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Measurements of sooting tendency in laminar diffusion flames of n-heptane at elevated pressure

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ABSTRACT

This research focuses on the effects of an increasing pressure on the soot formation during combustion of vaporized liquid fuel. Therefore soot formation is measured in a laminar diffusion flame, with n-heptane as fuel, over a range of pressures from 1.0 to 3.0 bar. The soot volume fraction in the diffusion flames has been measured using Laser-Induced Incandescence (LII) calibrated by means of the Line Of Sight Attenuation (LOSA) technique. The values of the calibration factors between LII intensities and soot volume fraction from LOSA are slightly varied for different pressure. The integral soot volume fractions show power law dependence on pressures, being proportional to p^n, with n being 3.4 ± 0.3 in the pressure range of 1.0–3.0 bar.

1. Introduction

Soot emissions from combustion of hydrocarbons have long been recognized as a significant problem to the environment and to the health of humans and other animals.

The understanding of the fundamental mechanism of soot formation has grown considerably by studies on laminar flames. Most literature has reported on soot formation in laminar flames at atmospheric pressure. It is well known that increased pressure has a large influence on the soot production in spray combustion, in premixed as well as diffusion flames [1–3]. Fuel pyrolysis and soot nucleation are enhanced by pressure, and the net effect is that soot growth and concentration are strongly affected by the system pressure [4,5].

Gülder's group at the University of Toronto has been focusing on soot formation research in laminar flame at elevated pressure for many years [6–10]. The soot formation which was defined as the mass flow of soot at a given flame height per unit of carbon mass flow in the fuel, in co-flow methane–air and ethane–air laminar diffusion flames at elevated pressures was measured at different fuel flow rates. Moreover, they reported on the exponential relationship between soot volume fraction and pressure or the fraction of fuel carbon converted soot. Their measurement methods mainly involve Spectral Soot Emission (SSE) and Line-Of-Sight Attenuation (LOSA), to provide information on spatially resolved soot volume fraction as well as temperature, for various fuels (methane, ethane, ethylene and propane), and pressure increasing from atmospheric to 60 bar. In a recent study [11], the authors summarized that the available high-pressure soot yield data from gaseous fuel diffusion flames behave in a unified way on reduced pressure when the soot yield is properly scaled. However, these studies have definitely addressed the issue of soot formation by gaseous fuels only. For liquid fuels, there is a clear scarcity of data.

Roberts' group at North Carolina State University also conducted a comprehensive investigation of soot formation in laminar flames [12]. Their results are expressed in terms of the Smoke Point Height by measuring the height of a laminar diffusion flame prior to emitting smoke, and in terms of soot volume fraction obtained by Laser Induced Incandescence (LII) with a calibration accomplished by LOSA. Similar to Gülder, only gaseous fuels, such as ethylene and methane, have been studied in their laminar flames over an ambient pressure range from 1 to 25 bar.

Smooke et al. at Yale University also investigated the soot formation in ethylene laminar diffusion flames [13]. The main purpose of their work was to provide quantitative and detailed data for the validation of numerical simulations. Therefore, the flame they studied was relatively simple (ethylene flame under atmospheric conditions), and the measurement method, a combination of LII and LOSA, was quantitative.

Recently, a new study on soot measurements in laminar diffusion flames of liquid fuels has been presented by Menon from Pennsylvania State University [14,15]. An experimental study on the effects of the addition of m-xylene to a nitrogen–diluted ethylene flame was presented [14]. A special setup for vaporizing liquid m-xylene introduced as an additive into a flow of gaseous fuels has

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been designed and used in this research. It was used to administer well defined quantities of m-xylene to the flame. Stable flames doped with m-xylene at low concentration levels were established at pressures of up to 5 bar. Although it is the first study of quantitative soot volume fraction measurements reporting on a liquid fuel (m-xylene) diffusion flame at high pressure, this research is limited to very small quantities of liquid fuel dopant (at most 5.0%) in a gaseous fuel flow.

Actually, there is no data on the sooting behavior of liquid fuel laminar diffusion flames at elevated pressures [16]. However, many practical combustion devices, such as internal combustion engines and gas turbines, operate on liquid fuels at high pressures. In our research, the soot formation in laminar diffusion flames of pure n-heptane up to 3 bar will be discussed.

One of the main challenges for measurements on flames in a closed chamber is the instability of the laminar flame itself. Laminar co-flow flames realized in experimental setups are required to be stable for a period long enough to apply the measurement techniques proposed. This challenge is greatly increased when working at elevated pressures. Several problems were faced by Thomson and co-workers [4,6] when stabilizing methane–air diffusion flames. Darabkhani provides extensive discussions on the stability of laminar diffusion flames [17,18]. Experiments were conducted in a high-pressure burner to investigate the influence of pressure (1–16 bar), fuel type (ethylene, methane and propane) and fuel flow rate on the shape and buoyancy induced instabilities in sooty co-flow diffusion flames [18]. The results show that the shape of the flame changes dramatically with increasing pressure, and the instability behavior of the flame depends on both fuel type and pressure. Again, all of this research was conducted on flames with gaseous fuels.

