

Study of Soot Formation and Oxidation in the Engine Combustion Network (ECN), Spray A: Effects of Ambient Temperature and Oxygen Concentration

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ABSTRACT

Within the Engine Combustion Network (ECN) spray combustion research frame, simultaneous line-of-sight laser extinction measurements and laser-induced incandescence (LII) imaging were performed to derive the soot volume fraction (f_v). Experiments are conducted at engine-relevant high-temperature and high-pressure conditions in a constant-volume pre-combustion type vessel. The target condition, called "Spray A", uses well-defined ambient (900 K, 60 bar, 22.8 kg/m³, 15% oxygen) and injector conditions (common rail, 1500 bar, KS1.5/86 nozzle, 0.090 mm orifice diameter, n-dodecane, 363 K).

Extinction measurements are used to calibrate LII images for quantitative soot distribution measurements at cross sections intersecting the spray axis. LII images are taken after the start of injection where quasi-stationary combustion is already established. In addition, by changing the LII timing relative to the injection, the temporal variation of the soot cloud is observed from initial soot formation until soot oxidization. OH-chemiluminescence imaging was used to determine the lift-off length, relative to the soot-forming region and used to interpret the soot measurements.

Results show that Spray A is a moderately sooting flame where signal trapping is not significant, aiding the potential for quantitative soot diagnostics. Maximum soot volume fractions around 2-3 ppm are obtained at the nominal ambient temperature defined for Spray A (i.e. 900 K) that rise to 12 ppm at elevated temperature (1030 K). At 1.5 ms nominal injection duration the Spray A soot cloud is mainly transient. Therefore, an extended injection duration of 4 ms at identical rail pressure was used to characterize the soot structure in quasi-steady mode. Variations of ambient temperature and oxygen concentration are carried out showing effects on soot formation and oxidation that are consistent with the literature.

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INTRODUCTION

The Engine Combustion Network (ECN) aims to improve the fundamental understanding of Diesel spray formation and combustion in engines, and to develop predictive models to optimize these engines. It brings experimental and computational research together for collaborative

comparisons of measured and simulated results at standardized conditions [1,4]. The community has investigated hydraulic properties, spray development, and ignition processes in the target sprays. Datasets from these measurements are now available for the further development of experiments and for the validation of model-based

simulations. Recent experiments [5] have quantified the mixture formation, spray and gas velocities, as well as the local soot distribution.

In engine combustion, soot is of major concern because the emission of particulate matter (PM) is subjected to stringent regulations. Additionally, soot affects the flame temperature via radiation heat transfer from incandescent particles which also implicitly influence NO_x emissions [6]. For engine researchers and manufacturers, these environmental concerns necessitate the understanding of soot processes comprehensively. A quantitative understanding of soot formation enables the development of methodologies to limit PM emissions and to develop CFD (computational fluid dynamics) models that accurately predict soot processes in Diesel combustion.

To avoid the complications of measurements in a real engine and to better decouple the impacts of different variables on fuel sprays, experimental studies in the ECN are conducted at engine-relevant high-temperature and high-pressure conditions in constant-volume pre-combustion type vessels and constant-flow test rigs. The ECN has defined target sprays that can be operated in various facilities available in the contributing labs. The target conditions are confirmed by comparing to existing databases include liquid penetration-length, vapor penetration-length, ignition delay and lift-off length measurements [2,3,4,6]. Well-characterized nominally identical injectors [3] are used in all contributing labs.

Soot investigations in optically-accessible engines as well as high-temperature, high-pressure combustion vessels have shown that soot formation in reactant fuel jets begins shortly after autoignition [5,7,8,9,10,11] at a finite distance from the initial high-heat-release reactions and then continues throughout the mixing-controlled combustion phase [5,9,10,12]. Investigations indicate that mixing-controlled Diesel combustion for quiescent Diesel engine conditions occurs in a lifted, turbulent diffusion flame mode. The diffusion flame takes place at the periphery of the spray. Inside this hollow high-temperature reaction zone, a fuel-rich mixture with relatively lower temperature and slower reaction rate takes place. Fuel and air in this core spray zone start to mix upstream of the flame lift-off, hence the position of lift-off is important for the mixture fraction downstream of this zone. For soot processes local availability of oxygen and temperature in the region are two key parameters. Therefore, the flame structure in the premixed phase and the lift-off lengths are crucial for the soot formation process [1,9,11,12].

Target Spray A (Table 1) acts as a reference case for low-temperature Diesel combustion in engines with moderate exhaust-gas recirculation (EGR) [2]. Extensive information about measurements on Spray A are given on the ECN web-site [5].

