Soot measurements in laminar flames of gaseous and (prevaporized) liquid fuels

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Abstract
A high pressure vessel and diffusion burner (HPVB) were designed, constructed and integrated with an evaporation system, to enable laminar flames studies of gaseous and vaporized liquid fuels. This experimental setup is designed to offer ample optical accessibility for laser diagnostics techniques; its capabilities and specifications are described. Soot volume fraction measurements are performed with line-of-sight attenuation (LOSA) and validated using data of ethylene flames from literature. The first measurements of a vaporized liquid fuel (n-heptane) are reported. A stable laminar n-heptane flame is achieved with a maximum standard deviation in soot volume fraction of 0.04 ppm.

Introduction
In the present work, it is attempted to build a bridge between the most simple combustion systems that have been studied in literature (laminar atmospheric burners, in which mostly simple, gaseous fuels are burnt) and the much more complex environment that is found in practical combustion engines. This is accomplished by the design and construction of a high pressure vessel and diffusion (co-flow) burner, and integrating this with a commercial evaporation system CEM (Controlled Evaporator Mixer), making it possible to study the combustion at elevated pressures of automotive (bio-)fuels, which are typically liquid at room temperature.

A set of gaseous and liquid fuels is selected for soot volume fraction studies, aiming to get new insights into the effects of fuel structure on soot formation. These experiments will also serve the purpose of integrating, debugging and validating the experimental setup, including its use at high pressures (up to 3 MPa). The first experimental approach comprises a characterization of different fuels in terms of soot formation tendency, using the line-of-sight attenuation (LOSA) technique.

Future investigations will include an investigation of the pressure effect on soot formation and soot formation/reduction tendency, using different oxygenated fuels blended with more conventional fuels. Further detailed studies will be carried out using advanced laser diagnostic techniques that will be assessed and/or developed for use in a high pressure and sooting environment.

Specific objectives
In this paper, the design of the high pressure burner setup, its capabilities and specification are described. The soot volume fraction measurements performed with LOSA are validated using ethylene flame data from literature. For first time, according to the author's knowledge, soot volume fraction measurements of a vaporized liquid fuel (n-heptane) performed in a laminar co-flow diffusion burner are presented.

Experimental methods

Pressure vessel, burner and evaporation system

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The burner dimensions are based on the Long and Smooke burner [1] and its design is adapted to take into account the fit of the burner inside the pressure vessel and the use of the burner at elevated pressures. It is a coflow diffusion burner with a fuel tube inner diameter of 4 mm, an oxidizer coflow inner diameter of 50 mm and a height of 100mm (see figure 2). The fuel nozzle is tapered to reduce recirculation from the burner tip and improves stability of the fluid–ambient interface. Aluminum beads, perforated plates and sintered metal are used as flow straighteners.


The high pressure vessel and burner are integrated with a commercial (Bronkhorst) evaporation system called CEM (Controlled Evaporator Mixer). In this unit, a liquid (fuel) and carrier gas are accurately mixed and then heated. The heating part consists of a heated spiral where the liquid droplets and carrier gas flow through. When the temperature of the heated block is sufficient, the droplets will vaporize. This process results in a stable and accurately controllable fuel vapor/carrier gas stream. To avoid condensation of the fuel vapor on its way to the burner, an electrically heated tube is used to connect the CEM to the burner. The burner is also heated by an electrically heated ring.

**High pressure soot diagnostics**

Soot formation has been studied in the literature using both physical probing of the flame and non-intrusive optical techniques. Among the non-intrusive optical techniques, soot volume fraction has been investigated using a variety of diagnostics including line-of-sight attenuation by soot particles (LOSA), also known as extinction, and laser induced incandescence (LII).

