CHARACTERIZATION OF CARBONACEOUS PARTICLES BY TIME-RESOLVED LASER-INDUCED INCANDESCENCE

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Abstract

Online-characterization of carbonaceous nanoscaled particles is important in different application fields, e.g., in carbon black production or in automobile raw exhaust. For the determination of mass concentration and primary particle size respectively specific surface area, a possible in situ measurement technique is time-resolved laser-induced incandescence (TIRE-LII). The basic principle of this technique is to heat the carbonaceous particles up to their sublimation temperature by means of a high-energetic laser pulse and the subsequent detection and analysis of the spectral and time-resolved enhanced thermal radiation. At later times after the laser pulse heat conduction to the ambient gas is the dominant heat loss mechanism and so particles with different specific surface area cool down differently. From the temporal signal decay the size distribution of the primary particles can be derived. Furthermore, the signal maximum is proportional to the mass concentration. Here, an overview is given on current potentials and problems using this measurement technique for the comprehensive characterization of carbon black and Diesel soot. Measurements have been performed at different carbon black reactors on the one hand and directly in the Diesel raw exhaust on the other hand. Thereby, the results were compared with established measurement methods like TEM, conventional adsorption analysis and gravimetrical techniques. Furthermore, a new approach applying this technique also for suspended particles in liquids is shown.

Introduction

The industrial production of nanoscaled particles has become very important in recent years due to the increased number of application areas of these substances. Exemplarily, carbon blacks are important nanoparticles, which are included as crucial additives in tires, paints and varnishes. One particle property, which determines together with other attributes the quality of these products, is the primary particle size. To measure and control this quantity during the production process, an appropriate size characterization method is required. For this, online and in situ process monitoring is necessary. Other areas in which the characterization of carbonaceous particles plays an important role, are soot measurements in Diesel raw exhaust and in ambient air.

Unfortunately, current measurement techniques like transmission electron microscopy (TEM), chemical analysis, dynamic light scattering (DLS), impactors or scanning mobility particle sizers (SMPS) do not allow fast, non-intrusive or selective determination, respectively. However, this is possible using time-resolved laser-induced incandescence (TIRE-LII). Laser-induced incandescence (LII) has been proven to be a powerful measurement tool for soot characterization in different flames for many years. However, in recent years it has been further developed for carbonaceous particle characterization in technical important applications (Santoro & Shaddix, 2002; Leipertz & Dankers, 2003; Schulz et al., 2006). Accessible parameters are besides the volume concentration, the primary particle diameter respectively the specific surface area. Meanwhile, it has been extended to evaluate even the size distribution of particles (Dankers & Leipertz, 2004). The potential and features of this technique are presented here exemplarily by a few selected measurements, like the online primary particle size characterisation within a carbon black production reactor (Dankers et al., 2003) or the soot analysis in engine exhaust (Schraml et al., 1999 & 2004). In particular, for engine applications the method has been implemented to a robust sensor, which allows a reliable soot measurement even for modern low emission vehicles. The system has a high temporal resolution of up to 20 Hz and a very low detection limit of just a few µg/m³, which qualifies the system also for immission measurement and environmental health control. Moreover, the potential of this measurement technique for particle sizing in suspensions is regarded.

Measurement Principle

The basic principle of laser-induced incandescence (LII) is the rapid heating of carbonaceous nanoparticles up to their sublimation temperature within a few nanoseconds by means of a short intense laser pulse, and the subsequent detection and evaluation of the enhanced thermal radiation. At first, the particles are heated up by absorbing the laser radiation, which results in an increased internal energy. The maximum particle temperatures reached are about 4000-4500 K for a short time period of several nanoseconds. The subsequent heat loss is dominated by different mechanisms, mainly sublimation, heat conduction and radiation. These are responsible for the time-dependent decrease of the particle temperature. By setting up the energy balance for one single particle, the resulting differential equation yields to

$$C_{abs} \cdot E_i - \Lambda (T - T_0) \pi d_p^{\,2} + \frac{\Delta H_S}{M} \cdot \frac{dm}{dt} - \pi d_p^{\,2} \int \varepsilon(d_p,\lambda) M_\lambda^{\,b}(T,\lambda) d\lambda - \frac{\pi d_p^{\,3}}{6} \rho \cdot c_P \cdot \frac{dT}{dt} = 0.$$

The temporally resolved particle temperature can be calculated by solving the differential equation including the laser absorption in the Rayleigh-Regime (absorption efficiency C_{abs}, laser energy E_i), heat conduction (Knudsen number dependent heat transfer coefficient Λ, particle temperature T, temperature T_0 of the surrounding gas), sublimation (sublimation enthalpy ΔH_V, molar mass of carbon M) and radiation (emission coefficient ε, spectral energy density $M_\lambda^{\,b}$, wavelength λ), as well as the change of the internal energy (mass density ρ, specific heat c_P). Thereby, the equation given above includes the assumption of spherical primary particles, which have only point contact between each other inside the aggregates. Applying Planck's radiation law the detectable signal can be determined (Santoro & Shaddix, 2002; Michelsen, 2003).