This paper addresses the measurement of local soot volume fractions in transient and quasi-stationary cases of Spray A. Measurements for the standard operating conditions

provide a basis for understanding the ambient and injector boundary conditions, and also provide the basis for interpreting the soot measurement results. Additional measurements with parametric variations (cf. Table 2) of the standard conditions permit investigation of the effect of the various operating conditions on the lift-off length and the soot level. The differences between nominal and actual gas temperatures mentioned in Table 2 will be explained in the results section.

Table 1. Operating conditions for Spray A

Ambient gas temperature	900 K
Ambient gas pressure	near 6.0 MPa
Ambient gas density	22.8 kg/m ³
Ambient gas oxygen	15% O ₂
Ambient gas velocity	Near-quiescent, <1 m/s
Nozzle diameter	0.090 mm
Nozzle K factor	1.5
Discharge coefficient	$C_d = 0.86$ (at 100 bar)
Number of holes	1 (single hole)
Fuel injection pressure	1500 bar
Fuel	n-dodecane
Fuel temperature at nozzle	363 K
Injection duration	1.5 ms

Table 2. Parametric variations of the Spray A operating conditions

Nominal ambient gas temperature (actual)	800 (830), 850 (880), 900 (930), 1000 (1030) K
Ambient gas oxygen	11, 13, 15, 21% O ₂
Injection duration	1.5, 4 ms

In this study, the Laser Extinction Method (LEM) [1,10,13,14,15,16,17] is used to quantitatively measure the soot concentration along a line in the spray. This information is coupled with Laser-Induced Incandescence (LII) imaging [1,6,7,8,9,10,11,12,13,16,17,18,19,20,21,22] for the quantification of the soot volume fraction maps for cross-sections through the flame. One important aim of the measurements presented here is to explore the suitability of Spray A to LII and LEM diagnostics. Therefore, effects of laser and signal trapping have been quantified (see below in the Diagnostics and Measurements section).

For each boundary condition, OH chemiluminescence imaging was used to determine the lift-off length. The following diagnostics and measurements section introduces the measurement principles. The experimental set-up and the validation tests are described in the experimental chapter. Experimental results for Spray A and its parametric variants are given in the results section.

DIAGNOSTICS AND MEASUREMENTS

Laser Extinction Method (LEM)

LEM is a non-intrusive zero-dimensional line-of-sight technique based on the attenuation of light when a laser beam passes through a particle cloud. The attenuation of light is caused both by elastic scattering and absorption in the particles [15,16]. The relation between the incident light intensity (I_o) and the light intensity after passing through the particle cloud (I) is given by Lambert-Beer's law, Eq. (1):

$$\frac{I}{I_o} = \exp(-KL_{LEM}) \quad (1)$$

where L [m] is the path length through the cloud and K [m⁻¹] is the mean extinction coefficient along this path length. Following Mie theory [10,16,23] the path-length-averaged extinction coefficient can then be related to the soot volume fraction as given in Eq. (2):

$$K = \frac{k_e}{\lambda} f_v \quad (2)$$

where λ [m] is the laser wavelength and k_e is a non-dimensional optical coefficient that depends on the soot morphology and the particle size. This coefficient can be related to the refractive index function at the detection wavelength. In this work, a value of $k_e = 8.7$ is used [19,26] which is assumed to be uniform throughout the spray for all operating conditions [12]. The accuracy of this value is debated in the literature and a systematic error on this value must be taken into account [16,24].

Laser-Induced Incandescence (LII)

LII is an optical diagnostics technique which uses a pulsed laser to heat soot particles to temperatures significantly above the ambient temperature and uses photo-detectors to record the incandescence emitted from these heated particles [28]. LII can be applied either in a point-wise or an imaging manner. For soot volume fraction measurements, the latter is mostly preferred, while time-resolved measurements for determining the particle size typically are restricted to point measurements [29,30,31].

The incandescence radiation is a blackbody-like radiation and strongly depends on the temperature of the soot particles. Therefore, the signal intensity per particle increases with the energy fluence (i.e., pulse energy per area) of the pulsed heating laser. This correlation is valid (but not linear) until soot sublimates at around 4500 K. Beyond this point, excess energy does not further increase the particle temperature or LII signal. In contrary, sublimation of particles leads to a decrease in the signal due to rapid evaporation of the radiating particles.

For soot "concentration" measurements it is important that the LII signal is proportional to the amount of soot

particles and their size. Assuming a homogenous temperature distribution of the laser-heated soot particles in a measurement volume the signal is proportional to the soot volume fraction f_v (i.e., the relative volume occupied by the particles in the respective gas sample). f_v is presented in terms of parts per million (ppm).

