Laser-induced fluorescence of NCN in low and atmospheric pressure flames

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Received July 31, 2008; revised September 17, 2008; accepted September 23, 2008; posted October 8, 2008 (Doc. ID 99611); published November 11, 2008

The cyanonitrene radical, NCN, is detected by laser-induced fluorescence in laminar, adiabatic, flat $\phi=1.3$ methane–air flames at 200 hPa and atmospheric pressure. Laser excitation of the $\tilde{A}^3\Pi_g(020)\rightarrow \tilde{X}^3\Sigma_g^-(000)$ band at 317 nm allows off-resonant fluorescence to be detected at 326 nm. Excitation and dispersed fluorescence spectra are presented, as well as profiles of NCN and CH versus height above burner.

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OCIS codes: 120.1740, 300.6170, 300.2530, 300.6540, 280.2470, 280.1740.
The LIF signal is observed at right angles to the laser beam with an intensified CCD camera (Princeton Instruments THM, 512×512 pixels, 16-bit dynamic range), which is mounted behind a spectrograph (Acton SP300i) equipped with a UV Nikkor f=105 mm, f/4.5 lens. The spectrograph entrance slit is parallel to the laser beam and the burner deck. Dispersed fluorescence spectra have been taken with a 2400 grooves/mm grating, yielding a total view of about 18.8 nm in one direction of the camera image (0.037 nm/pixel). The perpendicular image direction represents spatial information (at 0.15 mm/pixel) and captures more than the entire width of the flame. To increase the signal–noise ratio, however, spectra have been recorded while binning together pixels in the spatial direction over almost the entire width of the flame. In this way dispersed fluorescence spectra could be recorded rapidly and continuously while the laser excitation wavelength was scanned slowly, yielding two-dimensional excitation–dispersion spectra. Figure 1 presents an example of this, in which 100 laser shots are integrated per dispersed fluorescence spectrum. From these spectra separate excitation and dispersed fluorescence spectra are easily obtained. The spectrograph–camera combination is also used to record the position of the flame front, using CH⁻ chemiluminescence as a marker.

The LIF signal of NCN and spectrally nearby transitions of OH (A–X system) and CH (C–X system) are recognizable in Fig. 1 and the wavelengths of OH and CH lines are used for accurate spectral calibration of both wavelength axes using LIFBASE [20]. Employing an excitation–dispersion spectrum, as displayed in Fig. 1, shows that in case of nearly equal excitation wavelengths, species can still be identified and spectrally separated by their dispersed fluorescence wavelengths. Reference measurements of CH LIF are performed by exciting the P1(9)(0,0) C–X transition at 316.943 nm. The population of the lower level of this transition is relatively insensitive to temperature variations in the range encountered in this flame, so that its intensity is a coarse measure of the local CH density.

An excitation spectrum of the A–X (020)–(000) band head of NCN at 316.970 nm [10], derived from Fig. 1, is presented in Fig. 2. Only fluorescence between 325.67 and 326.00 nm is selected for extracting the excitation spectrum. The P2(15) (0,0) and P2(15) (0,0) OH are identified: NCN and P2/Q1 OH are explained in Fig. 2; a=P2(15) (0,0) OH, b/c=Q1/Q2(21) (1,1) CH (λdisp =322.7 nm for a–c). Low intensities are enhanced in a nonlinear way (detector saturated near laser wavelength).
The thickness of the flame front and its separation from the burner allow dispersed fluorescence spectra of NCN to be recorded versus \( h_{ab} \) at 200 hPa. To this end, a linear background is fit and the remaining net signal between 325.71 and 326.07 nm is integrated. A similar procedure is followed for dispersed fluorescence spectra of CH after exciting the \( P_{1}(9)(0,0) \) C-X transition. In this case the net signal of the \( Q \) branch is integrated between 313.87 and 314.79 nm, with integration of 200 laser shots per spectrum being sufficient. The resulting variations of net NCN and CH signals with \( h_{ab} \) are shown in Fig. 4.

From this data it is clear that the NCN signal peaks somewhat higher above the burner than CH, that the NCN distribution is somewhat narrower than that of CH, and also that the presence of NCN is limited to the flame front. These results are similar to those observed in [15–17]. In those studies the FWHM of the NCN distribution is slightly larger than 2 nm (at \(<53 \) hPa), whereas here it is about 0.9 nm at 200 hPa. This difference is caused by the higher pressure. The \( h_{ab} \) of maximum NCN signal presented here differs from that in [15–17], but apart from pressure effects this may be due to different burner–flame conditions in those studies.

In summary, clear spectra of the NCN \( A^3\Pi_u(020) - X^3\Sigma_g^+(000) \) band have been recorded in a \( \phi = 1.3 \) methane–air flame at both 200 hPa and atmospheric pressure. This extends NCN measurements to higher pressures than reported in earlier literature [15–17]. Using the off-resonant excitation–detection scheme presented here (\( \lambda_{exc} = 316.970 \) nm and \( \lambda_{disp} = 325.90 \) nm) allows unambiguous detection of NCN [10]. It also allows NCN LIF to be measured in situations with more laser light scattering, either from Rayleigh scattering at higher pressures or reflections off surfaces. Variation of the NCN signal (with respect to that of CH) with the height above the burner, as observed here at 200 hPa, is similar to that presented in [15–17].

Discussions with A. A. Konnov (Vrije Universiteit Brussel and Technische Universiteit Eindhoven) are gratefully acknowledged, as is technical support from L. Gerritsen and A. P. van Vliet and financial support from the Technology Foundation Stichting Technische Wetenschappen.

References

Fig. 4. (Color online) NCN and CH LIF signal versus \( h_{ab} \) in the 200 hPa flame. Gaussian fits guide the eye.