One attribute of this measurement technique is the non-linear behavior of the maximum peak signal and the excitation laser energy. An initial increase in laser fluence leads to a rapid rise in the peak LII signal, whereby it levels off to a so-called "plateau" region at a laser fluence of about 0.2 J/cm², from which no significant signal change is evident (Will et al., 1998). This observation is found in previous studies on aerosols and flames (Snelling et al., 2000; Bladh & Bengtsson, 2004). Noteworthy is the plateau width which is strongly dependent on the laser spatial energy distribution. In the plateau region the particles reach their peak temperature, sublimation acting as the limiting factor. At higher laser excitations the particles shrink due to surface sublimation of carbonaceous matter that, in turn, leads to lower signal intensities. Moreover, due to high laser fluences morphological changes can also occur possibly modifying the particle properties (Vander Wal et al., 1998 & 1999). For technical applications this "plateau" region is of advantage as changes in the laser fluence do not affect the signal behavior.

Under the assumption of sufficient high laser excitation energies the heat loss at peak particle temperature is dominated by sublimation. Hence, the maximal signal is proportional to the particle volume and by assuming a constant density also to the mass concentration (Melton, 1984; Will et al., 1995; Mewes & Seitzman, 1997). This incandescence signal is particularly selective on the elemental carbon (EC) mass fraction as volatile exhaust components will vaporize at such extremely high temperatures without emitting thermal radiation. A great benefit, which can be taken from this behavior, is the applicability to conditions at which liquid droplets are present.

At later times after the laser pulse, with falling particle temperature, heat conduction becomes the most important heat loss mechanism (Will et al., 1995), and therefore, particles with different specific surface areas cool down differently, which can directly be regarded in **Figure 1** (Roth & Filippov, 1996; Will et al., 1998).

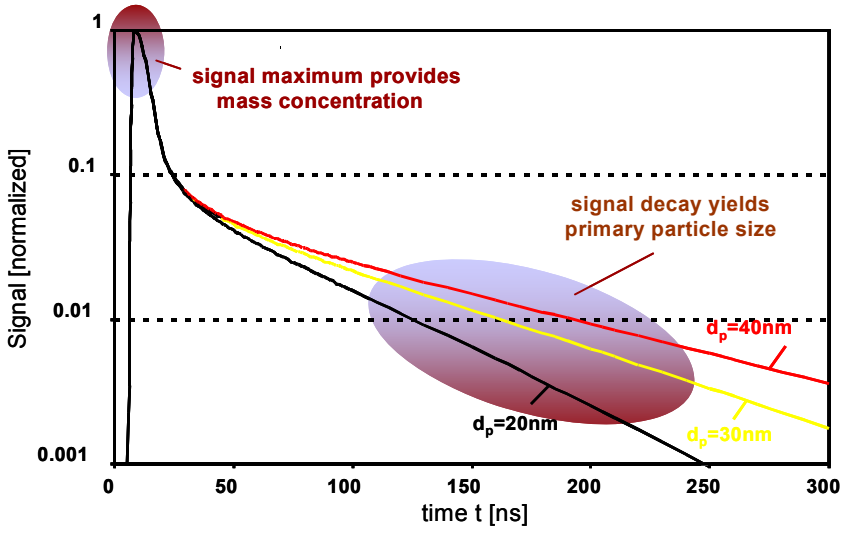

Figure 1. Temporal curve of the LII signal for different primary particle diameters

In the presented measurements temporal-resolved LII signals were detected with a fast photomultiplier tube, which guarantees a good signal-to-noise ratio. For the determination of a mean primary particle diameter d_p only one signal decay time τ has to be determined from experimental LII signal curves by an exponential fit in the time interval, in which heat conduction dominates the particle cooling (**Figure 2**). Under consideration of the surrounding temperature, τ can be compared with numerically calculated signal decay times assuming a monodisperse particle ensemble and a mean particle

diameter can be associated unambiguously. Hence, a simultaneous temperature determination of the ambient gas is necessary.

This approach is sufficient if the size distribution is narrow in technical systems, which is typically not the case. An optional extension of the mean primary particle size determination yields its distribution function. This method relies on the fact, that the superposition of several monodisperse signal curves gives, in contrast to monodispersial decay functions, a total signal with a non-exponential shape. In principle, any arbitrary distribution function can be recovered by employing an inverted Laplace algorithm. As this is a very time consuming issue and is very sensitive to statistical errors and noise on the signal course, a more robust method has to be chosen. One favorable way assumes the presence of any fixed distribution function, which is given by its median diameter and its distribution width. In this case the shape of the distribution function is assumed to be well characterized by these two parameters. A broadly used and well justified assumption for the distribution shape is given by a logarithmic normal one, which is typical for combustion processes (Dankers & Leipertz 2004; Liu et al. 2006).