Transmission electron microscope (TEM) analysis [32,33,34] shows that soot particles consist of agglomerated nearly spherical primary particles. Within one measurement location it is often assumed that the size of the primary particles and the aggregates are nearly the same [29,30]. With this simplification, the relation between the soot volume fraction (f_v) and the LII signal (I_{LII}) turns into a linear form as given in Eq. (3)

$$f_v = \alpha I_{LII} \quad (3)$$

where I_{LII} can be acquired by a camera and can be written in photon counts unit. The assumption of a homogenous temperature distribution among the heated soot particles is crucial for the accuracy of the volume fraction measurement. Considering the possible non-homogeneities in the spatial profile of the heating laser and the non-homogeneous initial temperature of particles prior to the laser pulse, maintaining a uniform temperature in the measurement volume requires an additional effort in data analysis. One simple way of assuring this uniformity is bringing the particles into the sublimation regime with excessive energy so that all particles are heated to the sublimation temperature. However, the laser profile has a key influence on the LII signal in the sublimation regime. As mentioned above, rapidly sublimating particles cause a decrease in the overall LII signal when all particles contributing to the signal are exposed to the same laser fluence (top-hat laser profile in direction of signal detection). An (approximately) stable LII signal intensity at increasing laser power can be maintained by a laser sheet with a Gaussian spatial profile. The reduced signal caused by the sublimating particles in the center line of the laser beam is compensated by the increasing temperature of particles exposed to wings of laser beam. The resulting "plateau zone" where the signal is insensitive to variations in laser fluence is often used in cases where beam attenuation and laser pulse-to-pulse energy variation needs to be compensated [36]. The threshold for this signal saturation regime is given as ~ 0.3 J/cm² at 1064 nm [23,24]. Detailed descriptions of the effects of the spatial laser profile have been reported [23,24]. The LII measurements carried out within this paper use the high-fluence approach with a laser light sheet where the laser fluence distribution is approximately Gaussian in the direction of signal detection.

Coupling LII and LEM

LEM provides quantitative but path-integrated information about the soot volume fraction. LII, on the other hand, yields information about the spatial distribution of the soot volume fraction in a selected cross section but is not

quantitative per se. By aligning the two systems in a way that the LEM line-of-sight is part of the LII cross-sectional area it is possible to use the LEM result to calibrate the LII signal integrated along the same direction of LEM pathway as long as the LII image covers a cross-section through the entire soot cloud. In a turbulent environment, the soot volume fraction at a given location can vary considerably with time. Therefore, it is important to consider the different temporal characteristics of LII and LEM measurements. While the LII signal shows the instantaneous spatial distribution of soot, LEM depends on the soot during the integration time (that can be considerably longer). For minimizing the effect of this, LEM signals recorded within short integration times at nearly identical instants should be used for the calibration of the LII images. In contrast, a phase-averaged calibration in a quasi-stationary part of the spray has been used in [6].

From the combined measurement, a calibration factor is derived that connects the LII signal intensity integrated along the path of the LEM laser and the laser attenuation and the laser attenuation with the soot volume fraction. The LII signal along the LEM laser path is integrated according to Eq. (4).

$$KL_{Lu} = \int_0^L I_{LII} dx \quad (4)$$

By combining Eq. (1), (2), (3) and (4) one can calculate the linear calibration constant α in Eq. (3) and use Eq. (5) for soot the determination of volume fractions for the entire LII image [18].

$$f_v(x, y) = \alpha I_{LII}(x, y) = \frac{\lambda}{k_e} \frac{KL_{LEM}}{KL_{LII}} I_{LII}(x, y) \quad (5)$$

In Figure 1, simultaneously recorded KL_{LII} and KL_{LEM} values for over 100 spray events are presented. A linear curve-fit to the entire data set (bold line) and two linear curve-fits to lower and higher edges of the data envelope (dashed-lines) based on the ordinary least squares solution are also shown. All these curve-fits are also forced to intersect the origin. The data was collected on a single distance from the orifice ($x = 58$ mm) at the spray axis; details about the experiment will be given in the next section. Note that these measurements were conducted for calibration purposes and to allow for correlation between LII and LEM on a larger range of operating conditions and soot volume fractions. Therefore, ambient temperatures were varied for the individual measurements around the given Spray A target (± 100 K).

