

LAMINAR SOOT PROCESSES (LSP) EXPERIMENT: FINDINGS FROM GROUND-BASED MEASUREMENTS

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INTRODUCTION

Processes of soot formation and oxidation must be understood in order to achieve reliable computational combustion calculations for nonpremixed (diffusion) flames involving hydrocarbon fuels. Motivated by this observation, the present investigation extended earlier work on soot formation and oxidation in laminar jet ethylene/air and methane/oxygen premixed and acetylene-nitrogen/air diffusion flames at atmospheric pressure in this laboratory [1-7], emphasizing soot surface growth and early soot surface oxidation in laminar diffusion flames fueled with a variety of hydrocarbons at pressures in the range 0.1-1.0 atm [8,9].

Sunderland et al. [1-3] investigated soot surface growth in laminar jet diffusion flames but were unable to evaluate available mechanisms of soot surface growth and oxidation because their measurements did not provide information about radical concentrations (e.g., H, OH and O) needed by the theories [10-14]. Xu et al. [4-6] continued the work of Sunderland et al. [1-3] by studying soot formation in laminar premixed flames. These measurements were used to evaluate the Hydrogen-Abstraction/Carbon-Addition (HACA) soot surface growth mechanisms of Kazakov et al. [10] and Colket and Hall [11]. It was found that the HACA soot surface growth mechanisms provided excellent correlations of the measurements using quite reasonable steric factors that appear in the theories. Xu and Faeth [7] extended the study of soot surface growth to diffusion flame environments, considering acetylene-nitrogen-fueled flames burning in coflowing air, and using the full suite of measurements developed during the premixed flame studies of Refs. 4-6. These results showed that soot surface growth rates in premixed and diffusion flames satisfy similar reaction rate expressions, and were well represented by the HACA mechanisms of [10] and [11]. The present investigation extends this research to consider the structure and soot surface growth and oxidation properties of laminar diffusion flames fueled with hydrocarbons other than acetylene (e.g., ethylene, propylene, propane and benzene) at pressures of 0.1-1.0 atm.

Early studies of soot surface oxidation by O₂ in nonflame environments were reported by Nagle and Strickland-Constable [12]. Subsequently, Neoh et al. [13,14] studied soot oxidation within laminar premixed flames. Soot surface oxidation in laminar diffusion flames has been studied as follows: in methane/air flames by Garo et al. [15,16], and in ethylene-nitrogen/oxygen-argon flames by Haudiquert et al. [17]. These diffusion flame studies supported the findings of Neoh et al. [13,14] of the dominant role of OH in soot oxidation at near-stoichiometric conditions in flames but yielded OH collision efficiencies that were not in particularly good agreement with the results of Neoh et al. [13,14] for premixed flames. One explanation for the disagreement was that optical scattering and extinction measurements were used to infer soot structure properties during the diffusion flame studies that were based on models that have not been very successful for representing soot optical properties [18-20]. Thus, the present investigation also sought to resolve problems of soot oxidation in diffusion flames using experimental methods developed during earlier studies of soot processes in this laboratory, which avoid the experimental problems of [15-17].

EXPERIMENTAL METHODS

The experiments considered here involved new measurements in diffusion flames at atmospheric pressure for various hydrocarbons burning in air and at pressures 0.1-1.0 atm

involving acetylene-nitrogen mixtures burning in air. Flame and soot properties in these flames were measured in the same manner as Xu and coworkers [4-7]. A total of six premixed and thirteen diffusion flames were used to evaluate mechanisms of soot surface growth and oxidation during the present study.

FLAME STRUCTURE

Typical of all flames considered thus far [1-7], soot in the present flames were aggregates of nearly spherical primary soot particles with the primary particle diameters being nearly monodisperse at given locations in a particular flame. Structure measurements (temperature, T , streamwise velocity, u ; primary particle diameter, d_p ; soot volume fraction, f_s ; and the mole fractions of stable and some radical (H, OH and O) species) were obtained along the axes of the test flames. Results are illustrated in Fig. 1 for an ethylene/air flame at atmospheric pressure which is typical of the other flames that were studied. Different flame conditions varied the concentrations of acetylene and H in the soot formation region, species that are largely responsible for soot growth through the HACA mechanism, but did not affect the fundamental soot surface growth mechanism. Thus, soot formation began where H-atom first appeared in the presence of significant concentrations of acetylene and ended where concentrations of acetylene became small in the presence of significant concentrations of H-atom. Similarly, varying fuel type varied concentrations of O_2 and OH that are largely responsible for soot surface oxidation but did not affect the mechanism of soot surface oxidation. Finally, soot oxidizing species, mainly OH and O_2 , are present throughout the soot formation region so that soot formation and oxidation proceed at the same time with the former dominating fuel-rich conditions and the latter dominating near-stoichiometric and lean conditions.

