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ABSTRACT

The predictions of “soot” concentrations from numerical simulations for nitrogen-diluted ethylene-air flames are compared with laser-induced incandescence and Raman spectra observed from samples thermophoretically-extracted using a rapid insertion technique. In some flame regions, the Raman spectra were obscured by intense, radiation that appeared to peak in the near visible flame region. The spectra consisted of overlapping bands between 1000 and 2000 cm⁻¹ dominated by the G-band, near 1580 cm⁻¹, and the D′ band in the upper 1300 cm⁻¹ range. Several patterns are explored to discriminate the data including 3- and 5-band models, and a Ewing/Williams/Phanes (EWP) model. Because the Raman signals were observed at heights where in situ-LII was not observed, we postulate that these signals may be attributable to smaller particles. The results suggest that the observed Raman signals are attributable to particulate with modests (1-3 nm) crystallite sizes. This observation is discussed in the context of current models for nascent particle formation.

Fitting Raman Spectra

Three different fitting schemes investigated to model the Raman references: G1, G2, and G3. (BWF) two-band (A); D1, G1, and G (BWF) five-band fit (C). In each panel are shown the raw data (black), fit spectrum calculated by summing the individual peak contributions (red), and the individual peak contributions (green). Observations of (a) blue line, (b) green line, (c) purple curve. The lighter gray curve is an empirical fit of filtered curves. The green shaded area region show the range of I/G observed in the present work.

Modelling Soot Morphology

The vertical bars show the corresponding calculated distribution of masses. Graph of computed flame temperature, the calculated sum of peak intensities is used to estimate the crystallite size, La. This quantity varies only slightly in all of the data and is estimated to be around 1.0-1.2 nm. These observations can be rationalized within the context of current models for soot growth. Frankel and Tidball found that particle size was determined primarily by physical coagulation and gas-phase reactions. For particles with crystallite sizes in the range of La = 1.0-1.2 nm, the PAH within some calculated nascent soot particles range from 1 to 2 nm, and the presence of a very few larger particles likely from coagulation.

Comparisons of (cy) calculated soot volume fraction from numerical simulation and (b) in situ-LII for each of the target flames: ethylene concentrations of 32, 40, 60, and 80 volume percent (left to right). Contours for computed hydroxyl radical, the sum of calculated soot classes 3 through 7. The observed ratio of D to G in regions where Raman scattering was not observed and could be quantified over the ex situ-LII signals. For clarity in the last panel, image where LII-LII dominated Raman in the middle of the flame are shown in dark blue.

Experimental

Flame Raman experiments were performed on flames at George Washington University and at Yale University. In each, a fuel mixture of ethylene and nitrogen flows through a 0.4 cm inner diameter vertical tube and air issues from the bottom of the tube and a 7.5 cm inner diameter concentric tube. Flames with different fuel mixtures, composed of ethylene and nitrogen, were studied with ethylene concentrations of 32, 40, 60, and 80 volume percent. For all flames, the fuel velocity at the burner surface was a parabolic profile with an average velocity of 35 cm/s and air velocity profiles at the burner surface were flow through 35 cm/s; physically this was realized by having a hemispherical cover the region of co-flowing air. The flame was unconfined, and the flow was laminar.

Raman Spectra vs. Position in Flame

Contours of computed flame temperature, the calculated sum of the concentrations of all soot classes with N ≥ 3 and the measured in situ-LII signal.

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