The LII technique allows measurements of a 2D soot volume fraction field and it is able to measure very low soot concentrations. However, in order to give quantitative results, LII must be calibrated using the LOSA technique and it has some drawbacks when used at elevated pressures: soot volume fractions at high pressure are much higher than at atmospheric conditions and signal trapping can occur when the LII signal is attenuated on its way to the camera by soot particles in the flame. Choi and Jensen [2] claim to correct for signal trapping in their measurements, however Liu and co-workers [3] strongly disagree with their method. Liu claims that correction of signal trapping in general is difficult, if not impossible, since it requires knowledge of the distributions of soot volume fraction and the morphology of soot particles.

This stated, and with high pressure application in mind, the LOSA technique was chosen for this work. LOSA is a relatively simple and straightforward diagnostic technique for quantification of soot volume fraction in flames. Its principle is based on the extinction of light by a medium (see figure 3). It offers high spatial resolution and does not rely extensively on prior knowledge of physical constants or morphological properties. Several researchers have obtained two dimensional spatially resolved soot volume fractions using LOSA in combination with inverse Abel transformation [4, 5].

Figure 3. Scheme of an extinction measurement.

Extinction consists of scattering and absorption. Assuming the soot particles small enough to be in the Rayleigh regime given by

\[
\frac{\pi d}{\lambda} \leq 0.3, \tag{1}
\]

where \(\lambda\) is the wavelength of the light and \(d\) is the particle diameter, scattering can be neglected and absorption is equal to extinction [6]. The extinction coefficient \(K_{\text{ext}}\) can be calculated by measuring the transmitted light intensity \(I_T\), the incident light intensity \(I_0\) and the absorption length \(L\) in the flame:

\[
K_{\text{ext}} = \frac{1}{L} \ln \frac{I_0}{I_T}. \tag{2}
\]

Assuming the soot particles to be spherical, the soot volume fraction can now be calculated:
In Eq. (3) $K_{e}$ is the dimensionless optical extinction coefficient. The calculation of $K_{e}$ demands knowledge of the soot refractive index ($m$) and scattering to absorption ratio ($\alpha_{sa}$). The latter is neglected in the Rayleigh regime. Recently reported values of $K_{e}$ are in the range of 8 to 10 at a wavelength of 632.8nm [7, 8].

The logarithm of the incident laser intensity divided by the transmitted laser intensity determines the line-of-sight integral projection data:

$$P(y) = \ln \left( \frac{I_0}{I_T} \right)$$

In order to convert line-of-sight integral projection data of an axisymmetric measurement field into a spatially resolved distribution $F(r)$, in this case spatially resolved $K_{ext}$ and subsequently soot volume fraction, inverse Abel transformation is applied:

$$F(r) = -\frac{1}{\pi} \int_{y=r}^{y=y_{max}} \frac{P'(y)}{\sqrt{y^2 - r^2}} \, dy$$

Numerically, the integral is replaced by a summation with segments between points $y_i$ and $P'$ is considered to be constant between these points. This leads to

$$F(r) = \sum_{y_i=r}^{y=y_{max}} -\frac{1}{\pi} P'(y) \int_{y=y_i}^{y=y_{max}} \frac{dy}{\sqrt{y^2 - r^2}}$$

in which the integral can be evaluated analytically, and the derivative $P'(y)$ is obtained from the measurements.

**Optical setup**

The LOSA layout and equipment used in this work are shown in figure 4.

The LOSA setup is designed to minimize all possible disturbances. The laser used for this research is a HeNe laser, with a power of 5 mW and a wavelength of 632 nm. To compensate for laser intensity variation, a reference detector is used. Making use of a wedge shaped beam splitter; approximately 4% of the laser intensity is split to the reference detector. The incident light intensity is determined from the measurement without flame and the reference signals. To filter out background light in the reference detector, a tube with a diaphragm is placed in front of it. To prevent errors due to non-uniformity of the reference detector a negative lens is placed behind the diaphragm to diverge the beam to the size of the detector and a diffuser is placed just before the detector to make the signal completely uniform. The mirrors used in the setup are first layer mirrors to prevent multiple reflections. A laser scan is created by an adjustable mirror and two positive anti-reflection coated lenses. The angle of the adjustable mirror can be changed with an electric step-motor; the length of the scan is approximately 30 mm. The aperture right behind the first lens will block the laser beam when it is far away from the flame and transmits the beam when it is close to or in the flame. The laser beam with a diameter of 0.8 mm is focused in the flame where its diameter is approximately 100 µm. The diameter of the second lens is 50 mm and it is large enough to compensate for beam steering. An integrating sphere is used to create a uniform light beam for the interference filter and the attenuation detector. The entrance of the integrating sphere is placed behind the second lens at its focal point to make sure that the total light from the laser beam will always be captured by the integrating sphere. The interference filter with a wavelength of 632 nm and a FWHM of 3 nm, filters out background light. The HPVB is placed slightly non-perpendicularly (~15°) to the laser beam, to prevent reflections in the same direction as the main laser beam.