SOOT SURFACE GROWTH RATE PROPERTIES

Soot surface growth rates, w_g , were interpreted using the HACA soot surface growth mechanisms of Refs. 10 and 11, expressed as follows:

$$w_g = \alpha_i R_i \quad (1)$$

where $i = FW$ and CH denote the mechanisms of Refs. 10 and 11, respectively, the α_i are steric factors on the order of unity, and the R_i are the complex HACA reaction rate expressions given by Xu et al. [4]. As a first approximation, the R_i are proportional to the product of the concentrations of hydrogen-atom and acetylene, $[H][C_2H_2]$. Thus, measured values of $w_g/[C_2H_2]$ are plotted as a function of $[H]$ in Fig. 2, to provide a direct test of the main features of the HACA soot surface growth mechanism without the intrusion of uncertainties due to the numerous empirical parameters in the original detailed mechanisms; the correlation is surprisingly good indicating similar soot surface growth rate behavior for premixed and diffusion flames involving various hydrocarbon fuels and pressures of 0.1-1.0 atm. A more direct evaluation of the HACA mechanism of Colket and Hall [11] appears in Fig. 3; this mechanism provides a reasonably good correlation of the measurements with a steric factor on the order of unity as expected for soot surface growth rates for premixed and diffusion flames involving various hydrocarbon fuels and pressures of 0.1-1.0 atm. Results for the HACA mechanism of Kazakov et al. [10] were similar.

SOOT SURFACE OXIDATION RATE PROPERTIES

Similar to Neoh et al. [13], soot oxidation rates (corrected for effects of soot growth) were converted into collision efficiencies, for oxidizing species, i , as follows:

$$\eta_i = 4w_{ox}/(C_i[i]\bar{v}_i) \quad (2)$$

where w_{ox} is the rate of soot surface oxidation per unit area, C_i is the mass of carbon removed from the surface per mole of species i reacting at the surface, $[i]$ is the gas phase concentration of i adjacent to the surface, and

$$\bar{v}_i = (8R_u T / (\pi M_i))^{1/2} \quad (3)$$

is the mean molecular velocity of species i , R_u is the universal gas constant, T is the temperature and M_i is the molecular weight of species i . The measurements indicated that O_2 , CO , H_2O and O did not provide a good correlation of collision efficiencies for early soot surface oxidation. Results for OH , however, illustrated in Fig. 4, provided an excellent correlation for all the test flames, as well as for the earlier results in premixed flames due to Neoh et al. [13], yielding a steric factor of 0.13 with an uncertainty of 0.05, essentially independent of laminar flame type (premixed for diffusion), fuel type and pressures in the range 0.1-1.0 atm.

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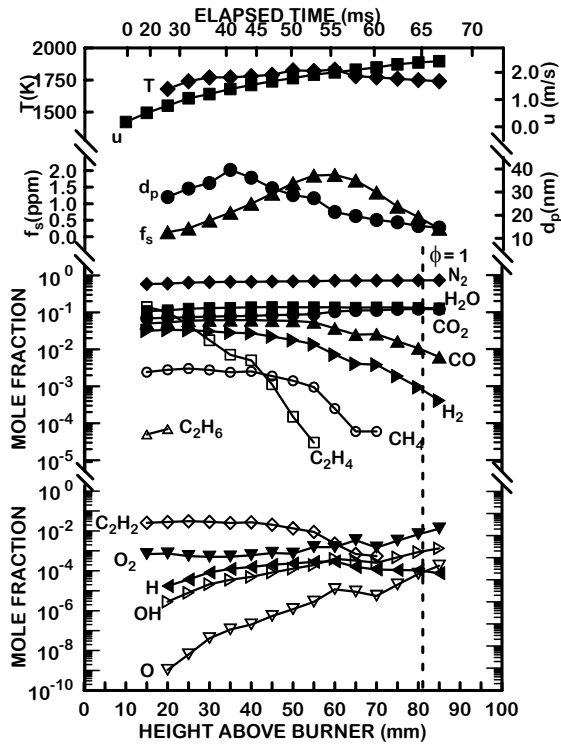


Fig. 1. Measured soot and flame properties in an ethylene/air diffusion flame at atmospheric pressure. From El-Leathy et al. [8].

