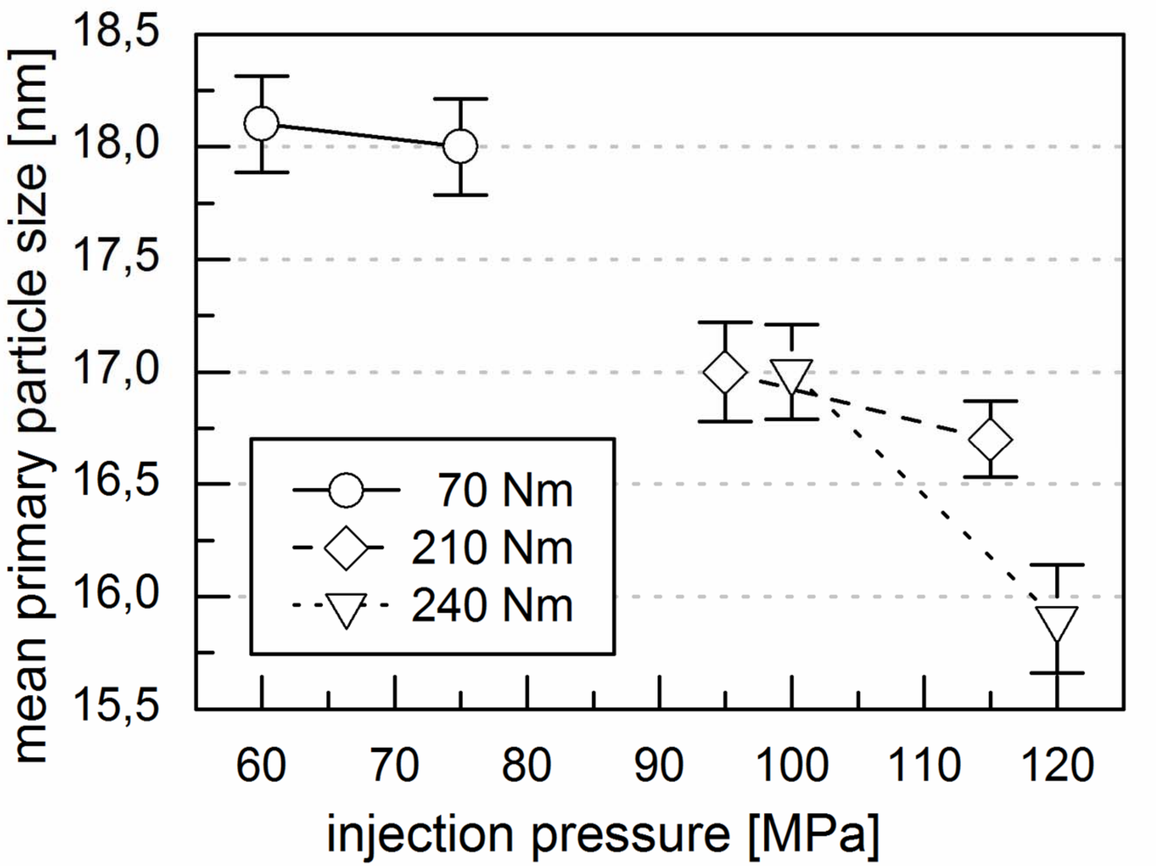

Thermogravimetry

Oxidation of Soot Samples

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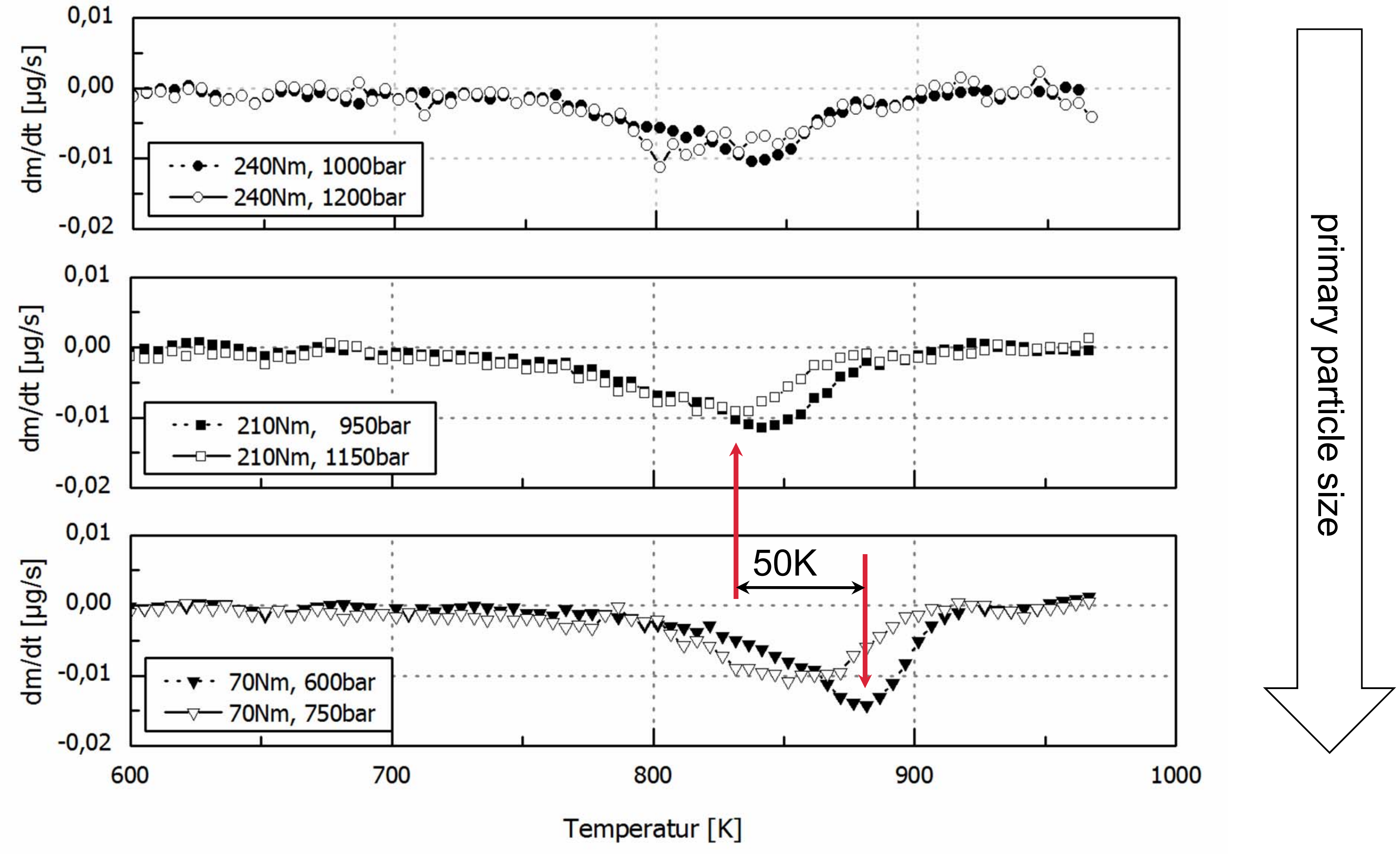