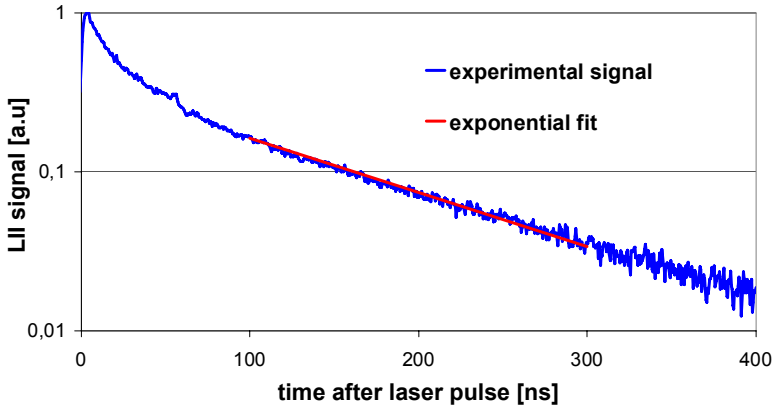

Figure 2. Experimental LII signal with exponential fit

An easy to use online determination method has been developed, that relies on the fact that the ratio of the contribution to the LII signal of different size classes changes with time after the induced laser pulse. Smaller particles cool down faster and therefore, a broad size distribution leads to a deceleration of the signal decay as the long lasting signal of bigger particles become more important at later times, what leads to a non-exponential decay. To comprise this effect a second signal decay time has to be determined by an exponential fit at later times. For comparing the experimental decay times with numerical ones, calculating of two signal decay times τ_1 and τ_2 has to be done under variation of median particle size $d_{p,med}$, distribution width σ and ambient temperature yielding the functions $d_{p,med} = f(T_0, \tau_1, \tau_2)$ *and* $\sigma = f(T_0, \tau_1, \tau_2)$. These functions can be stored in a data base, which makes an online determination in principle possible. The reconstruction of a size distribution can then be accomplished under consideration of the ambient temperature and the assumption of a lognormal function. Basically, an evaluation of the same curve under assumption of a monodisperse size distribution provides a mean particle diameter d_P, which is bigger than $d_{p,med}$, as d_p is a volume-weighted average. The reconstruction of particle size distributions from LII signals, however, is relatively sensitive and thus, needs a good signal-to-noise ratio (Dankers & Leipertz, 2004).

Another problem is the influence of the aggregate size on the signal decay, when the primary particles are packed together to dense structures, what strongly affects the particle cooling (Liu et al., 2006). In the following measurement examples this effect was not observed as the regarded particles had chainlike aggregate structures, what was validated by TEM. Thus, the primary particle sizes determined by LII showed reasonable agreement to conventional methods.

Even more flexibility of LII can optionally be achieved if it is combined with elastic light scattering, which is discussed in more detail elsewhere, see, e.g. Will et al. (1996). By this also the agglomerate size is accessible. This is, more precisely, an optical equivalence diameter of the agglomerates (radius of gyration), which correlates with the widely determined diffusion diameter of the particles. In this context it should be pointed out that elastic light scattering in contrast to LII is not selective to the carbon fraction of the particles. As also other particle components, e.g., volatiles contribute to the scattered signal, an appropriate sample conditioning is required for performing combined scattering/LII experiments.

Technical Applications

Production Process Control

Characterization of carbon black primary particle size during the production process was realized at a research plasma reactor and a furnace black production reactor (Dankers et al., 2003; Sommer et al., 2004). In **Figure 3** a typical LII measurement setup for such applications is shown, whereby here the detection of the enhanced radiation is in perpendicular direction relative to the incident laser beam.