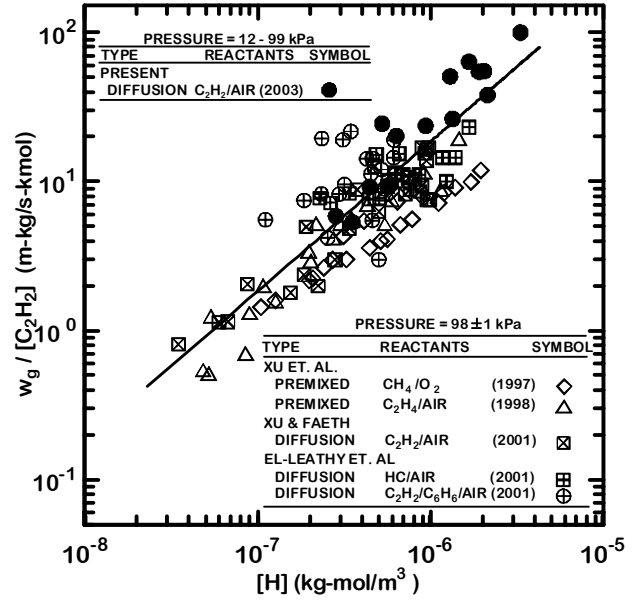


Fig. 2. Soot surface growth rates (corrected for soot surface oxidation) as a function of acetylene and hydrocarbon-atom concentrations for laminar premixed and diffusion flames (involving various fuel types and pressures).

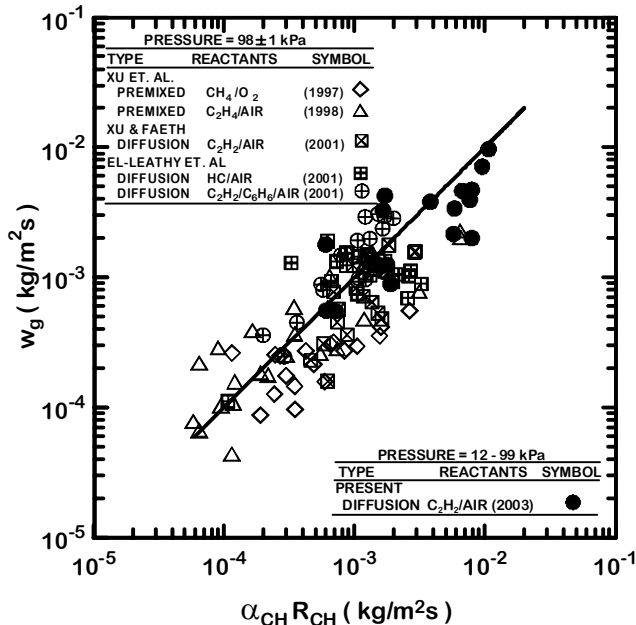


Fig. 3. Soot surface growth rates (corrected for soot surface oxidation) in terms of the HACA mechanism of Colket and Hall [11] for laminar premixed and diffusion flames involving various fuel types and pressures.

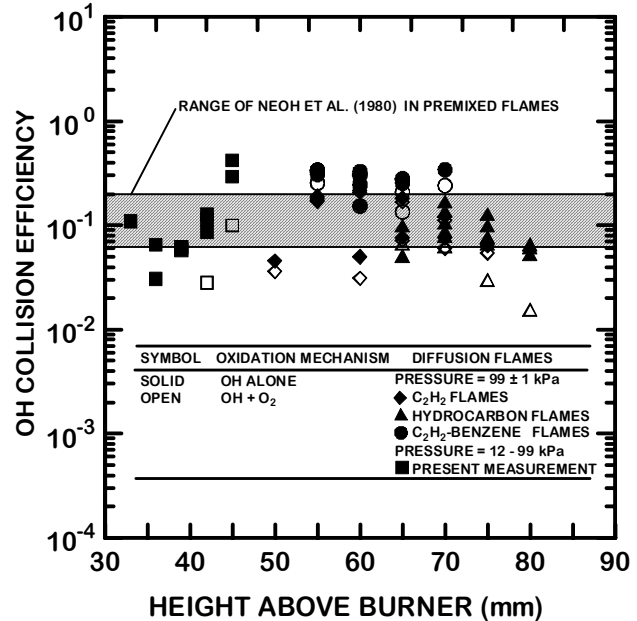


Fig. 4. Soot surface oxidation collision efficiencies for early soot oxidation assuming soot burnout due to OH and O₂ (with the O₂ rate taken from Ref. 12) for laminar premixed and diffusion flames involving various fuel types and pressures.