**Measurement procedure**

The first measurements of soot volume fraction performed with this experimental setup are carried out in ethylene and n-heptane laminar diffusion flames at atmospheric pressure. In order to validate the LOSA measurements on the HPVB, results of soot volume fraction measured in laminar diffusion ethylene flames are compared with available data from Smooke and co-workers [9] using the same dimensionless extinction coefficient ($K_{e}=10$). The oxidizer is air and the fuel is a mixture containing varying ratios of ethylene and nitrogen. Two flames are selected and named according to the ethylene/nitrogen fractions: 60% ethylene/40% nitrogen flame and 80% ethylene/20% nitrogen flame.
To generate laminar n-heptane diffusion flames, the n-heptane is vaporized in the CEM using nitrogen as a carrier gas and air is used as a coflow oxidizer. Two flames are generated: flame 1 has an n-heptane mass flow of 4 g/h, nitrogen flow of 0.052 Ln/min and coflow air of 30 Ln/min; flame 2 has an n-heptane mass flow of 4 g/h, nitrogen flow of 0.070 Ln/min and coflow air of 25 Ln/min. One set of measurements consists of an average of six unique measurements (a radial scan in the flame). Three sets of measurements are carried out at different days to evaluate the precision.

The soot volume fractions in these flames were measured at different flame heights. The flame height zero (the flame base) is accurately determined by changing the burner height until the laser beam is attenuated by the fuel tube tip. The scan-velocity of the laser beam is measured by scanning the beam over a known distance and analyzing the signal on the oscilloscope. This velocity is used to determine the radial position of the laser in the flame. The middle of the flame is determined by assuming the flame to be axially symmetric. To examine whether this assumption is valid, the attenuation signal of both sides of the flame is compared with every measurement. The laser scans the flame from right to left with a speed of 4.63 mm/s and two signals are collected at the same time: one by the attenuation detector, providing the main signal with flame (I_F), and another by the reference detector, the reference signal (I_F_ref). The data are read out with a digital oscilloscope and from the oscilloscope the data are transferred to the computer and interpreted there. The same scan is made without a flame, collecting I_NF and I_NF_ref ('no flame'). These data are also transferred to the computer; the two scans together form one measurement. After the measurement the laser is blocked, and the natural flame emission I_E is measured with the attenuation detector. This value is subtracted from the main measurement with flame (I_F) to determine the transmitted laser intensity I_T:

\[ I_T = I_F - I_E \]  

(6)

The incident intensity (I_0) is determined using the measurement without a flame and the reference signals according to equation 7. This is done to take into account possible variations in laser intensity during the measurements.

\[ I_0 = I_{NF} \left( \frac{I_{F_ref}}{I_{NF_ref}} \right) \]  

(7)

Results and Discussion

Ethylene flames

In the subsequent figures we plot the radial profile of soot volume fraction measured in two ethylene flames along with the data provided by Smooke and co-workers. The ethylene flames generated in this work show differences in height of 5% when compared to those of Smooke. It is likely that these differences in flame heights are caused by the differences in accuracy of the mass flow controllers used in both setups. Due to this fact, the flames were compared at three different vertical positions: 30%, 50% and 70% of the respective flame heights. The flames measured in this work have 60% ethylene/40% nitrogen and 80% ethylene/20% nitrogen in the fuel tube. Figures 5 to 7 show the results obtained with the flame 60% ethylene/40% nitrogen.
The flame measured in this work showed very good stability with maximum standard deviations of the soot volume fractions of 0.06 ppm. Peak soot volume fractions are located on the flame wings and considering the heights measured, the peak value of 1.2 ppm appears at 50% of the flame height. The comparison with Smooke data shows very good agreement in the absolute soot volume fractions values and in the location of the maxima.

