MECHANISMS OF SOOT FORMATION AND OXIDATION IN FLAMES

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Introduction

Soot formation and oxidation must be understood in order to develop reliable computational combustion methods for flames involving hydrocarbon fuels. Motivated by this observation, the present investigation extended earlier work on soot formation and oxidation to consider broader ranges of test conditions than considered before. In particular, past studies of TEM-observable primary soot particle surface growth involved measurements in laminar jet premixed and nonpremixed (diffusion) flames fueled with various hydrocarbons (CH_4, C_2H_2, C_2H_4, C_3H_6, C_3H_8 and C_6H_6) and having gas temperatures of 1500-1850 K at atmospheric pressure and yielded major findings as follows, see [1,2] and references cited therein: surface growth of primary soot particles is similar in premixed and diffusion flames, is independent of hydrocarbon fuel type, and can be correlated based on the Hydrogen-Abstraction/Carbon-Addition (HACA) mechanisms of soot surface growth due to Colket and Hall [3] and Kazakov et al. [4]. In addition, past studies of TEM-observable primary soot particle surface oxidation for the same range of flame conditions yielded the following major findings [1,2]: primary soot particle surface oxidation in laminar premixed and diffusion flames is similar, is independent of hydrocarbon fuel type, and can be correlated using the OH/O_2 mechanism of Nagle and Strickland-Constable [5] and Neoh et al. [6]. Finally, the properties of TEM-observable primary soot particles have received a great deal of attention, see [1-7] and references cited therein. Based on these studies, it is generally agreed that TEM-observable primary soot particles consist of nearly spherical and monodisperse primary soot particles having diameters of 10-50 nm, that are collected into mass fractal aggregates. More recent studies suggest that the formation of soot precursor particles that are not visible by TEM involves coalescence of large Polycyclic-Aromatic-Hydrocarbon (PAH) molecules that form by the HACA mechanism, see [8] and references cited therein. Unfortunately, effects of temperatures greater than 1850 K, and pressures other than atmospheric pressure on soot surface growth and oxidation are unknown. In addition, past work has not yielded a way to estimate rates of nucleation of TEM-observable primary soot particles. Thus, the objectives of the present investigation were to address these limitations by conducting additional experiments at flame temperatures up to 2350 K, pressures of 0.1-1.0 atm, and including observations of the nucleation rates of TEM-observable primary soot particles.

Experimental

Measurements were made along the axes of laminar jet premixed and diffusion flames as discussed by Kim et al. [1] and El-Leathy et al. [2]. This involved measurements of soot volume fractions by laser extinction, soot temperatures by multiline emission, soot structure by thermophoretic sampling and TEM, gas species concentrations by isokinetic sampling and gas chromatography, radical species concentrations by Li/LiOH atomic absorption, and gas velocities by laser velocimetry. These measurements were used to find TEM-observable primary soot particle surface growth rates, surface oxidation rates, and nucleation rates, as discussed by Kim et al. [1] and El-Leathy et al. [2].

Results and Discussion

Measured TEM-observable primary soot particle surface growth rates were correlated successfully for the extended range of test conditions mentioned earlier based on the HACA mechanisms of Colket and Hall [3] and Kazakov et al. [4] yielding unknown steric factors in these mechanisms on the order of unity as expected. The only exception was that it was necessary to delete Reaction 3 of Colket and Hall [3] in order to obtain the correct pressure dependence for this mechanism.

TEM-observable primary soot particle nucleation rates were correlated successfully by assuming that nucleation was controlled by the formation of large PAH particles. This yielded an effective TEM-observable nucleation rate correlation over a range of nucleation rates of six orders of magnitude that was first order in the concentration of acetylene and second-order in concentration of H.

Finally, TEM-observable primary soot particle early oxidation rates (before oxidation of more than 70 % of the maximum mass of the primary soot particles) were correlated successfully based on a collision efficiency of OH molecules of 13 %, supplemented by a relatively small contribution due to direct oxidation by O_2 based on the measurements of Nagle and Strickland-Constable [5], as proposed by Neoh et al. [6] based on measurements of soot surface oxidation in laminar premixed flames.

References

[1] Kim CH, El-Leathy AM, Xu F and Faeth GM. Soot surface growth and oxidation in diffusion flames at pressures of 0.1-1.0 atm. Combustion and Flame 2004; 136 (1/2): 191-207.
[2] El-Leathy AM, Kim CH, Faeth GM and Xu F. Soot surface reactions in high-temperature laminar diffusion flames. AIAA J. 2004; in press.
[3] Colket MB and Hall RJ. Success and uncertainties in modeling soot formation in laminar premixed flames. In Bockhorn H, editor. Soot Formation in Combustion, Berlin: Springer-Verlag, 1994: 442-470.

[4] Kazakov A, Wang H and Frenklach, M. Detailed modeling of soot formation in laminar premixed ethylene flames at a pressure of 10 bar. Combustion and Flame 1995; 100(1/2): 111-120.
[5] Nagle J and Strickland-Constable RF. Oxidation of carbon between 1000-2000 °C. Proceedings of 5th Carbon Conf., 1962; 1: 154-164.
[6] Neoh KG, Howard JB and Sarofim AF. Soot oxidation in flames, in Siegla DC and Smith BW, editors, Particulate Carbon, New York: Plenum, 1980: 261-277.
[7] Ishiguro T, Takatori Y and Akihama K. Microstructure of diesel soot particles probed by electron microscopy: first observation of inner core and outer shell. Combustion and Flame 1997; 108(1/2): 231-234.
[8] Zhao B, Yang Z, Wang J, Johnston MV, Wang H. Analysis of soot nanoparticles in a laminar premixed ethylene flame by scanning mobility particle sizer. Aerosol Science 2003; 37: 611-620.