Soot Study Part I
Single Cylinder Engine
Variation of Operating Parameters

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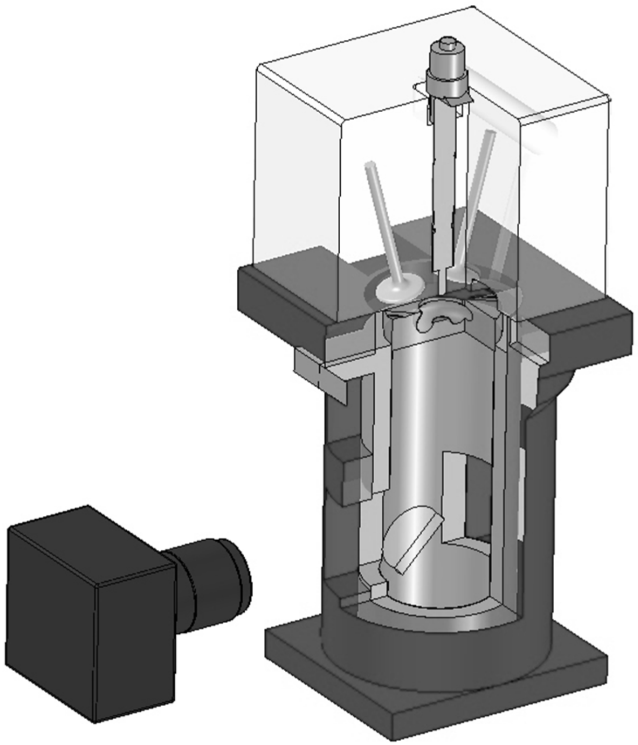
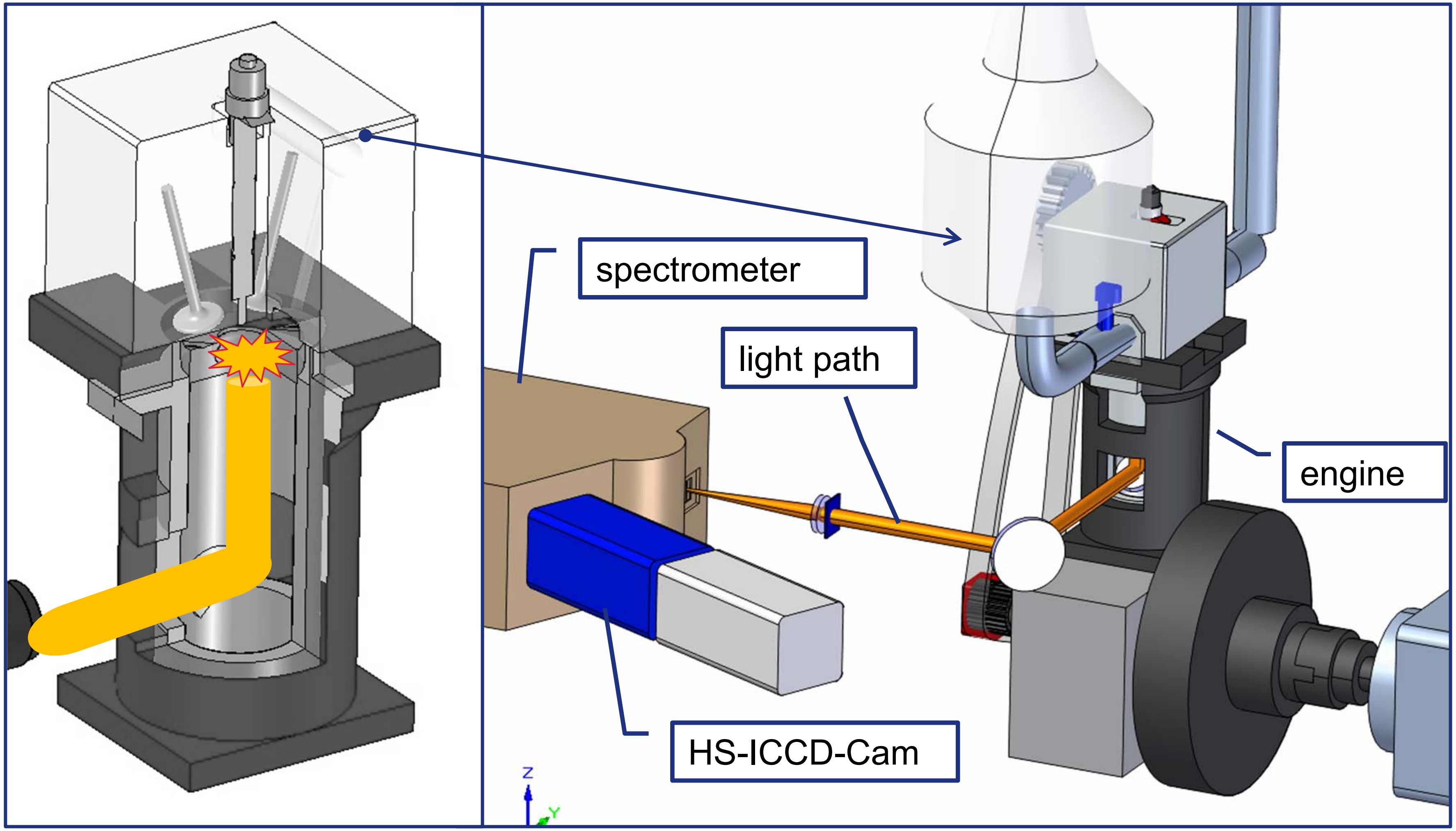
- Optical investigation of the in-cylinder processes
- Simultaneous sampling of the engine-out soot emissions



speed in rpm	600; 800; 1000
injection pressure in MPa	80; 100; 130
injection timing in °CA ATDC	-10; -6; -3
boost pressure in MPa	0,105; 0,125; 0,145
EGR-Rate in %	0; 25; 50
fueling rate in mg/stroke	17

Spectroscopy

Single Cylinder Setup



Chemiluminescence of Combustion

Variation of Boost Pressure

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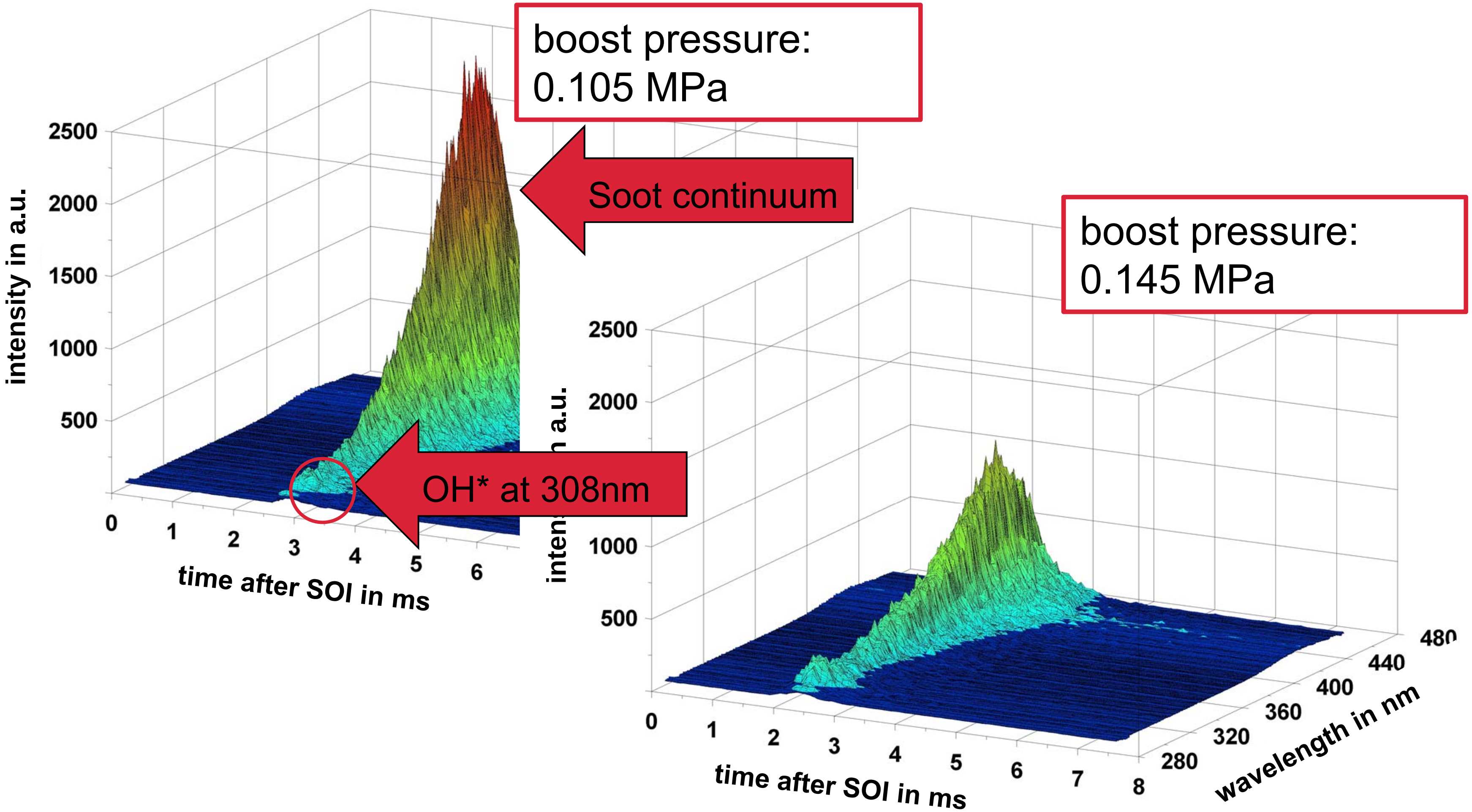