Unfortunately, as compared to flames of gaseous fuels, the instability of laminar diffusion flames of liquid fuels turns out to be more pronounced, especially at elevated pressures. That is probably the main reason why data on the sooting behavior of liquid fuels in laminar diffusion flames at elevated pressure are not available [16]. Apart from the factors governing the instability of the flames of gaseous fuels [4,6,17,18], some obstacles particular to laminar flames of liquid fuels make the realization of stable flames particularly difficult. In fact, it is difficult to produce stable flames of liquid fuels even at atmospheric conditions, mainly because the homogeneity of the temperature in the whole system is not easy to maintain during the measurement process, which in turn could bring about incomplete evaporation of the liquid fuel or condensation of the pre-vaporized liquids in the fuel line towards the burner.

Consequently, a specially designed burner and fueling system are proposed in this paper, which significantly improve the homogeneity of the temperature of the whole fuel line of pre-vaporized liquid fuels. The stability range of the flames is extended up to 3 bar.

It is important to develop diagnostic methods for soot formation measurements which can work under elevated pressure. Generally, as already mentioned in the previous part, two main methods have been used for soot detection: a luminescence method, called Laser-Induced Incandescence (LII) and an extinction method, called Line Of Sight Attenuation (LOSA).

LOSA is a widely used diagnostic method for the quantification of soot volume fractions in flames. Its principle is straightforward. The optical transmissivity of the flame of interest is determined from the difference in intensity of a narrow laser beam that has or has not passed through the flame. This yields the total extinction coefficient, integrated along the whole path of the laser beam through the flame. Typically, this "line of sight" corresponds to a tangential path, at a fixed height above the burner exit (HAB = Height Above Burner). From a series of measurements at various lateral positions at the same HAB, the radial soot distribution can be reconstructed. By also varying the HAB, the whole 3-D soot volume fraction field can be derived. Performed in this way, this method relies on several assumptions. The flame must be stable during the whole measurement series, and it must be axis-symmetric in order to reconstruct the radial distributions at fixed HAB (this involves an inverse Abel transform). Finally, the method requires an independent calibration or knowledge on the optical properties of the soot responsible for the attenuation, and it assumes that soot is really the only cause of attenuation.

Laser-Induced Incandescence (LII) is an imaging diagnostic technique, that is based on recording the grey-body radiation of soot that is instantaneously heated to far above the local flame temperature by means of a short laser pulse. Comprehensive overviews of the technique and the physics involved have been given by Schulz et al. [19] and by Michelsen [20]. The incandescence yield is nearly, but not quite, proportional to the local soot volume fraction [21]. This technique provides the advantage of instantaneous 2D measurement, but requires a calibration (often obtained by LOSA) to yield absolute soot volume fractions [22].

Compared to LOSA, the most important advantage of LII is that it is capable of providing 2D images of the soot distribution directly. However, there are a few limitations. According to research by Bassi et al. [23], at high pressure the soot particle density may be so high that the infrared radiation is not able to penetrate the flame. Incandescence trapping by flame regions in between the probe volume and the detector is a concern in highly sooting flames and may cause an underestimation of the actual soot volume fraction [24]. Shaddix and Smyth [25] claims that signal trapping becomes significant at soot volume fractions above 10 ppm. Liu et al. [26] used the uncorrected LII intensities and the particle temperatures derived from wavelength dependent LII intensity ratios and argued that the soot volume fraction inferred from the absolute LII intensity technique is higher than the true value, especially when the detection location is on the flame centerline and the soot loading is high. Thus, he claims that a correction for signal trapping in general is difficult, if not impossible, since it requires knowledge not only on the distribution of the soot volume fraction, but also on the morphology of soot particles.

Accordingly, at present there is no broadly accepted model to extract soot volume fractions directly from LII data. Consequently, some researchers prefer a qualitative comparison by comparing relative LII signal intensities [27]. Alternatively, in order to take advantage of LII for 2D soot measurements, as a compromise, some investigations have been accomplished by a combination of LII and LOSA [12–15,23,25,28]. According to this method, one flame will be measured using both methods in otherwise completely similar configurations, and the soot volume fraction of the flame is calculated from the LOSA signals. By means of this process, a calibration factor between LII intensity and LOSA soot volume fraction will be established. As a result, the LII intensity can subsequently be interpreted in terms of the soot volume fraction.

In LOSA, the soot particles are considered to be in the early stage of soot formation and small enough to warrant the Rayleigh assumption [4]. The soot volume fraction f_v can be calculated from [4]:

$$f_v = \frac{\lambda}{k_e} K_{ext} \tag{1}$$

where λ is the wavelength of the laser, K_{ext} is experimental extinction coefficient and k_e is the dimensionless extinction coefficient. K_{ext} can be calculated by measuring the transmitted light intensity I_T, the incident light intensity I_0 and the affective absorption length L in the flame: