

SOOT PRODUCTION IN FLICKERING METHANE, PROPANE, AND ETHYLENE DIFFUSION FLAMES

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Diffusion flames originating from gaseous jets, liquid pools, or even solid materials frequently exhibit a periodic flickering behavior, whose effect on the chemical fields within the flame is not understood. In particular, the slow rates of soot particle inception chemistry and of carbon monoxide oxidation might be expected to result in strong sensitivity of soot and CO production to the complex, time-varying flowfields present in flickering flames; this would have important consequences for flame radiation and the emission of smoke and CO. Indeed in an earlier study of OH• laser-induced fluorescence and elastic scattering from soot in steady and time-varying methane/air flames, it was found that the soot scattering intensity in flickering flames was significantly greater than that observed for the steady (i.e., unforced) flame with the same mean fuel and air flow rates [1]. Measurements of the soot volume fraction fields in the steady and a moderately flickering flame reveal a factor of 5 enhancement in the peak soot volume fraction and a factor of 4 increase in the time-averaged, volume-integrated soot volume fraction in the flickering flame [2]. Mie analysis of the soot volume fraction and vertically polarized scattering measurements suggests that in the flickering methane/air flame soot particle number densities generally are comparable to steady flame values, whereas mean particle sizes are as much as 50% larger. Since diffusion flame soot chemistry is highly sensitive to fuel type [3] and methane is the weakest sooting hydrocarbon fuel, the trends in soot production observed for flickering methane flames may not hold for other hydrocarbon fuels. Therefore, measurements of soot scattering and soot volume fraction, as well as visible flame luminosity, have been made on steady and flickering propane/air and ethylene/air diffusion flames.

The burner and phase-locked imaging setup for studying time-varying diffusion flames have been described in detail previously [1]. In brief, unconfined laminar diffusion flames were stabilized on a coannular burner with a loudspeaker attached to the plenum below a 1.1 cm diameter fuel tube. The mean fuel flowrates were normalized to the methane flame fuel flowrate (7.7 cc/s) on a carbon atom basis, giving visible flame heights of ~ 85 mm and 88 mm for the propane/air and ethylene/air steady flames, respectively, compared to 79 mm for the methane/air flame. Measurements were performed on these steady flames and on moderately and strongly flickering flames produced by applying sine waves of magnitude 0.75 V and 1.5 V (pk-to-pk) to the plenum loudspeaker. Figure 1 shows previously measured OH• radical fluorescence and soot scattering for the two flickering methane flames. The flickering propane and ethylene flames show similar dynamics but are somewhat thinner than the methane flames, consistent with the decreased effect of buoyant expansion for these more heavily sooting fuels [4].

Figure 2 shows the optical arrangement for measuring polarization-specific soot scattering at visible wavelengths. An intensified CCD camera, with attached glass or dielectric filters, was used to record planar images at 90° to the direction of propagation of the laser light. For the scattering measurement, an attenuated reflection of the incident laser sheet was directed to the side of the CCD pixel array, allowing shot-to-shot correction of the measured signals for laser energy in the vertical plane. Comparisons of laser extinction derived soot volume fraction profiles and laser-induced incandescence (LII) signals in the steady methane flame have shown good agreement [2], lending support to the interpretation of properly calibrated LII as soot volume fraction. In this study soot volume fraction was measured by removing the sheet-forming lenses and polarization optics from the scattering setup and focusing the laser beam at the burner centerline to produce energy-saturated incandescence, which was recorded as a line image on the CCD. Visible flame luminosity was measured simply by turning off the lasers and using a short-pass filter (415-550 nm) in front of the camera.

The peak values of soot scattering and soot volume fraction measured in the different flames are shown in Table I and reveal some interesting trends. For moderately flickering conditions, the propane and ethylene flames both show an enhancement in the peak soot volume fraction of approximately 60% and in soot scattering of 170–200% in comparison to their corresponding steady flames. These increases in soot concentration and soot scattering are substantially less than the factors of 5.3 and 37 observed in the methane flame, but result in visible emission of smoke from the flickering propane and ethylene flames. For all three fuels investigated, there is no further increase in the peak soot scattering or soot volume fraction as the flickering intensity is changed from moderate to strong. Table I also shows the peak luminosity signals in the different flames, relative to the peak

intensity in the steady methane flame. The flickering methane flames exhibit substantially larger relative increases in peak luminosity than the propane and ethylene flames, as might be expected based on the soot volume fraction results. Otherwise, the trends in the peak soot volume fraction noted above are not well followed by the luminosity, presumably due to the strong dependence of soot radiation on the local temperature and thus the local dynamics of the flowfield.