Chemiluminescence of Combustion

Variation of Injection Pressure

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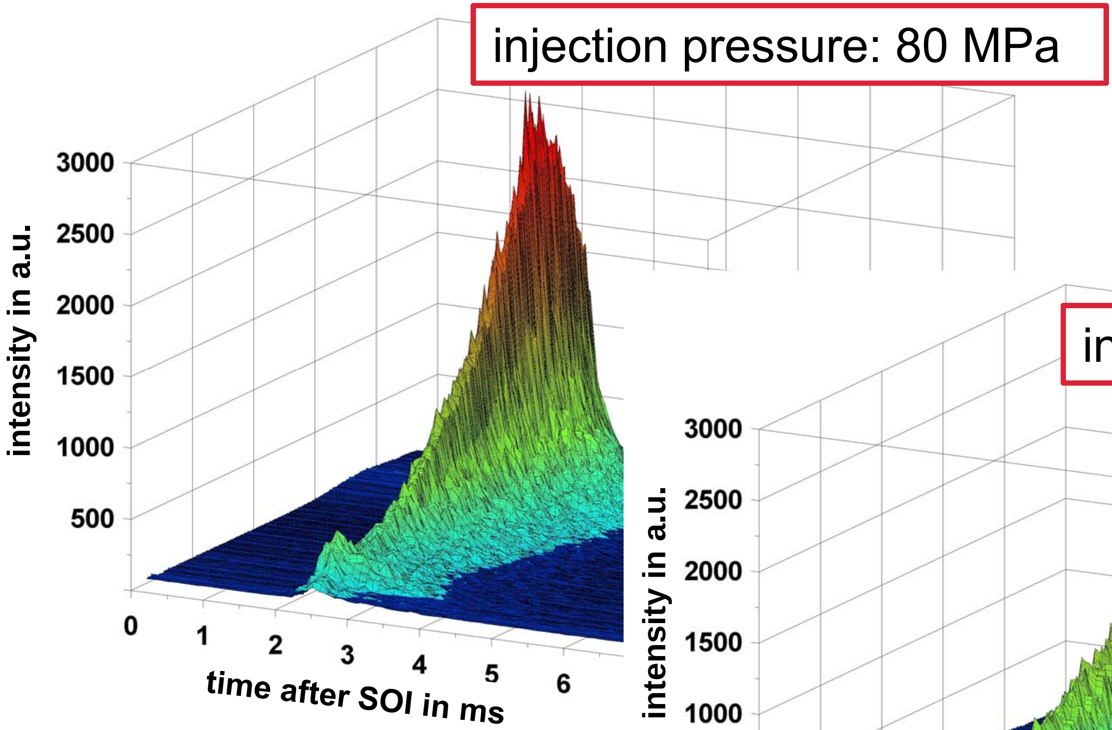

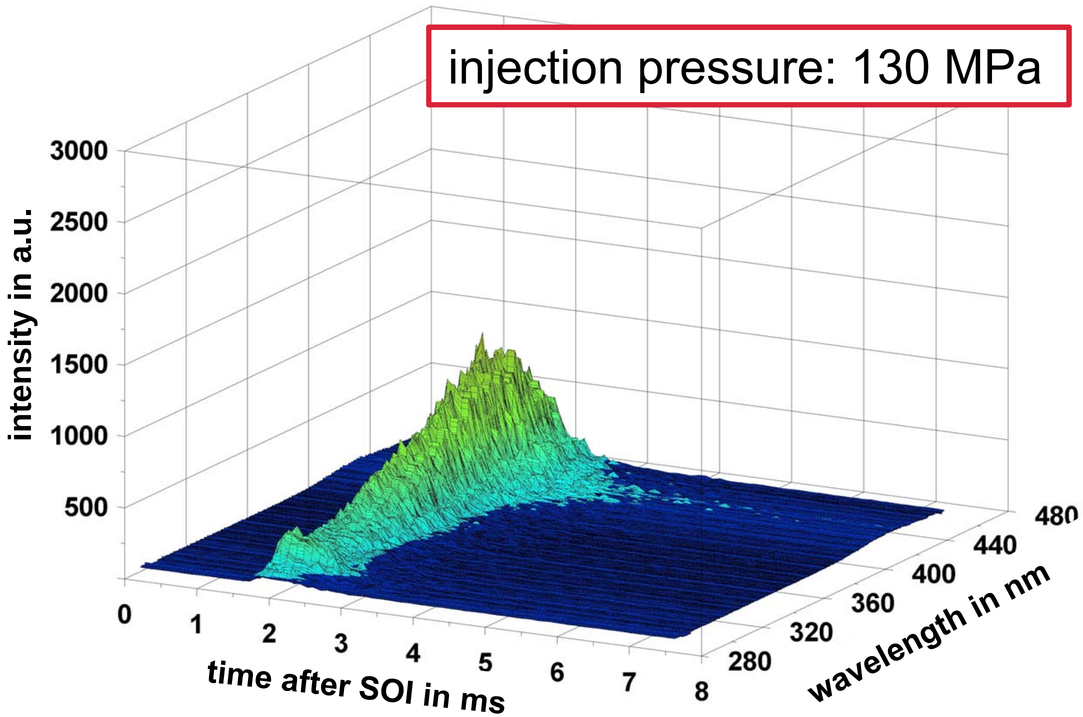

Spatially Resolved OH*/Soot Imaging

Single Cylinder Setup

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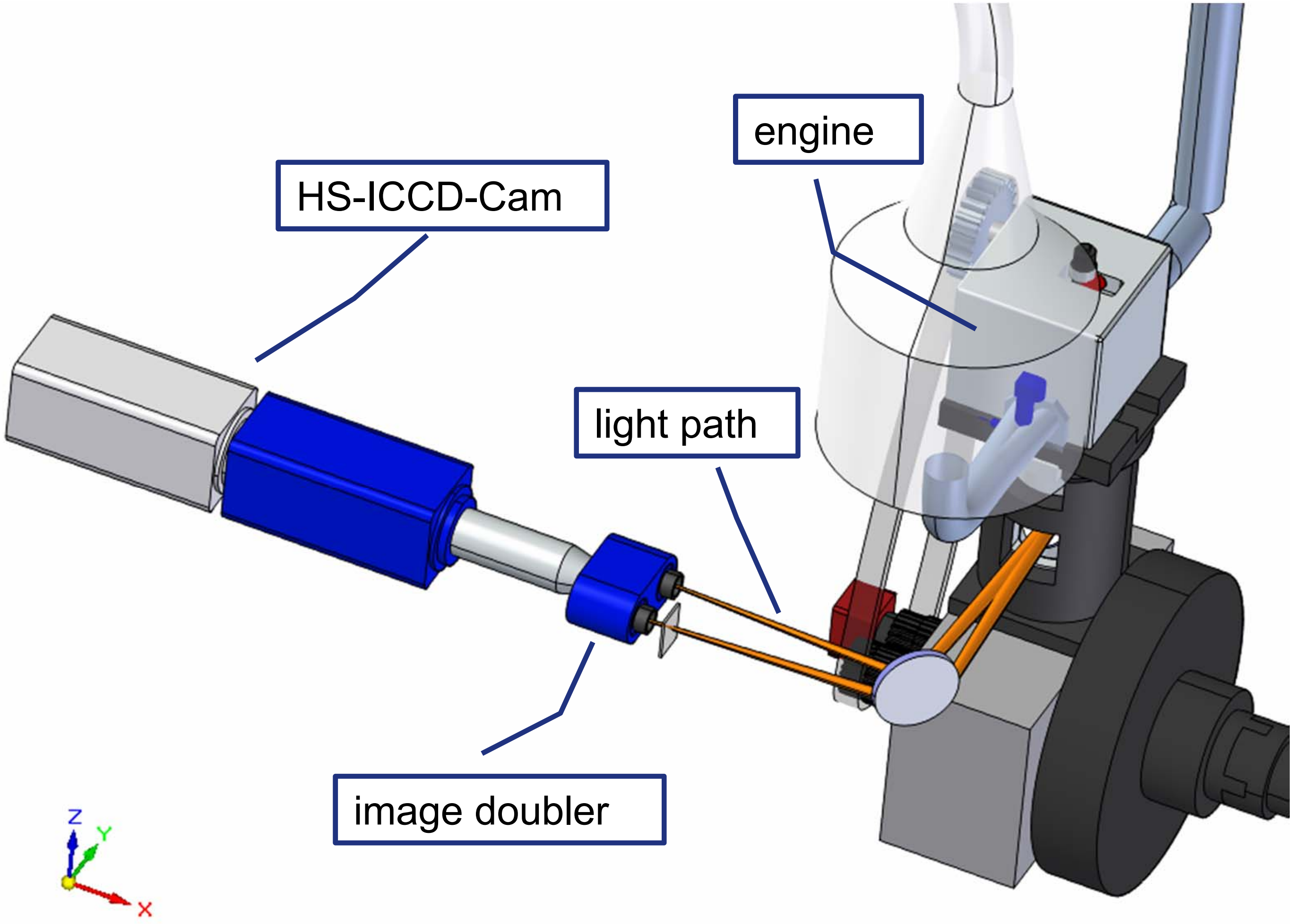