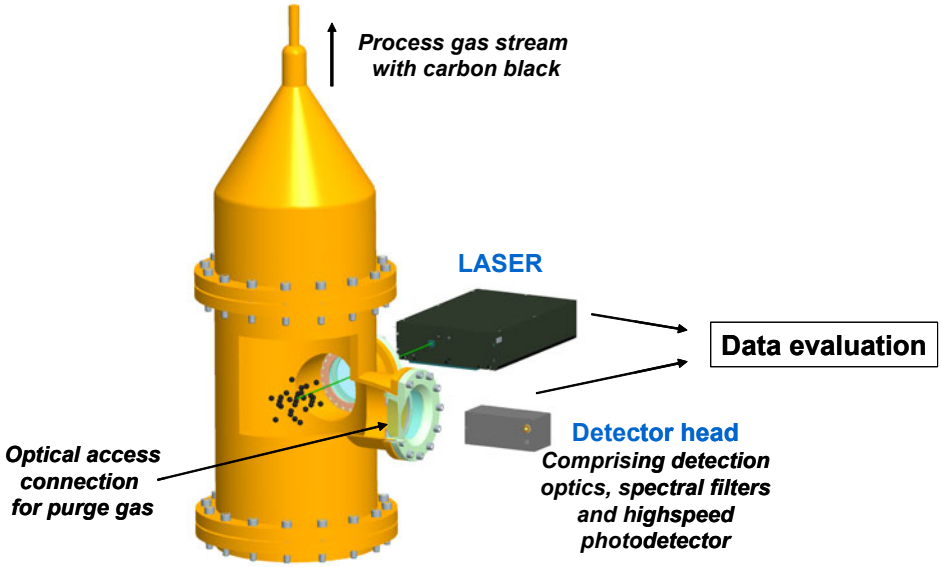

Figure 3. LII measurement setup at a carbon black reactor

Unfortunately, at most of technical reactors only one optical access is available, so LII setups in backscattering geometry have to be realized. Thereby, the carbon black particles are irradiated by a frequency-doubled Nd-YAG laser beam leading into the measurement volume by a mirror and a beam splitter, which are both highly reflective for 532 nm and transmittive for other wavelengths. The LII signal is detected within an appropriately selected spectral range by a fast photomultiplier tube (PMT), fed to an oscilloscope, processed and evaluated by a computer. In the regarded plasma reactor optical access was possible in three different heights, see **Figure 4**. To obtain an appropriate radial resolution, glass tubes were pushed into the reactor and kept free by means of purge gas. Absorption of the laser beam is so strong that the laser energy is sufficient to heat up the particles only for a short distance after entering the reactor, if the carbon black concentration is high enough. Thus, the outside edge of the LII measurement volume is determined by the end of the glass tubes, as the laser beam cannot irradiate particles until entering the reactor. However, in the course of the measurements it turned out, that the carbon black concentrations were lower than expected, and absorption of the laser beam within a short distance (<1cm) was not given. The laser irradiance was sufficient to induce incandescence over a long path within the reactor. Thus, the local allocation of the measuring volume became difficult as the signals are integrated over a certain radial distance. Furthermore, measurements with a 90°-setup were carried out in order to compare them with the results of the backscattering geometry. Thereby, the LII signal is detected through a second optical entrance perpendicular to the laser beam, which could only be realized for the lowest local position inside the plasma reactor. The temperature distribution in the reactor was measured with thermocouples, which is reasonable for the relatively low carbon black concentrations. The radial temperature gradient is steeper than the axial one which particularly holds in the upper part of the reactor.

For all examined reactor settings within the optically accessible range, temperatures do not exceed 1000°C, i.e. the particle formation is expected to be terminated. As the temperature is an important input parameter for evaluating the primary particle size, in case of the backscattering geometry the diameter can only be determined under consideration of the temperature range in the respective measurement downstream position. Nevertheless, these primary particle sizes reveal a good correlation with the results obtained in a perpendicular setup configuration and with calculated particle sizes based on the surface area from chemical analysis. Determinations in different downstream positions provided identical particle sizes for each reactor setting, which is in good agreement with the expectation that primary particle formation has already been terminated.

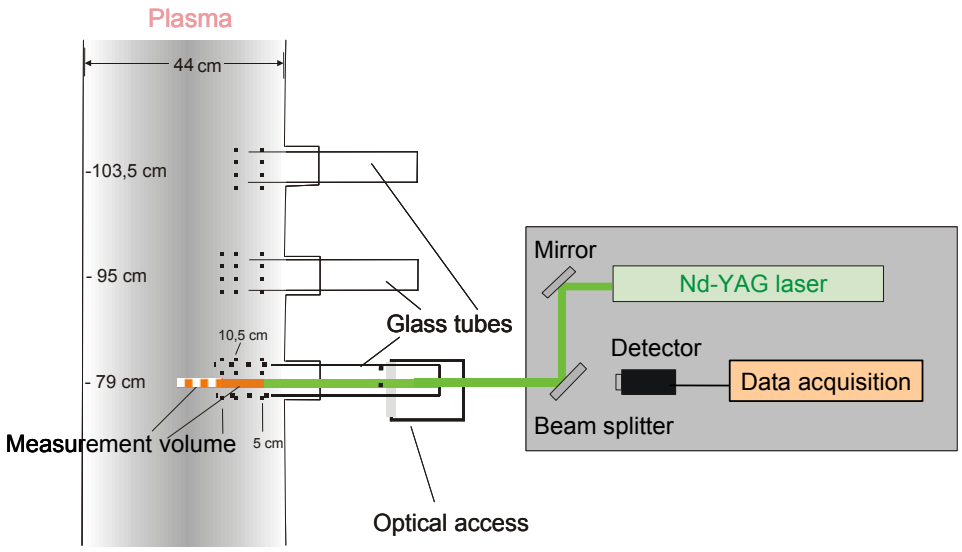

Figure 4. Backscattering configuration at a research plasma reactor (Sommer et al., 2004)

As stated above LII signals also contain information about the size distribution. To compare the influence of different plasma powers on primary particle diameters, different ways of size evaluation have been accomplished. It could be shown by assuming a monodisperse distribution, that the mean primary particle diameter is 31 nm for 30 kW and 33 nm for 70 kW. In contrast to that, under the assumption of a lognormal distribution, and by applying a second decay time, the determination yields a different result which can be seen in **Figure 5**. Size distributions with median sizes of 17 nm and 28 nm and standard deviations of 0.39 and 0.18 for 30 kW and 70 kW were observed, respectively. Unfortunately, the reconstruction of particle size distributions is relatively sensitive on the signal behavior and therefore, a good signal-to-noise ratio is required.

