LASER-INDUCED INCANDESCENCE STUDY OF THE INFLUENCE OF NITROGEN DILUTION ON SOOT FORMATION IN LAMINAR FLAMES

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Introduction

Along with molecular species such as carbon monoxide and unburned hydrocarbons, soot is an important product that is typically formed from incomplete combustion of hydrocarbons. "Soot" is a general term for particulate matter based on carbonaceous nanoparticles. In principle, there are applications where soot formation is wanted, e.g., in the production of carbon black or when heat should be carried away from the combustion process by thermal radiation. In other applications the formation of soot should be avoided as it is a pollutant, e.g., in automotive and industrial exhaust gases.

For pursuing both strategies, i.e., either the controlled production or the prevention of soot, in a rational way a detailed understanding of soot formation, growth and oxidation in combustion environments is essential. Although we have a general idea about how soot is formed including the generation and growth of polycyclic aromatic hydrocarbons PAH, the fundamental processes and how they are influenced by thermodynamic parameters and the chemical environment are still object of research and represent a major challenge for combustion engineers and scientists.

The fuel, or rather the fuel composition, and how it is mixed with the oxidizer plays a key role, e.g., non-premixed flames are often mentioned to be sooty [1]. Moreover, fuel additives like inert gases can influence the sooting behaviour significantly. Various experiments have been conducted in order to study the influence of transport properties on the soot formation mechanism using laminar diffusion flames adding inert gas like Helium and Argon or further fuels [2-5]. The results showed that three possible phenomena cause the reduction of soot concentration: (1) the effect of mixture dilution between gas and fuel results in changing the amount of carbon per unit of weight; (2) a thermo-effect due to a change in flame temperature and (3) a soot reduction through the chemical interaction caused by altered combustion boundary conditions.

Concerning soot diagnostics laser-induced incandescence (LII) is an established tool capable of determining soot concentration and particle size in principle in real time [6]. Therefore it is well suited for the investigation of the formation and destruction processes of soot particles inside flames.

In the present work we investigate the influence of adding an inert gas (nitrogen) to the fuel supply of a non-premixed propane flame. In order to study its impact on the local soot concentration and particle size, point-wise LII measurements are performed at different flame positions in dependence on the nitrogen dilution.

LII in a Nutshell

The principle of LII is based on the absorption of radiation of a short laser pulse by the carbonaceous particles. Promptly the particles are heated up close to their evaporation temperature. After the laser pulse, they cool down due to thermal radiation and heat conduction. While the latter is the dominating heat loss mechanism, the radiation represents the signal which is recorded time-resolved using a photomultiplier tube. Since small particles cool down faster than large ones owing to a higher ratio of surface area and volume, the temporal signal decay contains information about the particle size distribution. Moreover, the maximum signal intensity yields an estimate for the particle mass concentration. For quantification purposes, the energy balance for particle heating and cooling has to be set up and solved numerically. In the final step, the calculated data are compared to the experimental signal decays. A detailed description along with analyses and modeling of all involved heat and mass transfer mechanisms can be found in the literature, see e.g. [6,7].

Experiment

Atmospheric pressure laminar propane diffusion flames were stabilized on a Bunsen burner with a diameter of 12mm. A co-axial tube of 51mm diameter provided a stabilizing symmetric co-flow of air. The gas flow rates were controlled by mass flow controllers. The propane and air flow rates were kept constant at 0.09kg/h and 1.87kg/h, respectively. The nitrogen mass flow rate was stepwise varied from 0 to 0.047kg/h. All flow rates given are related to the standard conditions 0°C and 1013kPa. The N_2 was premixed with the C_3H_8 after the mass flow controllers. The resulting visible flame height was approximately 60mm.

The LII experiment was based on a pulsed, frequency-doubled Nd:YAG laser (532nm) with 9ns pulse duration. The beam crossed the flame centre axis of the burner. As the burner was mounted to a vertical translation stage the measurement position could be located at different heights. The measurement height above the burner exit was varied from 10 to 40mm in steps of 10mm. The mean power density of the laser was 10^8W/cm^2. The LII signal was collected in direction perpendicular to the laser pathway using a fused-silica lens, detected by a photomultiplier tube and recorded by a digital oscilloscope.

Results and Discussion

Measurements were carried out at four different positions ranging from 10 to 40mm height above the burner, where the flame instabilities were negligible. The detected time-resolved LII signals recorded at different heights in the undiluted flame are shown in Fig. 1. The signals were normalized by dividing the entire signal trace by its max value.