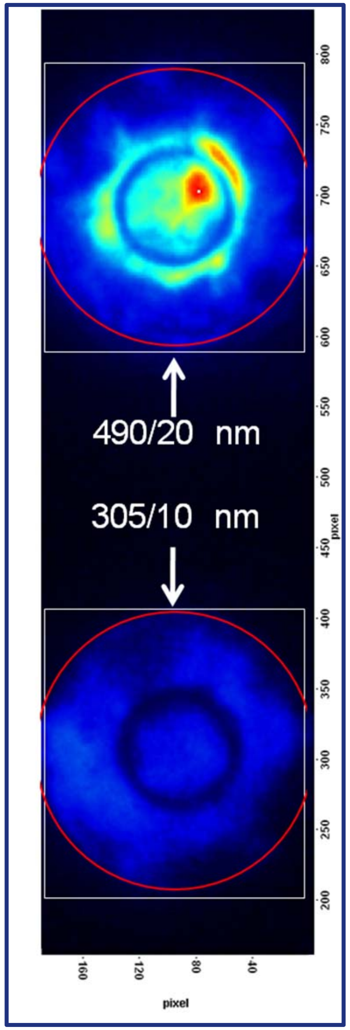

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Integrated Chemiluminescence

OH* (308nm) vs. Soot (490nm)

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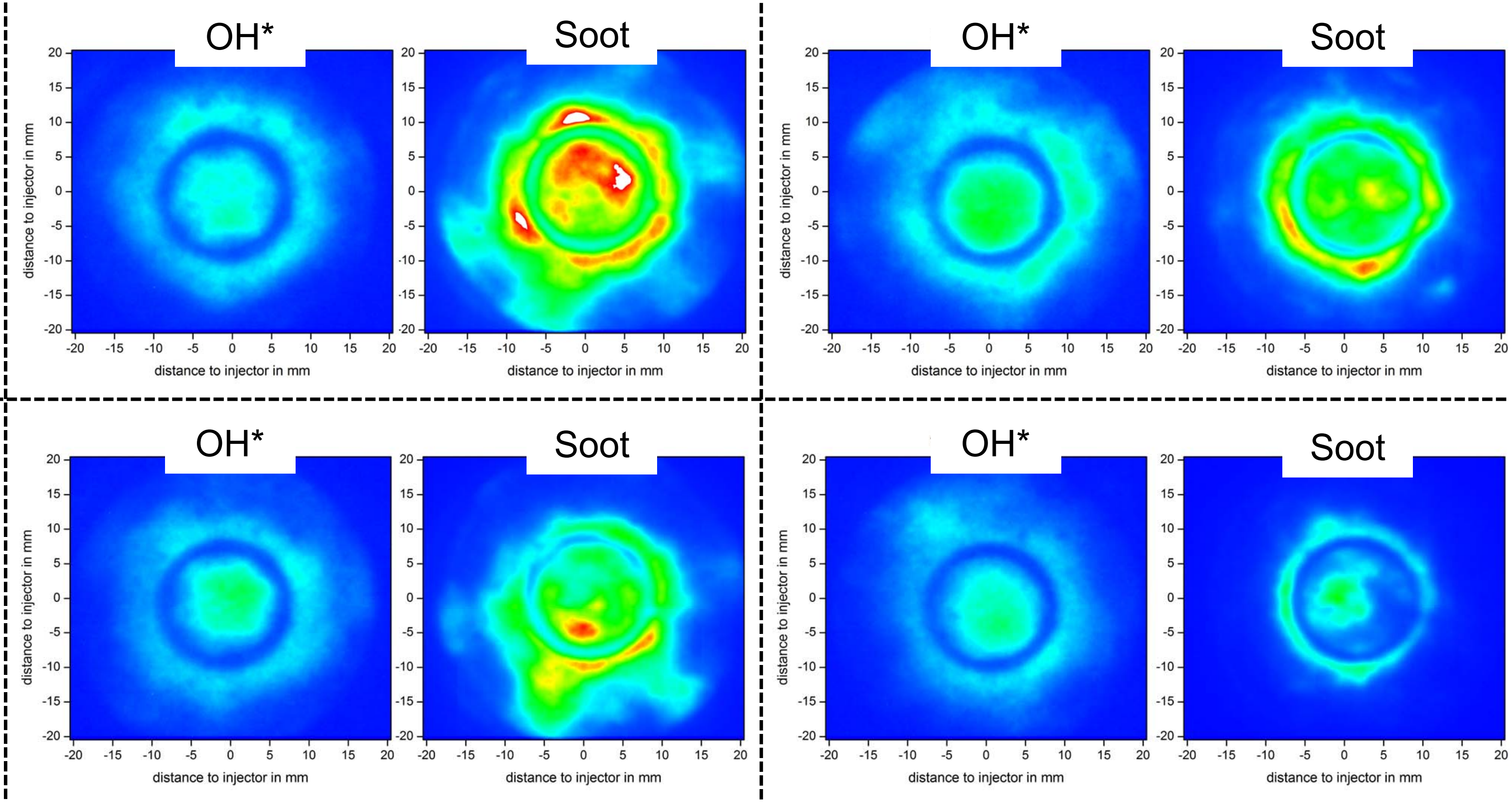
P_{boost}: 0.105 MPa | P_{boost}: 0.145 MPa

P_{inj} 80 MPa

P_{inj} 130MPa



Chemiluminescence of Combustion Ratio OH* (308nm) / Soot (490nm)