A similar experimental setup was used to perform online size characterization during the production process within a furnace black reactor (Dankers et al., 2003). For different operation conditions, LII measurements were carried out and compared with different chemical offline analyzing methods. For example, the CTAB (hexadecyltrimethylammonium-bromide) value is an adsorption technique, which characterizes the primary particle size without consideration of small porosities in contrast for example to the iodine number. Finally, it turned out that the obtained results correlate very well with CTAB adsorption numbers **(Figure 6),** whereas correlation with iodine numbers was not observable. This underlines the fact, that LII is a measure of the enveloping surface area.

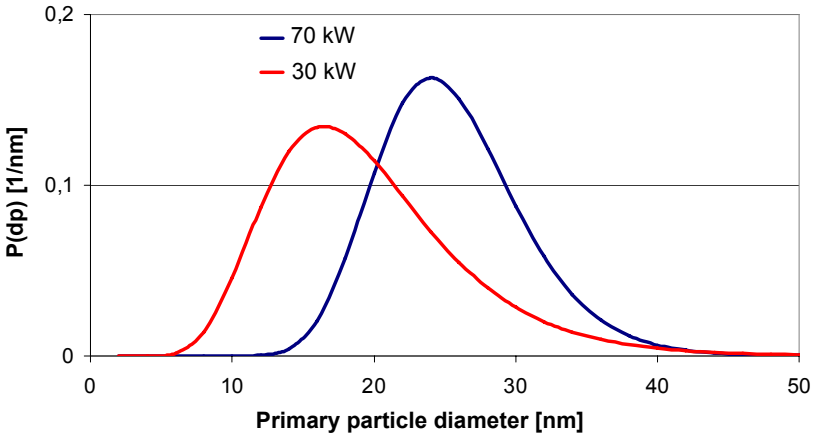

Figure 5. Primary particle size distributions for different plasma power (Sommer et al., 2004)

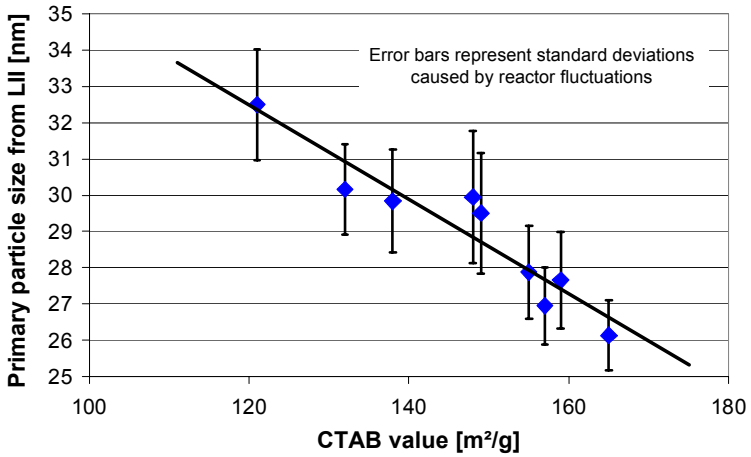

Figure 6. Correlation of primary particle size determined by LII and CTAB value (Dankers et al., 2003)

Diesel Raw Exhaust Measurements

For raw exhaust measurements in the automobile industry a compact sensor (Laser-Induced Incandescence Soot Analyzer - LI²SA) has been developed (Schraml et al., 1999). Selective particle characterization of elemental carbon (EC) is essential to appraise the performance characteristics of diesel exhaust after treatment systems and of specific engine optimization. For this, real time investigations are important in order to resolve fast particle property changes.

The system consists of a water cooled ring adapter with purge air supply (**Figure 7**) and of a control and evaluation unit. The laser beam with an excitation wavelength of 1064 nm is focused in the measurement volume by a light fibre and the enhanced thermal radiation is captured by an appropriate detector head perpendicularly. It can be applied directly in the raw exhaust without dilution up to exhaust gas peak temperatures of 700°C. Its sensitivity (1µg/m³) and variability for real-time (20Hz) soot characterizing in different applications is shown.

