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Further experimental and modelling evidences of soot fragmentation in flames

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Abstract

Particle oxidation is one of the steps still not completely understood in combustion. Most of the approaches are based on semi-empirical reaction rates. Correct evaluation of oxidation is needed to predict the final emission of particles in diffusion flames. Fragmentation has recently been proposed to be a controlling step in determining global soot burn out as well as the size of particles emitted.

The oxidation and fragmentation of soot particles is studied in a counterflow diffusion flames with in situ optical diagnostics, laser-induced incandescence and elastic light scattering. A sectional modeling approach is used to predict particle formation and burnout.

Two counterflow diffusion flames have been chosen, a soot formation (SF) and a soot formation/oxidation (SFO) flame.

Experimental data supported by model predictions show the role of fragmentation in controlling the burn-out and the size distribution of particles in flames. SF flame, where no soot oxidation occurs, shows large particles. By contrast in the SFO flame, the mean diameter of particles shows that when fragmentation is active coagulation is less effective, aggregates are hardly formed and primary particles with small size are mostly formed.

Keywords: Oxidation-induced fragmentation; Soot modeling; Laser induced incandescence; Counterflow flames

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1. Introduction

The understanding of the formation and burnout mechanisms of combustion-generated particles is still a challenge for the combustion community. Despite progress in diagnostic tools and modelling approaches there are some steps in particle formation and oxidation in flames which still remain elusive [1].

Particle oxidation is one of the steps not described at elementary level and semi-empirical reaction rates are used to account for its complexity. Soot oxidation is a mechanism which needs to be understood since controls the final amount of the particles emitted.

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Recently, it has been shown that during oxidation of soot particles in high temperature environments, fragmentation of particle agglomerates and even fragmentation of primary particles may occur [2]. The former happens if the oxidation of the particle occurs at a weak point of contact between two primary particles of a soot agglomerate, or at the bottom of primary particle pores. In these cases, fragments of soot agglomerates and of particles are formed and the oxidation and fragmentation processes control not only the final amount of the particle emitted but also their size distribution and morphology.

The processes described above have recently been incorporated into a sectional model [3] for particle evolution in flames. The model has been tested in premixed and diffusion flames. Other modelling approaches can be found [4,5], however none account for both aggregate and primary particle fragmentation and none have been tested against experimental data in both premixed and diffusion flames. The two stage burner experiment of Echavarria et al. [2] showed the increase of soot number concentration when a large amount of O_2 is present. No other approaches have tried to investigate this process.

Decoupling the processes of particle formation and burnout in diffusion flames is difficult. This is particularly true in coflowing flames where the two processes occur simultaneously close to the flame front and are difficult to be followed accurately. Also most of the soot burn-out is measured at the tip of the flame where flickering does not allow accurate spatial resolution.

Counterflow diffusion flames offer the possibility to decouple the two processes. By changing the fuel and oxidizer compositions and the flow rates it is possible to stabilize two different flame configurations. In the first configuration, named soot formation (SF) according to Chung and coworkers [6,7], soot forms on the oxidizer side of a stagnation plane, and once formed is convected away from the flame toward the stagnation plane. Soot oxidation is almost absent and restricted to OH which is present at low concentrations in the zone where soot is formed.

In the second flame configuration – soot formation/oxidation (SFO) – the main oxidation zone of the flame is located in the fuel side with respect to the stagnation plane. Soot formed in the fuel side of the flame is transported toward the flame front and it is finally oxidized by OH and O_2.

The main focus of this paper is to follow the processes of soot formation and oxidation in both SF and SFO counterflow flame configurations. Ethylene has been chosen because it allows different flame configurations to be realized. Laser induced incandescence and laser light scattering measurements between the two nozzles was carried out. These flames were modelled using a recently developed particle model which also includes the oxidation-induced fragmentation mechanism as well as the particle formation and oxidation steps. Comparison of model results with experiments is carried out to help understand the experimental data.

2. Experimental methods

The experimental apparatus consists of a counterflow burner, a Nd-YAG laser operated with its fourth harmonic at 266 nm and an ICCD camera. Two opposed jet nozzles (ID 2.54 cm) are vertically separated by 1.5 cm. with the oxidizer stream in the upper nozzle and the fuel stream in the lower nozzle. Screens are used at the exit of each jet to establish uniform gas flow velocities to generate stable, flat flames. The burner system is operated with a shield of nitrogen to protect the flame from the surrounding air. Using a mild vacuum through the holes in the annular section of the bottom burner, combustion products and shield gas are vented out of the system.

In the SF flame configuration, a flame is stabilized on the oxidizer side of the stagnation plane by feeding 75% C_2H_4, and 25% Ar as fuel stream, and 22% O_2 and the remaining Ar as oxidizer stream. The oxidizer and fuel stream velocities are fixed at 16.1 cm/s and 13.2 cm/s at standard conditions (1 bar and 298 K). The stagnation plane position estimated by visual observation of seeded micron particles in the oxidizer stream is located at about 4.2 mm from the fuel nozzle. Visual position of maximum flame luminosity is located at about 8 mm from the fuel nozzle.

The SFO flame was stabilized on the fuel side of the stagnation plane by feeding 15% C_2H_4 with 8% of O_2 and 77% Ar as fuel stream, and pure O_2 as oxidizer stream. Oxidizer and fuel stream velocities are fixed at 11.6 cm/s and 13.2 cm/s at standard conditions. The stagnation plane is located at about 8.5 mm from the fuel nozzle and the visual position of the maximum flame luminosity is about 7.3 mm from the fuel nozzle.

Measurements were performed over a range of positions between the nozzles by moving the entire burner assembly up or down with respect to the sampling point using a translation system, with a spatial resolution of 0.1–0.2 mm.

Laser Induced Incandescence (LII) measurements were performed using the fourth harmonic radiation (266 nm) of a Nd:YAG laser as excitation source. The laser beam was focused in the flame and a beam diameter at focal point of 350 μm was obtained. The energy of the laser pulse was kept constant at 0.8 mJ with a pulse duration of 8 ns. Higher energies of the laser pulse increased species fragmentation and consequently the interference of C_2 emission on fluorescence. Conversely, incandescence emission was enhanced.