Variation of Injection Pressure

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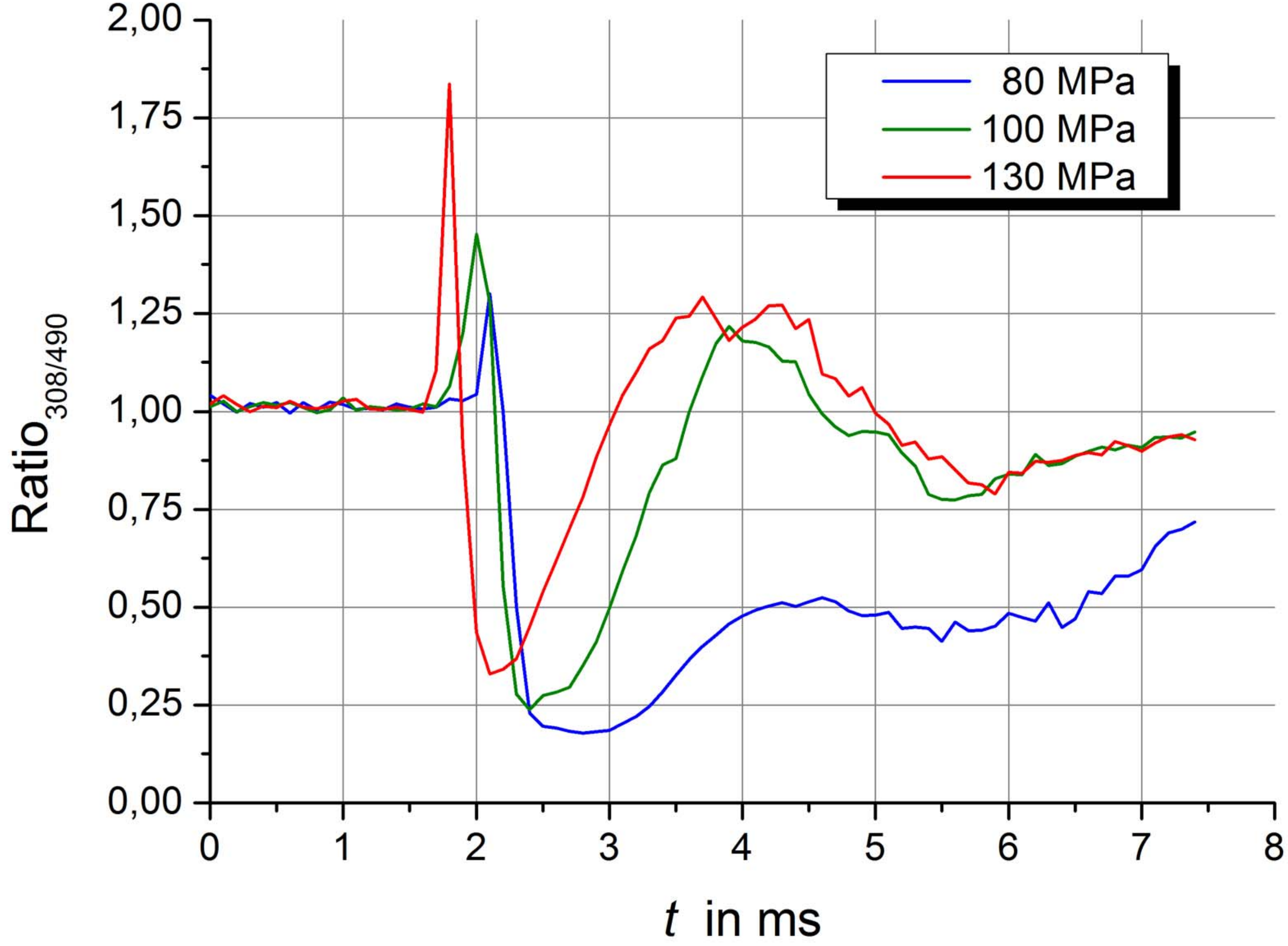

Soot Emissions

Correlation with Chemiluminescence

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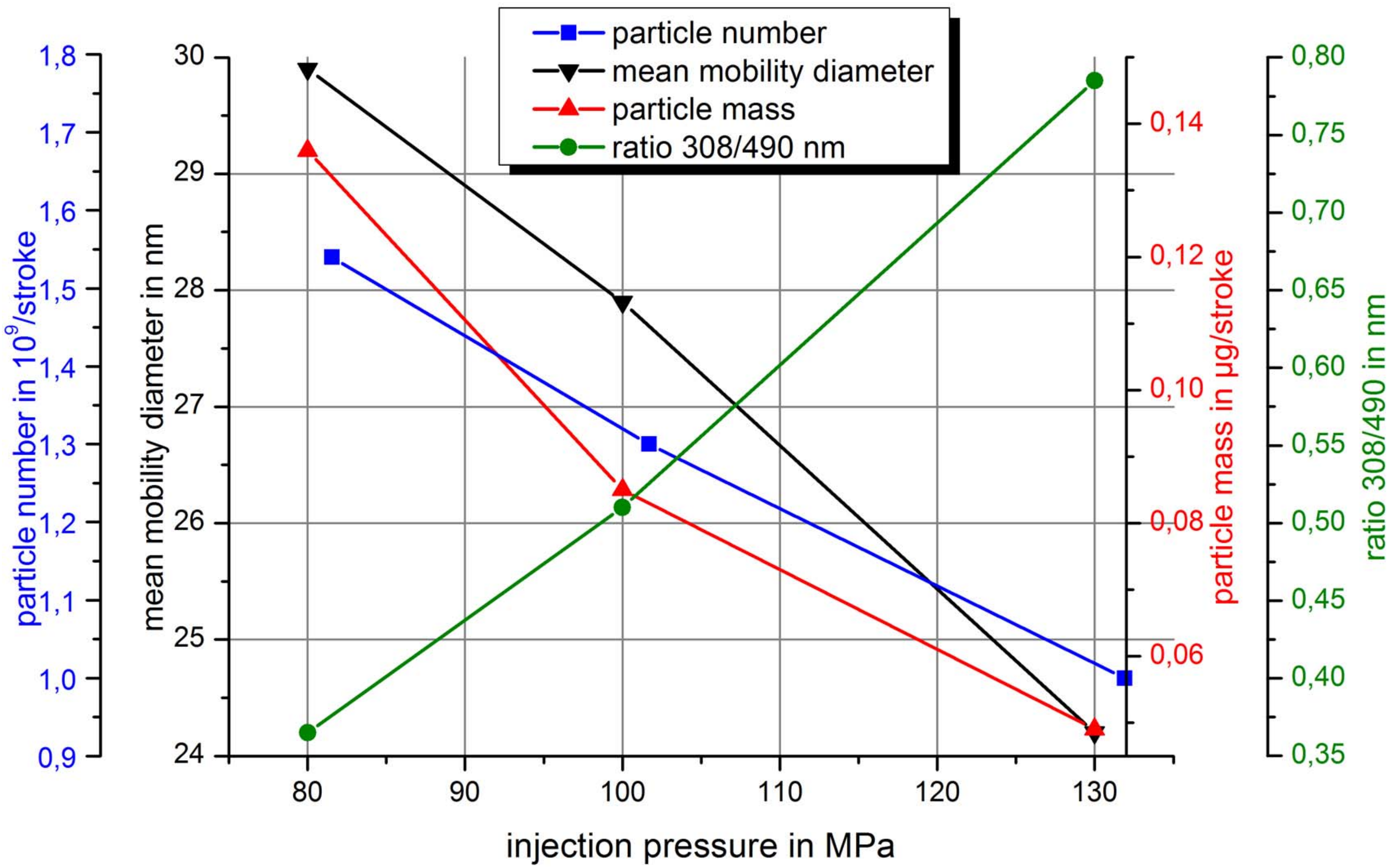