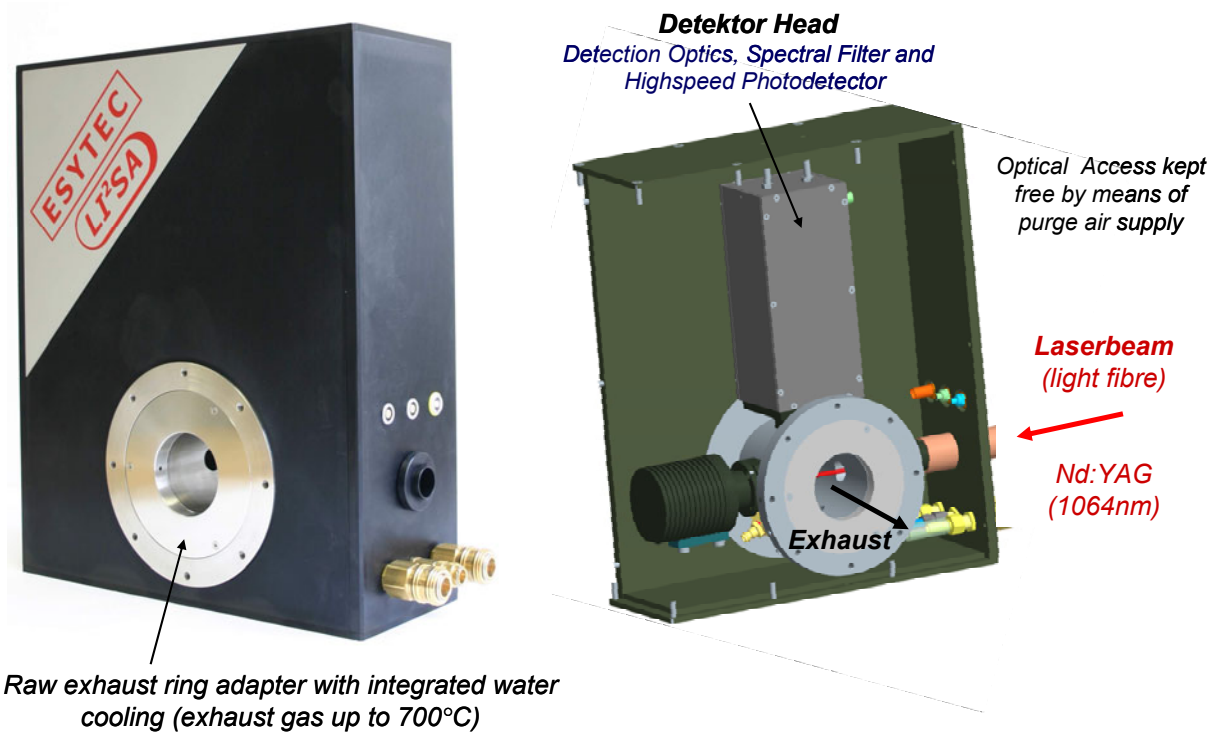

Figure 7. LI²SA soot sensor (raw exhaust ring adapter)

As already stated above, laser-induced incandescence is only sensitive to elemental carbon, particle fluctuations as ashes, soluble and volatile fractions are not determined, as they are vaporized by the high intense laser pulse. This was shown in many previous studies in comparison to conventional standard reference methods like coulometry, gravimetry or opazimetry (Schraml et al., 2004, and references therein). Exemplarily, correlation coefficients of just $R^2<0.53$ have been found for standard gravimetry. However, if the chemical composition of the particles remains fairly constant, which might be true in some cases for diesel engines, correlation coefficients increase to 0.8. A better correlation is consequentially given if the results are compared to thermogravimetric measurements. The only comparable technique, which determines only the EC content, is coulomteric filter analysis. Therefore, the mass concentration calibration for the sensor is done in comparison to coulometry at a combustion aerosol standard (CAST) (**Figure 8.**).

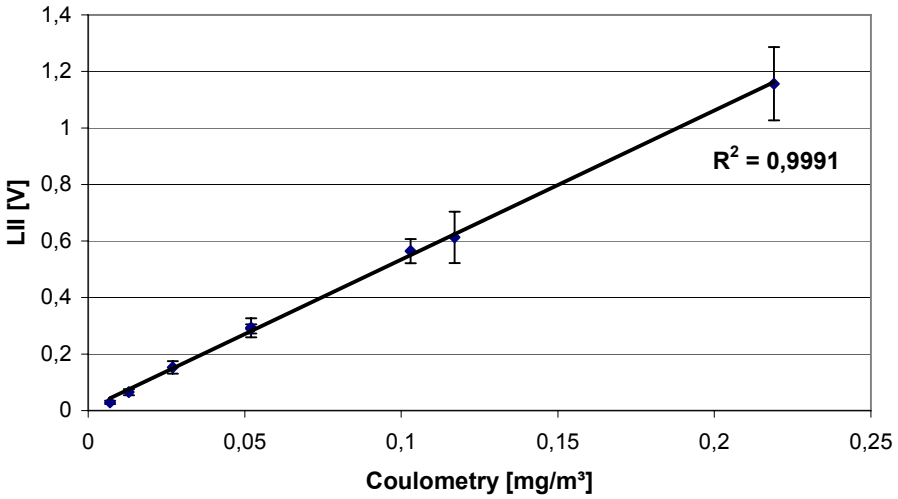

Figure 8. Correlation of the LII signal to coulometric filter analysis obtained at a CAST (Schraml et al., 2004)

The system has been applied in several configurations on a variety of test benches within different projects, which are here just briefly reported to illustrate the broad applicability and performance of the LI²SA sensor.
One possible application of the systems is the investigation of exhaust after treatment systems. Beside its high sensitivity, which is important for the characterization of diesel particle filters and fine dust monitoring, also the fast in situ measurement up to 20 Hz makes it possible to optimize filter regeneration strategies or engine combustion even under highly transient conditions. Especially, the investigation of exhaust after treatment system behavior during different test cycles is a very important task in research and development.