The coefficient of determination for the entire data set linear curve-fit is calculated as 0.54 where the slope of the curve is calculated as 90.9. It can be speculated that this data set acknowledges the linear assumption between LII and LEM signals. However, the spread of the measurement points around the mean line is significant and represents the combined uncertainties of the measurement strategies applied here. To show the effect of this uncertainty on LII-LEM coupling quantitatively, the slopes of linear curve-fits to maximum and minimum data points are measured as 145.5

and 36.5, respectively. For these fits, data points are manually chosen from the scatter plot. Concerning LEM, although the experiment was designed to minimize uncertainties due to beam steering and the natural soot incandescence (details will be given in the next section), interference with forward-scattered laser light may remain [18]. The LEM detection is also affected by instabilities in the alignment and by variable transmission of the windows. Additionally, LEM uses an integration time of $2 \mu s$ and thus does not observe exactly the same conditions as LII (with a detection time in the tens of nanosecond regime) which can affect the correlation in a strongly turbulent and spatially inhomogeneous case. The magnitude of the total uncertainty on LEM was measured as 5% in a previous study not including the temporal variations that might occur in a turbulent application.

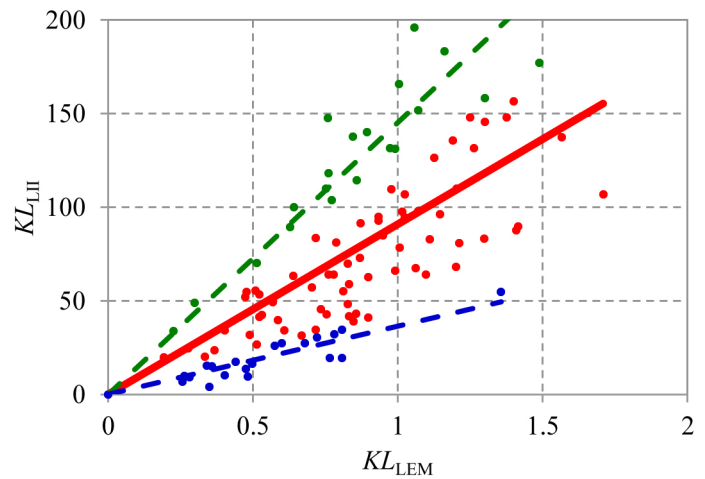


Figure 1. Correlation between KL_{LII} and KL_{LEM} over 100 individual measurements.

For LII, our present experiment was affected by a ± 20 ns random jitter of the camera gate timing relative to the laser pulse which was noticed only after finalizing the measurements. The lifetime of the LII signal under the conditions of our experiments was less than 100 ns (cf. Figure 2). The LII signal is detected within a 50 ns gate. Its beginning is set 15 ns before the laser Q-switch trigger. The intended position of the gate is shown by the red dotted lines in the figure. In case of a 20 ns positive jitter, shown with dashed green lines, the collected signal increases by $\sim 5\%$. In the case of a 20 ns negative jitter, the collected signal reduces $\sim 50\%$ (dash-dotted, black lines). This jitter was caused by an electronic problem in the timing circuit and is thus responsible to a large extent for the deviations seen in Fig. 1. With an improved system, a correlation like that demonstrated in [18] is expected.

Assessing laser and signal attenuation

Under conventional Diesel spray conditions, high soot concentrations can lead to severe attenuation of the LII signal and the laser light. In case of randomly distributed soot between the laser-heated plane and the camera this signal

trapping is difficult to correct for. Attenuation of the laser light on the other hand prevents the application of low-fluence LII (where the signal depends on the knowledge of local laser intensity) and even high-fluence LII in the plateau zone can be limited in its applicable range because of the laser intensity can fall below the plateau fluence after traveling through a dense soot cloud for a longer distance. Both effects limit the applicability of LII under conditions with high soot load and larger geometric dimensions [37].

Thus, a verification of the applicability of LII diagnostics is required for the present case. From Figure 1 it can be derived that Spray A and its variants provides moderately sooting conditions with limited laser attenuation and signal trapping that allow the application of LII. In case of signal trapping, a systematic decrease in KL_{LII} would take place for increasing soot concentration showing a systematic deviation from the linear correlation with increasing KL_{LEM} . Furthermore, it has been found that at the given axial distance ($x = 58$ mm) of Spray A, laser intensity loss due to extinction is less than 35% leading a KL_{LEM} roughly below 0.5. Compared to KL_{LEM} values in the literature [12] where signal trapping takes place, signal trapping is not expected at Spray A conditions.