Figures 8 to 10 show the results obtained with the flame 80% ethylene/20% nitrogen. This flame also showed good stability, although it was less stable than the flame 60% ethylene/40% nitrogen, with maximum standard deviations of soot volume fractions being 0.08 ppm. Peak soot volume fractions also appear on the flame wings with a peak value of 2.5 ppm at 50% of the flame height. The comparison with Smooke data also shows good agreement in the curve profile and location of the maxima. However, the differences in the absolute soot volume fractions values are larger when comparing with the flame 60% ethylene/40% nitrogen.

\textbf{n-Heptane flames}

Figure 11 shows the first measurements of soot volume fraction in a vaporized liquid fuel flame performed with this experimental setup. The n-heptane flow is 4 g/h, nitrogen (carrier gas) flow is 0.052 Ln/min and the coflow air is 30 Ln/min. The measurements were carried out in five different days at 90% of the flame height.

The results show soot volume fractions in the center line (flame axis) varying from 1.3 ppm to 1.5 ppm and a large deviation of one measurement (set 4) near the flame edge. It can be seen that it was not possible to achieve the same repeatability as for ethylene with this flame, which has a maximum standard deviation of 0.17 ppm within the flame area.

Efforts to improve the flame stability were made changing the carrier gas and oxidizer flows. A more stable flame was visually observed when using an n-heptane flow of 4 g/h, nitrogen and air flows of 0.070 Ln/min and 25 Ln/min, respectively. The improved flame stability was reflected in the soot volume fractions measurements as can be seen in figure 12. The measurements were carried out in three different days also at 90% of the flame height.
Figure 12. 4 g/h heptane/0.070 Ln /min nitrogen flame soot volume fraction at 90% of the flame height.

As expected, the flame with 0.070 Ln /min of nitrogen has lower soot volume fractions when compared with the flame with 0.052 Ln /min of nitrogen due to the dilution effect. The maximum standard deviation of this flame was 0.04 ppm within the flame area.

As in the case of ethylene flames, the wiggles observed in these two n-heptane flames can be caused by the fact that the line-of-sight integral projection data is smoothed less than in the center of the flame in these areas, as a result of the variable smoothing method used. Efforts are going on to improve this.

Conclusions

An experimental setup capable to burn gaseous and vaporized liquid fuels in laminar diffusion flames was constructed. The high pressure vessel and burner were designed to work at elevated pressures and to offer ample optical accessibility to apply laser diagnostic techniques. The first experiments with this setup were performed in ethylene and n-heptane flames at atmospheric pressure. The flames show good overall stability. Spatially resolved soot volume fractions were measured with line-of-sight-attenuation (LOSA) in combination with inverse Abel transformation. The measurements were validated with data from Smooke and co-workers in ethylene diffusion flames. Overall correspondence was observed, both in peak values and location of maxima in soot volume fractions.

Soot volume fraction measurements were also performed in a vaporized liquid fuel (n-heptane) flame. The flame dynamics was clearly affected by the carrier gas and oxidizer flows. A reasonable stable flame was achieved with the flame of 4.0 g/h heptane/0.070 Ln /min nitrogen and 25 Ln/min coflow air. The results suggest that although it is possible to create a stable flame from a vaporized liquid fuel, more studies under different operational conditions should be performed with this setup to better evaluate the flame stability.

This experimental setup enables a variety of studies on liquid fuels at elevated pressures. It is also a useful tool for investigation of advanced laser diagnostics techniques in high pressure environments, allowing for improvements and new developments.

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