In **Figure 9** the temporal course of the EC mass concentration is depicted for two different measurement locations during an ESC test cycle (European Stationary Cycle) of a medium duty truck engine equipped with an SINOx© SCR catalyst together with the deduced EC mass reduction. The first measurement location was directly in front of the catalyst, whereas the second was immediately afterwards, each utilizing a full-flow sensor head. It could be observed that the EC mass reduction is significantly different for the individual operation points during this 13 stage stationary test cycle. Although the SINOx system has been designed to reduce the nitrogen oxide emission by a selective catalytic reaction after urea injection, also an EC mass reduction of about 10-30% was found. This reduction is strongly dependent on the individual operating conditions, e.g., a significant higher mass reduction occurs under high load conditions. Amazingly, no simultaneous change was observed in specific particle surface or primary particle size (not depicted), respectively. Thus, the dominating reduction process cannot be caused by a particle size reduction in this case, but in particle number. It seems to be most likely that some of the particles passing the SCR catalyst are completely oxidized, whereas other ones pass it entirely unaffected. A possible mechanism is impaction and subsequent oxidization of these particles on the catalyst surface. This example shows that by selective particle characterization of the mass concentration as well as the primary particle size conclusions can be drawn for the underlying reduction processes in such systems.

In another application a heavy duty diesel engine was equipped with a continuation regeneration trap (CRT). From these measurements it could be seen that LII is feasible to detect EC mass concentrations with high signal dynamic and high temporal resolution even at very low, near ambient emission levels. In this application, the very good reproducibility of engine operation conditions allowed a detailed investigation of the system repeatability under ultra low emission, which has been proven to be excellent (Schraml et al., 2004).

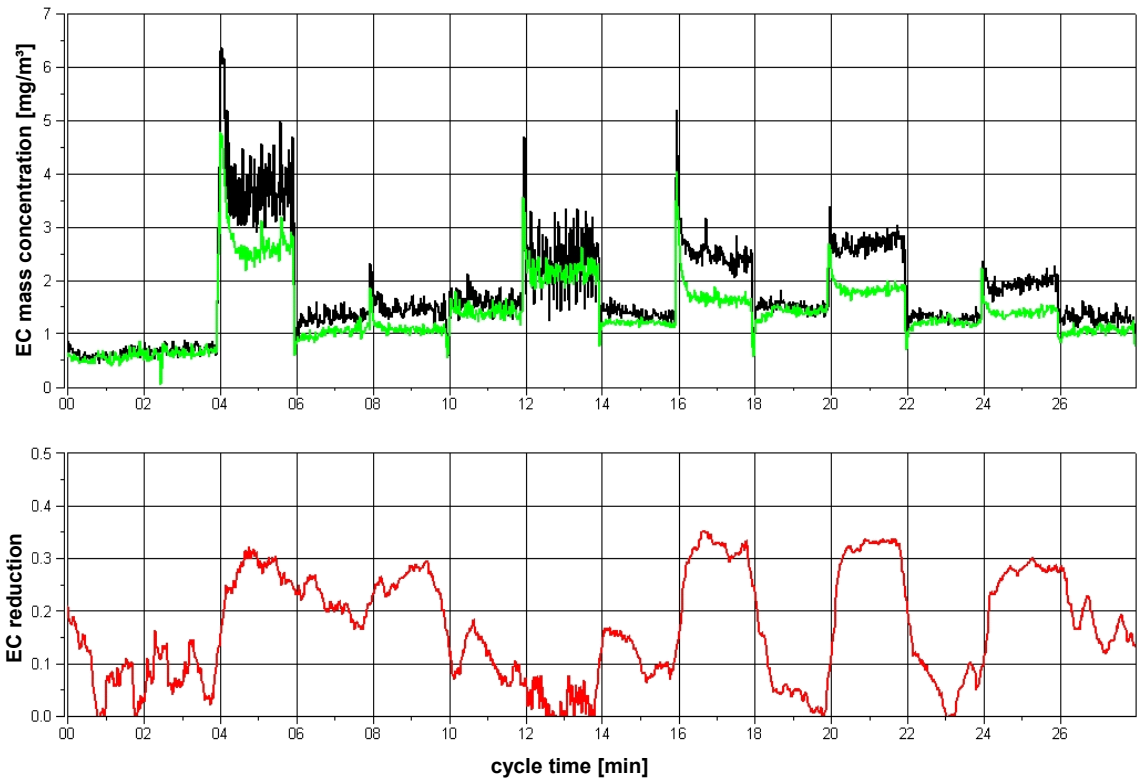

Figure 9. EC mass concentration during ESC test cycle (top) in front of (dark curve) and after (green curve) a SINOx catalyst and the resulting EC mass reduction (bottom)
(Schraml et al., 2004)

Carbon Black Particle Suspension

To this day, LII has only been applied for aerosol processes without the consideration of particles dispersed in liquids. First investigations with re-dispersed carbon blacks were carried out in order to show the principle applicability of TIRE-LII in liquids. Apart from furnace blacks (Printex A, G, 25, 35, and 55) also various gas black particles (FW 18, Colour Black S160 and S170, Printex U and U140) were suspended in different liquids. In order to break dense agglomerate cluster and to equally disperse the aggregates, the suspension was excited by ultrasonic. The stability of the suspension was recorded by measuring the aggregate size distribution (diffusion diameter) with dynamic light scattering (DLS). Moreover, this was done before and after the LII measurements in order to control the stability of the particle suspension. The LII measurements were carried out in a quartz crystal, using the same excitation and detection equipment as mentioned before.

In the course of our investigations it was possible to detect LII signals for all particle suspensions studied. First, the dependence of the maximal incandescence signal on the laser fluence is documented. The initial increase in laser fluence leads to a rapid rise in the peak LII signal, until the sublimation-limited region is reached from which no significant signal change is observable. These results are comparable to those from aerosols and flames as mentioned already before. Fluence dependency as described here was representative for all carbon blacks tested in this study (Sommer & Leipertz, 2007).

To evaluate the relation of the exponential signal decay time, which is determined 100-250 ns after the laser pulse, and the size of the primary particles, which has been measured by TEM, the laser fluence was set at 0.17 J/cm². This corresponds to the beginning of the plateau region. A linear correlation between both quantities was found, which is illustrated in **Figure 10**. FW 18 is not included here, as its signal decay was too fast. Furthermore, no influence of the aggregate size could be found in these investigations. A comparison of Printex U and Printex 55 in particular, which have the same TEM primary particle diameter, confirms this observation. The LII signal decay time was almost the same for both, showing that for the considered carbon black suspensions the exponential decay time is only a measure of the primary particle size.

In this context, the influence of different solvents on the signal decay behavior was also considered. Despite different thermal conductivities, the signal decay time was not influenced by the solvent in question. This factor and the signal relation to the laser fluence can possibly be described by vapor layers enclosing the particles, which is described in more detail elsewhere (Sommer & Leipertz, 2007).

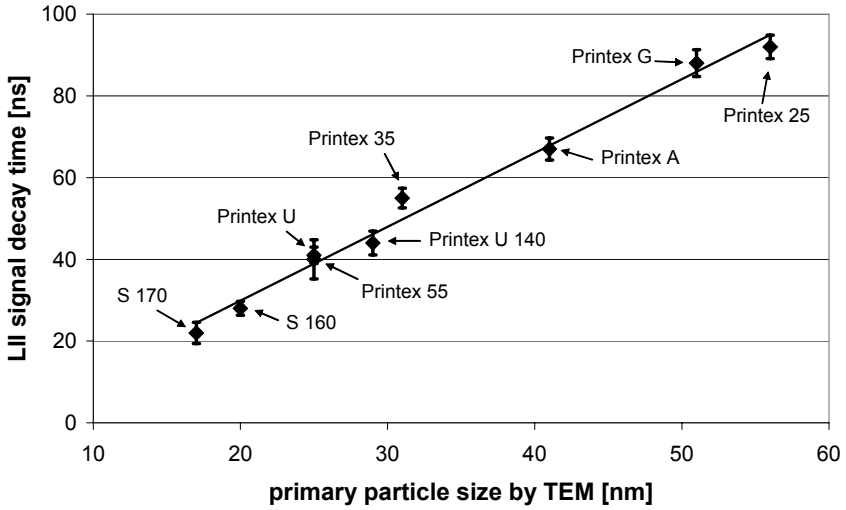

Figure 10. Correlation of LII signal decay time and primary particles size measured by TEM for different carbon blacks dispersed in Ethanol (Sommer & Leipertz, 2007)

Conclusion

In this paper the features of time-resolved laser-induced incandescence are shown in two important technical application fields. The technique has been successfully applied both at a carbon black reactor and directly in Diesel raw exhaust. An online determination of the mean primary particle size of carbon blacks during the particle formation has been carried out and compared to standard chemical analysis indicating very good agreement. Furthermore, primary particle size distributions for different reactor settings were reconstructed by the determination of two signal decay times from the experimental LII signals in comparisons to numerically calculated ones.

For automobile investigations one of the main benefits of LII, in comparison to conventional particle measurement systems, lies in the exceptional high sensitivity and the possibility of a simultaneous evaluation of EC mass concentration and primary particle size. This feature enables a convenient investigation of several interesting effects, which up to now, have not been observed in detail – either because of the lack of possibility or because of the complexity of alternative solutions. Furthermore, the high dynamic range and temporal resolution qualifies LII as an ideal tool for research and engine development as it enables the user to optimize the engine with respect to soot emission even for transient operation conditions. This is particularly important as time consuming data evaluation is not necessary with this system and so extensive parameter studies can be performed very fast. Therefore, LII is able to comply with all forthcoming developments of new combustion concepts, traps or other exhaust gas after treatment systems. Furthermore, with this measurement technique it is possible to determine very low soot concentration also in ambient air (Sommer et al., 2005).

For the first time, time-resolved laser-induced incandescence has been successfully applied for the characterization of suspended nanoparticles. Re-dispersed carbon blacks were investigated in different solvents, whereby a linear correlation between the exponential LII signal decay time and the primary particle size determined by transmission-electron microscopy was found. Moreover, the signal decay time was thereby influenced neither by the aggregate size distribution, nor by the used solvents. Further studies will focus on the understanding and theoretical modeling of the heat transfer mechanisms under these new measurement conditions.

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