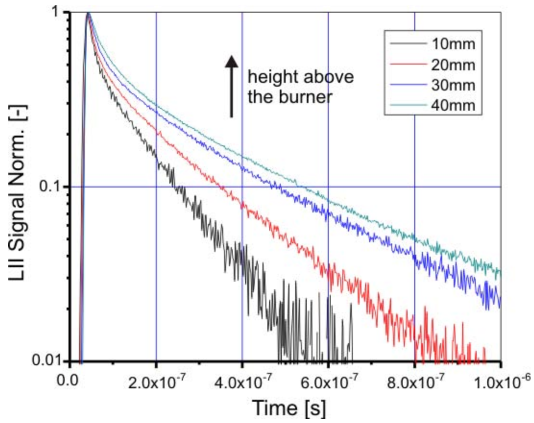

Fig. 1 Experimental LII signals from the undiluted propane diffusion flames recorded at different height above the burner. The normalized signals are displayed on a log scale indicating different decay times thus particle sizes.

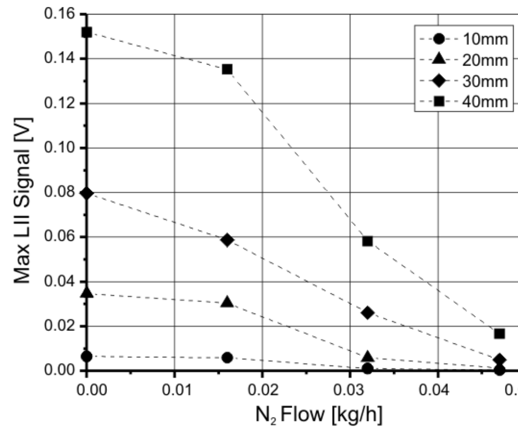

Fig. 2 Maximum LII signal as functions of nitrogen mass flow rate at different flame height.

After approximately 100ns after the laser pulse the particles cool down slowly and in accordance with their specific size: bigger particles cool down slower owing to their small surface/volume ratio, for small particles it is vice versa. At times after 150ns the signal shape is almost perfectly linear in the log-scale diagrams. The strong linearity indicates an exponential decay from a fairly mono-disperse particle size distribution. Looking at the height-dependent curves in Fig. 1 allows obtaining information about the soot formation behaviour. The characteristic signal decay is becoming slower indicating that the particles are growing. The slope of the decay, however, is changing significantly only between 10 and 30mm. Therefore we assume that particle growth is not continued or at least negligibly small at positions higher than an estimate of 50mm.

The maxi signals dependent on nitrogen flow rate and measurement height are displayed in Fig. 2. For the discussion of the influence of the nitrogen dilution only the signals at 20mm height above the burner are considered first. Starting from the undiluted flame which provides the strongest signal indicating the highest amount of soot, a reduction of soot concentration can be observed with increasing nitrogen flow rate. Adding 0.016kg/h N_2 the soot reduction effect, however, is already considerably. By adding 0.032kg/h and 0.047kg/h N_2 a reduction of 70% respectively 96% is achieved. The signal temporal shape is also changed with dilution but still shows rather linear decay behaviour. The heat transfer process is strongly dependent on the gas kinetics. The fuel dilution by N_2 changes the mean free path between molecules which is dependent on temperature and pressure. Both parameters temperature and pressure are used to get the Knudsen number which explains the decrease of primary particle size by increasing the inert gas. An increasing Knudsen number implies the reduction of the primary particle size. The decay between 150ns and 1000ns becomes much faster when increasing the nitrogen flow rate. The diluted flame indicates that a substantial fraction of the entire soot has a smaller primary particle size. This conclusion is supported by the results of the studies discussed in the introduction. When the hydrocarbon fuel is diluted with a non-hydrocarbon gas the probability of collisions between the different soot precursor molecules is smaller, hence the particle growth is hindered.

Summary

In the present work we studied the influence of nitrogen dilution on the sooting behaviour of laminar non-premixed propane flames. For this purpose time-resolved laser-induced incandescence was employed. The obtained signals revealed changes in soot volume fraction as well as primary particle size and size distribution. Dilution with the inert nitrogen gas was found to be an effective way to significantly reduce the amount of soot generated in the combustion process. Furthermore, the mean particle size decreased with increasing nitrogen flow rate.

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