Soot Emissions

HR-TEM Analysis

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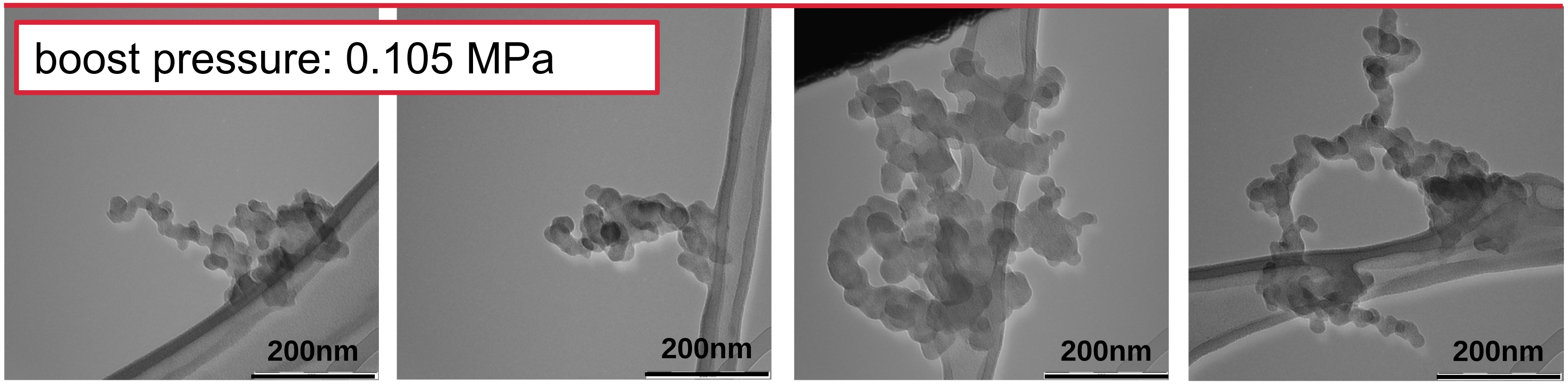
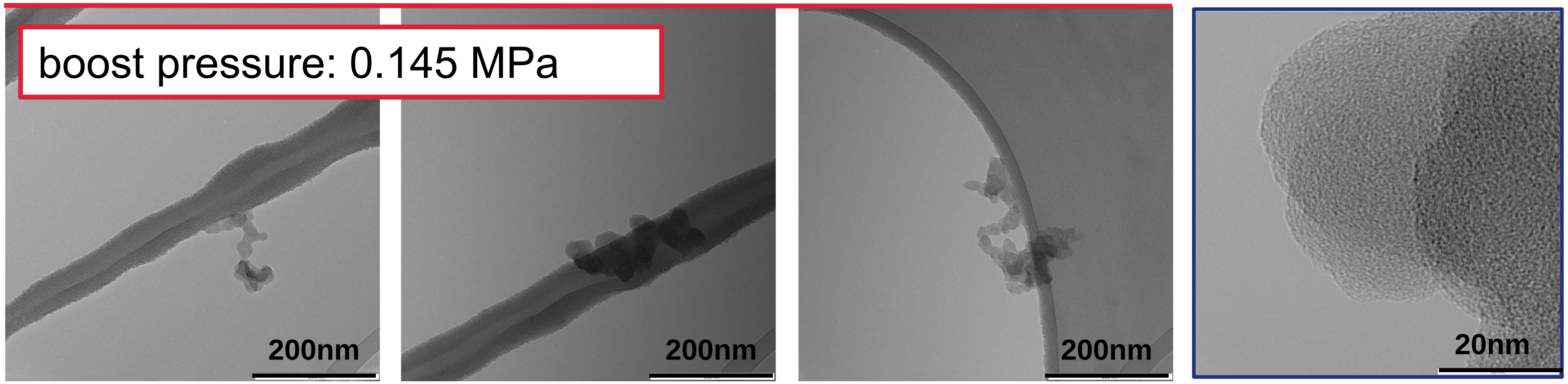
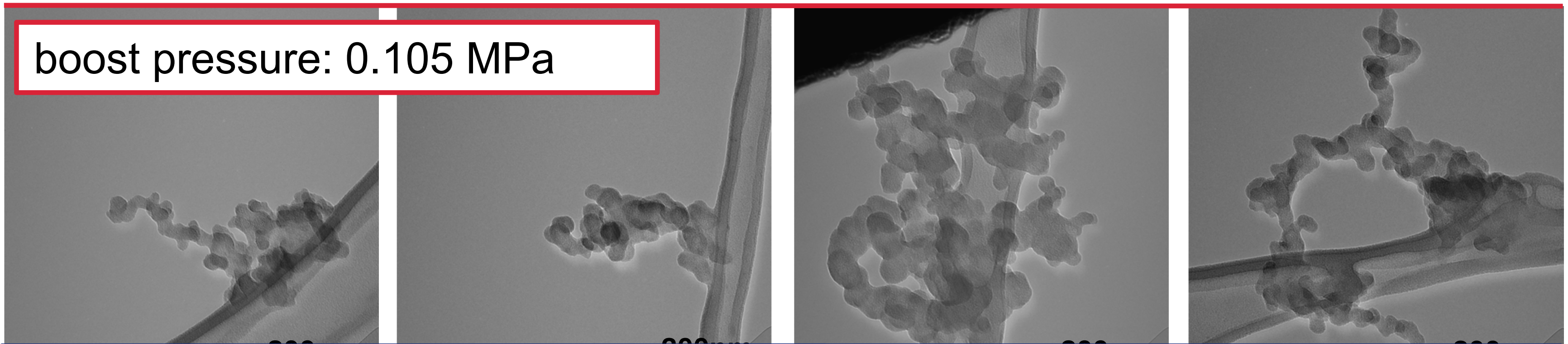


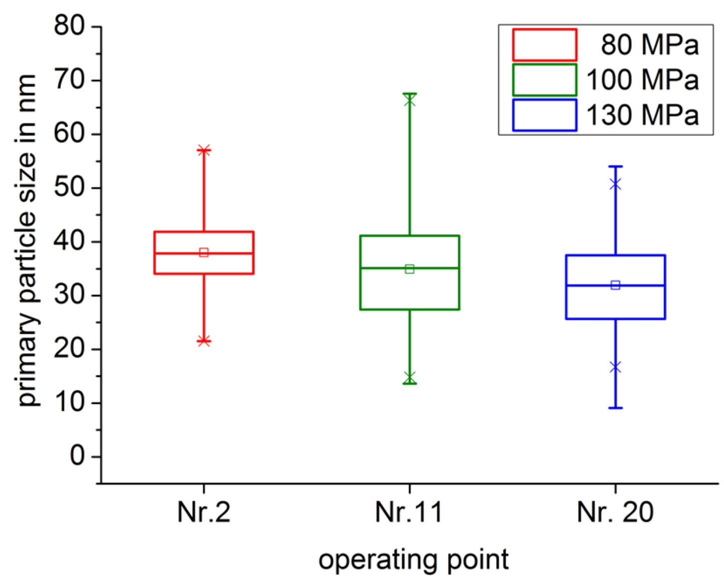


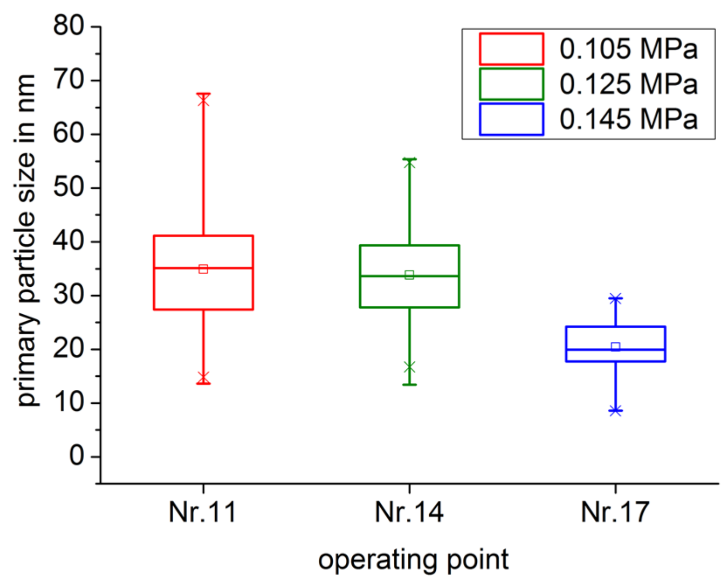
Soot Emissions

HR-TEM Analysis

boost pressure: 0.105 MPa







Summary

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- Rising p_{boost} and p_{inj} lead to decreasing:
  - mean mobility diameter, particle mass and particle number
  - mean primary particle diameter
- Rising p_{boost} and p_{inj} correlates with a rising ratio of OH*/Soot
  - Physical mixing effects
  - chemical effects of oxygen
- Soot agglomerates with smaller primary particles are oxidized at lower temperatures

→ **improved mixing reduces fuel rich zones and local soot production**

→ **OH*-concentration strongly influences soot concentration :**

- **Early OH* reduces soot inception and particle growth**
- **Late OH* increases soot oxidation rate**

