

Soot particle size distributions in premixed stretch-stabilized flat ethylene–oxygen–argon flames

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Abstract

Evolution of particle size distribution function (PSDF) was studied in premixed stretch-stabilized flat flames for the first time. The purpose is to demonstrate that stretch-stabilized flames can broaden the experimental flame condition space for studying soot formation in a pseudo-one dimensional flow configuration. PSDFs were measured in three series of atmospheric-pressure ethylene–oxygen–argon flames with maximum temperatures around 1980, 2000, and 2160 K. The measured PSDFs show a strong effect of flame temperature as nucleation and growth of soot is found to be suppressed towards high temperatures. Simulations using a population balance soot model show reasonably good agreement with the 1980 and 2000 K series of flames, but it significantly overpredicts the number density and size of soot in the highest temperature flame. Numerical tests suggest that the discrepancy can be caused by high-temperature reversibility in surface growth and other processes.

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1. Introduction

It is well known that soot formation is greatly impacted by thermodynamic reversibility towards high temperatures. This reversibility causes soot yield to follow a rise-then-fall behavior or bell-shaped dependency with respect to temperature in

shock tubes [1–3] and flames [4–7]. At a detailed level, the kinetic causes for the reversibility of polycyclic aromatic hydrocarbon (PAH) formation and growth have been discussed some time ago [8,9]. For soot nucleation and size growth involving PAH clustering, PAH condensation and particle–particle coagulation, the importance of microscopic reversibility has been also discussed (see, e.g., [10–16]). More recently, detailed modeling of soot formation has generally considered the reversibility of soot particle nucleation and coagulation in the form of a stick probability (see,

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e.g., [12,14,15,17,18]), which is expected to be dependent on size, temperature and pressure. Physical and quantitative understanding of these dependencies is currently lacking. In particular, reversibility is amplified at high temperatures (>2000 K) yet experimental observations in flames with low-dimensional flow geometry (e.g., the McKenna type burner) above this temperature have proven to be difficult for reasons to be discussed below.

Previously we have shown that the burner-stabilized stagnation flame (BSSF) approach coupled with mobility sizing can yield a wealth of information about the competition between various soot growth processes [19–21]. The approach allows for the evolution of the soot particle size distribution function (PSDF) to be followed quite closely while reducing the problem of probe perturbation and ill-defined boundary conditions [21,22]. The data generated have been useful to model development and comparison (see, e.g., [14,18,23,24]). However, the BSSF approach has limitations. Fundamentally, flame stabilization is achieved through heat loss to the burner in burner-stabilized flames, thus it is generally difficult to access high temperature flames (>2000 K) where the effect of thermodynamic reversibility is expected to be amplified. High temperature flames also hasten non-uniform porosity changes in the porous plug thus creating a non-uniform radial velocity profile after some period of measurement – an experimental issue that would impact data quality [21]. This porosity change problem is further exacerbated in experiments for liquid fuels as keeping the fuel in a vaporized state usually requires heating of the porous plug. In addition to porosity changes, the elevated temperature increases heterogeneous reaction rates in the porous plug, which can lead to coking and changes in the composition of the unburned gas due to fuel cracking.

For reasons discussed above, it would be beneficial to develop a new burner that would remove the aforementioned problem and still allow for particle size distributions to be followed in a one-dimensional or quasi-one-dimensional flame system. In this work, we show that premixed stagnation flat flames offer this alternative configuration and demonstrate that such flames can be used to extend the experimental range for studying nascent soot formation through mobility sizing, while still maintaining well defined boundary conditions in a pseudo-one dimensional geometry. The so-called stationary premixed planar flame in stagnation flow (alternatively known as a stagnation flame or premixed stretch-stabilized flat flame) is commonly used to examine fundamental combustion phenomena like flame speed and extinction [25]. Stretch-stabilized flat flames have certain advantages over burner-stabilized flames. The flame stabilization mechanism of stretch-stabilized flames is based on flow stretch rather than heat loss. For this reason, stretch-stabilized flat flames have

maximum flame temperatures, $T_{f,max}$, approaching the adiabatic flame temperature, and in many cases exceeding the adiabatic flame temperature due to Lewis number effects [25]. Studies of low-vapor pressure, liquid fuels become significantly more reliable because stretch-stabilized flat flames are issued from a simple nozzle. In this work, experimental observations of the PSDF in a series of ethylene–oxygen–argon flames are compared to predictions from a population balance model. Relatively high flame temperatures are accessed with stretch-stabilized flat flames to assess the accuracy of a soot model against the measurement.

2. Experimental

The experimental setup, summarized in Fig. 1, consists of a burner with an aerodynamically shaped nozzle, a stagnation surface/sampling probe assembly and a scanning mobility particle sizer (SMPS). The aerodynamic shape of the burner nozzle body is designed to achieve plug flow at the burner exit (1.43 cm nozzle exit diameter). The distance between the burner nozzle and stagnation surface, L , is held constant at 1.0 cm. The standing distance between the flame and stagnation surface/sampling probe, L_s , may be varied by changing the unburned gas flow rate.

Flame sample was drawn into a micro-orifice (127 μm diameter, 125 μm wall thickness) embedded within and flush to the stagnation surface on a water-cooled aluminum disc. The techniques for sampling and dilution ratio calibration (based on CO_2 concentration measurement) are identical to our earlier work [21]. Figure S1 of the Supplemental material shows the dilution ratio calibration for the current stretch-stabilized flame. An optimum dilution ratio was established in accordance with previously established procedures [21,26,27]. The PSDF was determined in terms of mobility diameter as measured by the TSI SMPS (models and settings found in [19]) with previously described mobility diameter corrections [28,29].

The gas temperature at the nozzle exit, T_n , was measured with an uncoated fine-wire (125 μm wire diameter) Pt–Rh thermocouple placed near the centerline of the flow. Similar to the BSS flame sampling technique [19], the stagnation surface doubles as a sampling probe. The temperature at the stagnation surface, T_s , was measured by a type-R thermocouple (0.2 cm wire diameter) placed flush with the surface such that the bead is exposed to the sample gas 1 cm away from the centerline of the flame. The gas temperature at the nozzle exit was determined to be $T_n = 347 \pm 10$ K; and the stagnation surface temperature was $T_s = 385 \pm 25$ K. The uncertainty values quoted here are one standard deviation across all flames studied. Numerical sensitivity calculations show that within the uncertainty values quoted the flame structure exhibits

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