REFERENCES

1. Smyth, K. C., Harrington, J. E., Johnsson, E. L., and Pitts, W. M., *Combust. Flame* 95:229-239 (1993).
2. Shaddix, C. R., Harrington, J. E., and Smyth, K. C., 25th Symp. (Intl) on Comb. (1994), in press.
3. Glassman, I., 22nd Symp. (Intl) on Comb., The Combustion Institute, Pittsburgh, 1988, pp. 295-311 and references therein.
4. Kaplan, C. R., Baek, S. W., Oran, E. S., and Ellzey, J. L., *Combust. Flame* 96:1-21 (1994).

TABLE I

Fuel	Peak scattering at 560 nm $Q_{VV, 90^\circ}$ ($10^{-5}/\text{cm-sr}$)			Peak soot volume fraction f_v (ppm)			Peak visible luminosity (relative to steady CH_4)		
	steady flame	0.75 V flame	1.5 V flame	steady flame	0.75 V flame	1.5 V flame	steady flame	0.75 V flame	1.5 V flame
	methane	7.6	280	310	0.30	1.6	1.7	1.0	2.6
propane	650	1300	1300	5.2	8.2	6.6	5.1	5.5	8.4
ethylene	1400	2400	2200	8.0	12	14	5.6	9.3	11

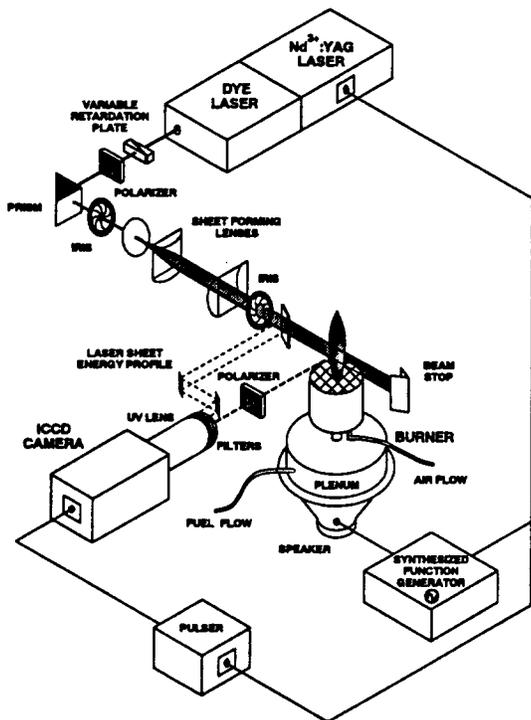


Figure 1. Experimental set-up for 1-D or 2-D imaging of axisymmetric diffusion flames which are acoustically excited and phase-locked to the pulsed dye laser system operating at 10 Hz. Images are recorded using an intensified charge-coupled device (ICCD) camera.

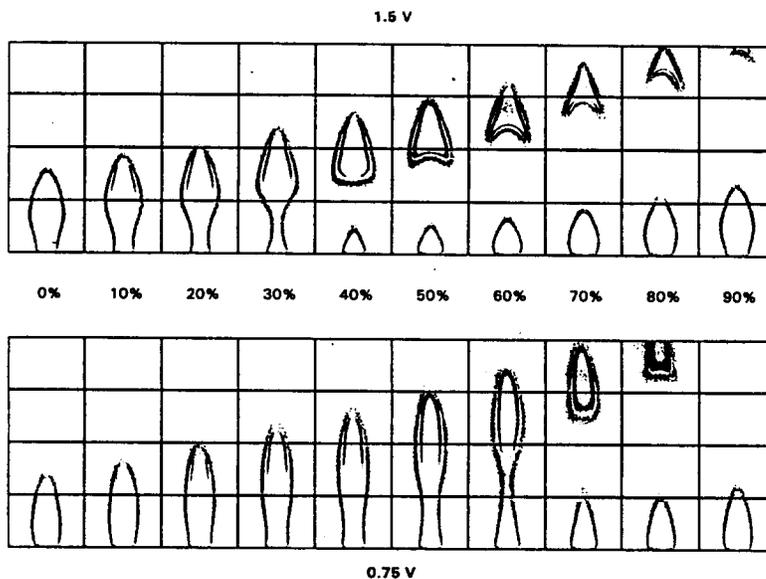


Figure 2. Soot scattering and OH^\bullet laser-induced fluorescence in flickering methane/air diffusion flames using horizontally polarized light at 283.55 nm. The OH^\bullet fluorescence signals surround intense scattering from the soot particles. Ten phases (arbitrary zero) are shown at two excitation voltages, corresponding to time intervals of 10 ms.