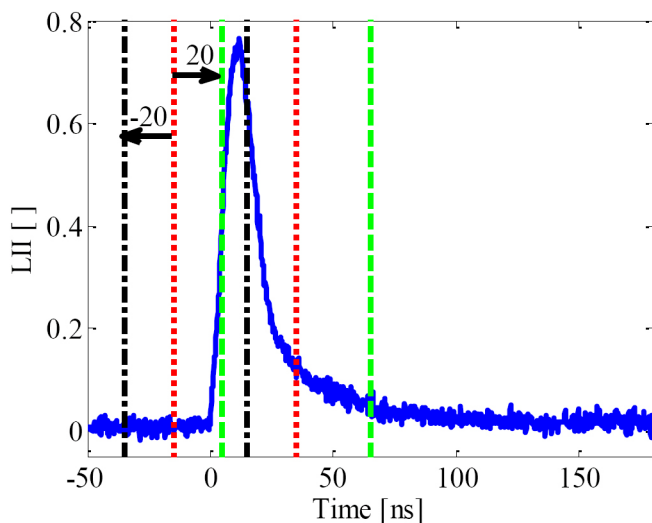


Figure 2. Temporal variation of the LII signal. The red dotted lines show the intended temporal position of the 50-ns detection gate, black (dash-dotted) and green (dashed) lines indicate the shift of the gate caused by the unintended ± 20 ns jitter that occurred in the present measurements.

OH Chemiluminescence

In near-stoichiometric, high-heat-release chemical reactions, excited-state OH radicals (OH^*) are formed as intermediates. Unlike ground-state OH, which exists as an equilibrium product in regions of high temperature, including hot exhaust gases, OH^* only exists in the flame zone [12] and emits chemiluminescence at around 310 nm. Thus, with an

appropriate band-pass filter it is possible to locate zones that can be correlated with near-stoichiometric, high-heat-release conditions [7,10,31,32]. Because stoichiometric combustion is expected at the flame base [10,33], i.e., at the lift-off location, chemiluminescence imaging is also the appropriate method to identify the lift-off length of a spray flame. During the quasi-stationary phase of the spray, the lift-off length is not varying systematically. However, small deviations occur due to turbulent fluctuations. In order to suppress these fluctuations and achieve a steady lift-off length measurement, time-averaged imaging is employed via longer camera gates (500 μ s) within the quasi-stationary phase of the flame.

Figure 3 shows an OH^* -chemiluminescence image. OH^* chemiluminescence begins at a well-defined distance from the injector orifice, initially rising very sharply, and then leveling off. Two lobes of intense chemiluminescence on the top and bottom of the jet are typical near the lift-off length. The lift-off length was determined by finding the distances between the injector and the first axial locations above and below the spray centerline with an intensity greater than approximately 50% of the leveling-off value. The average of these two axial distances is defined as the lift-off length [5]. The intensity increase downstream is due to broad-band blackbody-like radiation of soot particles.

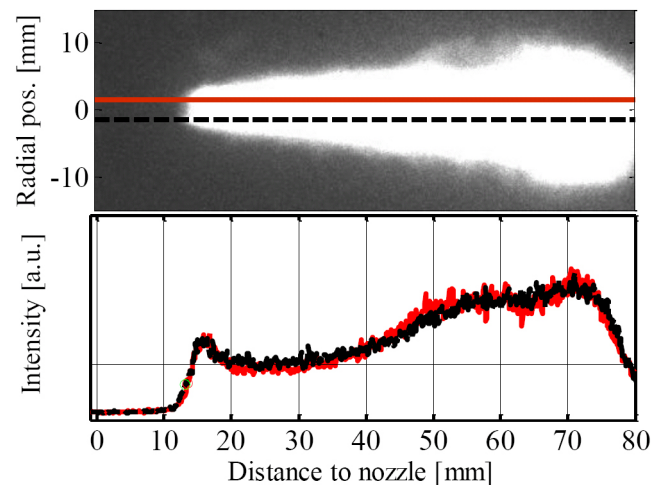


Figure 3. OH^* -chemiluminescence imaging of Spray A. Intensity profiles along the lines indicated in the image show the rapid intensity increase that allows identifying the lift-off length. The signal increase towards the right is due to interference from soot incandescence.

Determination of the Local Fuel/Air Ratio

An estimate of the amount of air entrained into a fuel jet was used to help analyze the soot and lift-off measurements. As shown in Figure 3, Spray A is a lifted flame. The amount of fuel and air premixing that occurs upstream of the lift-off length affects the composition of the reactants downstream of the lift-off. Because soot formation and burnout is directly related to oxygen availability in a region, air entrainment into the jet is crucial for the soot formation rate and its location.