Work in Progress

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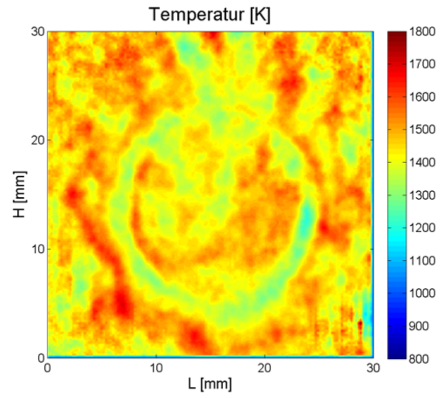
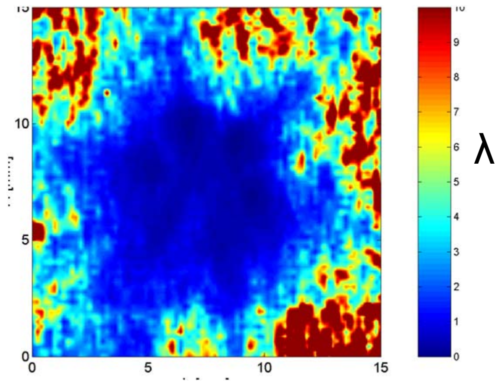
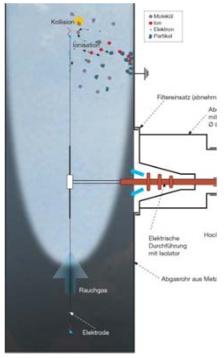
2-color-pyrometry



Mixture formation by LIEF

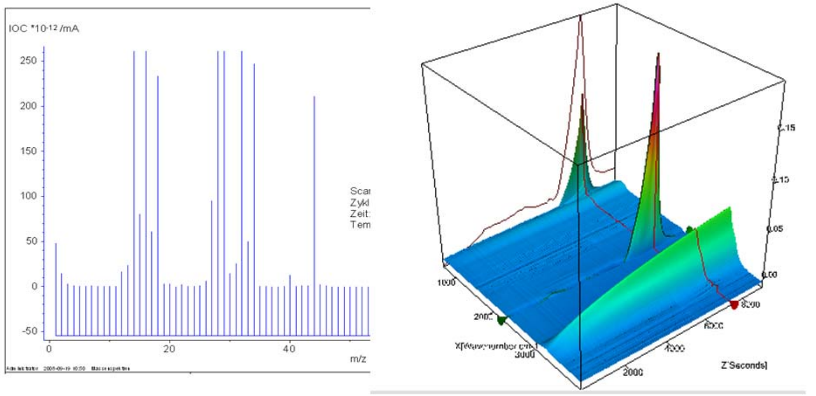


BET Surface Area

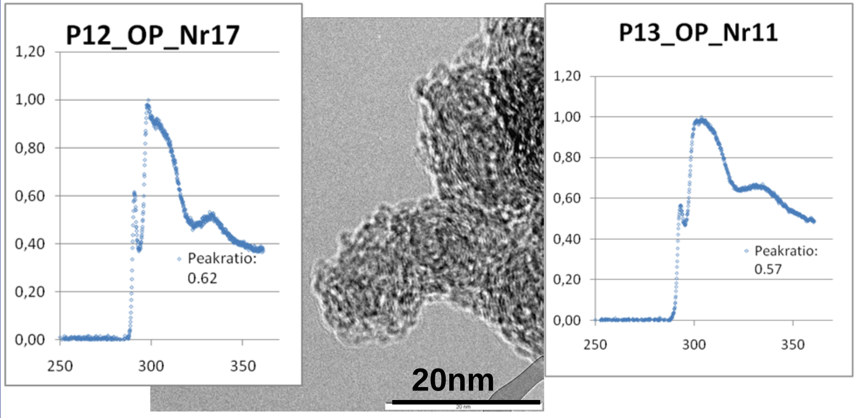


- OP 1 (70 Nm):
  Soot (untreated): ~72 m^2/g
  Soot (preheated): ~94 m^2/g
- OP 3 (210 Nm): ~38 m^2/g
- OP 5 (240 Nm): ~39 m^2/g

TG-FTIR-MS



Electron Energy Loss Spectroscopy



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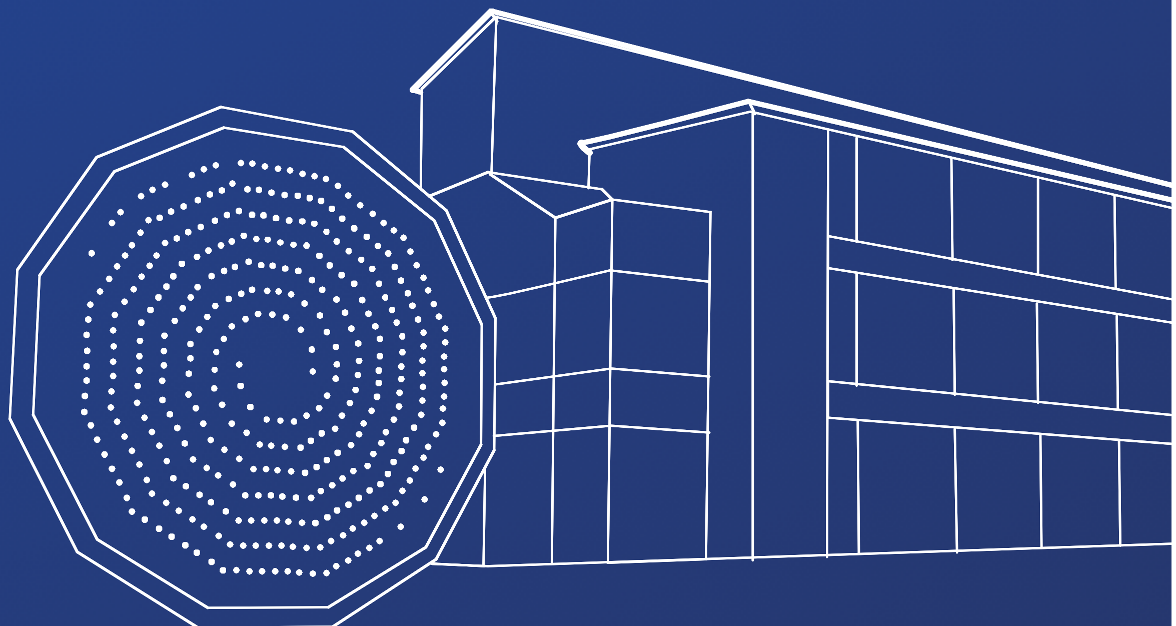
Thank You!



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Highspeed-Imaging

Penetration Depth and Combustion

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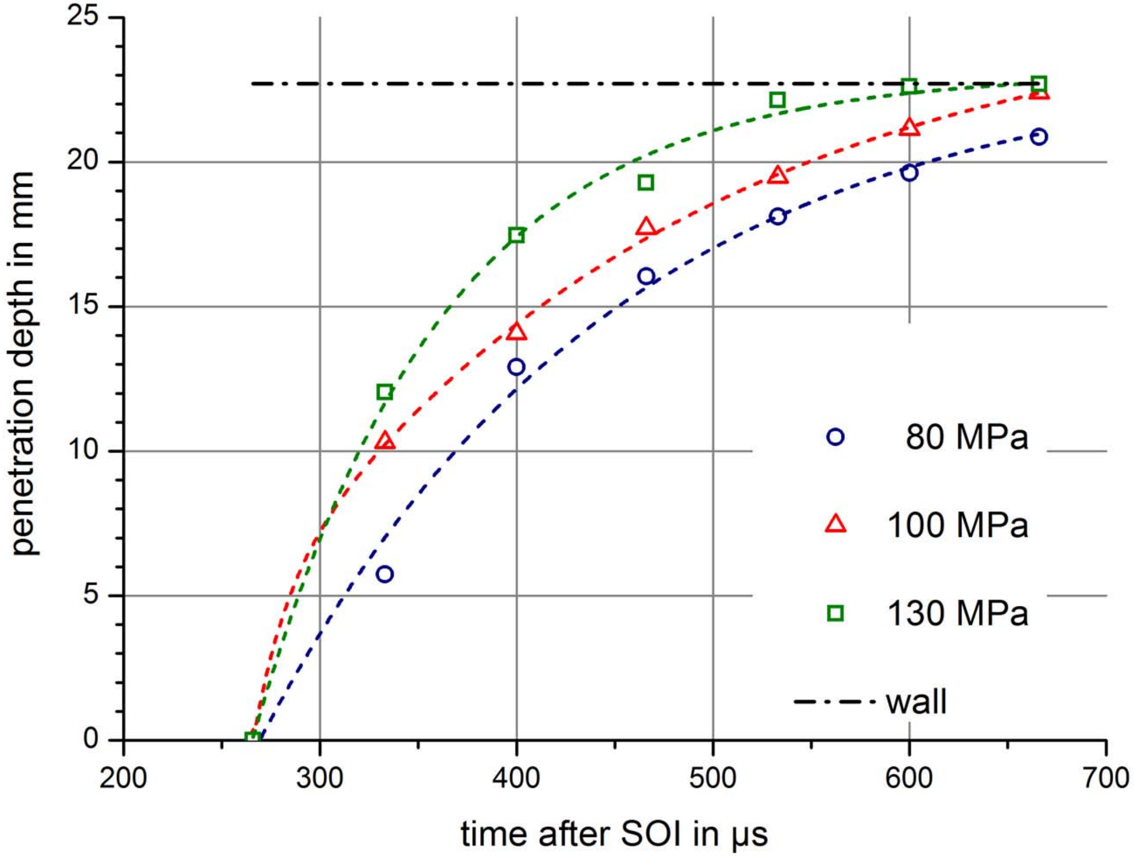

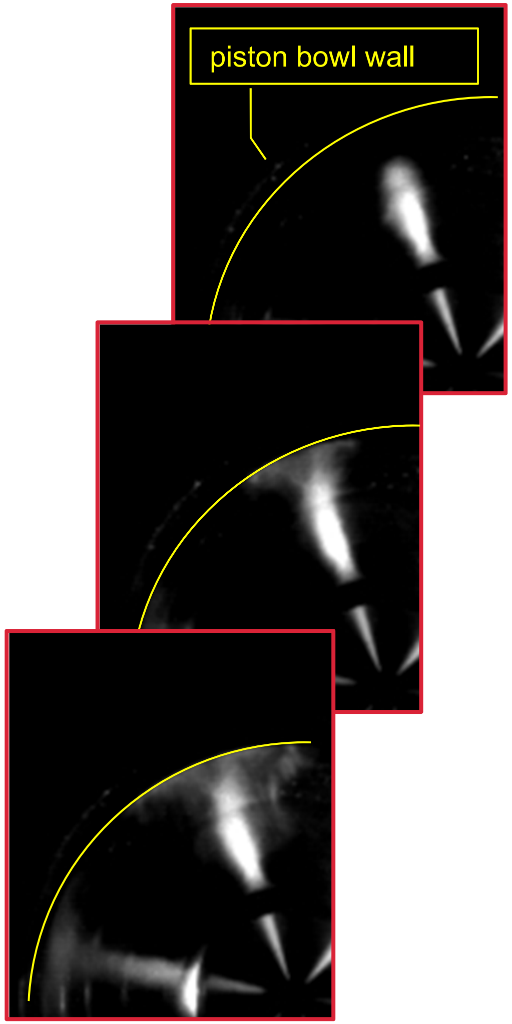

Pressure Indication

Variation of Injection Pressure

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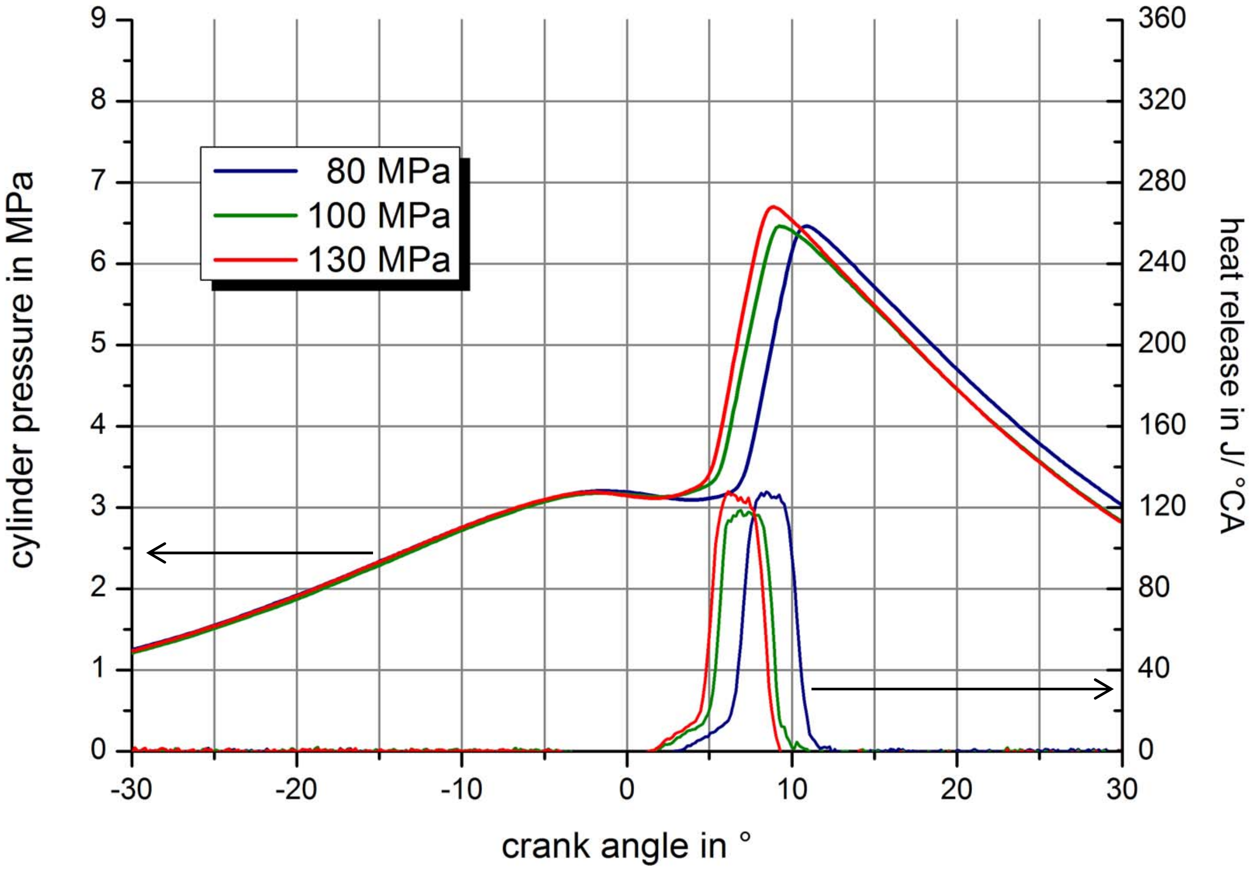
n = 1000/min, p_L = 0.105 MPa, EGR = 0%, ϕ_i = 6 °CA



Injection pressure influences:

- ignition delay
- pressure rise rate
- heat release
- fuel conversion
- max. pressure
- combustion temperature

Evaluation/Overview

Correlation Table

LEHRSTUHL FÜR TECHNISCHE THERMODYNAMIK UND TRANSPORTPROZESSE
PROF. DR.-ING. D. BRÜGGEMANN

Parameter & *range* / Measurement	Speed [1/min]	EGR-rate [%]	Injection pressure [bar]	Boost pressure [bar]	Injectin timing [°CW ATDC]	
	600 → 1000	*0 → 50*	*800 → 1300*	*1,05 → 1,45*	*-10 → -6*	*-6 → -3*
Maximum pressure and burnrate	+	–		+ +	+	+
Thermodynamic and optical ignition delay	–	+	–	– –	–	+
Fuel conversion	+ +	–	+	+ +	+	+
Fuelspray velocity and penetration depth			+ +	–	(+)	(+)
Particle characteristics	– –	+	– –	– –	+	+
OH*-intensity	–	– –	=	+ +	+	–
Soot-intensity	–	=	– –	–	=	=
Ratio OH*/soot	+	–	+ +	+ +	+	–