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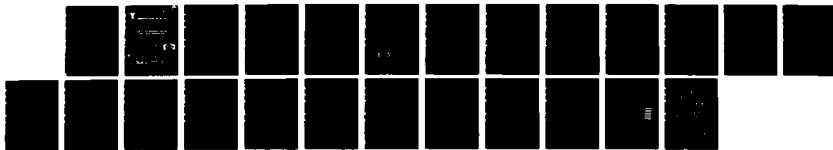
RADICAL FLUORESCENCE STUDIES WITH LIQUID PROPELLANTS
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20. Abstract (Cont'd):

effect of the LP on these flames at atmospheric pressure. NCO was likely present, but would not have been observed under the conditions of these observations. From the convolution of light intensity and detector response, the radical CN is the clear choice for maximum signal-to-noise ratio. Preliminary two-dimensional imaging of the fluorescence was done to demonstrate the utility of the technique for the study of liquid propellant sprays. Thus, the probability of success for active laser probing of LP spray combustion looks excellent, with the prime candidate for probing being the CN molecule. Other candidates such as C_2^H and NO_2^H are possible candidates with continuous or high rate metal vapor lasers.



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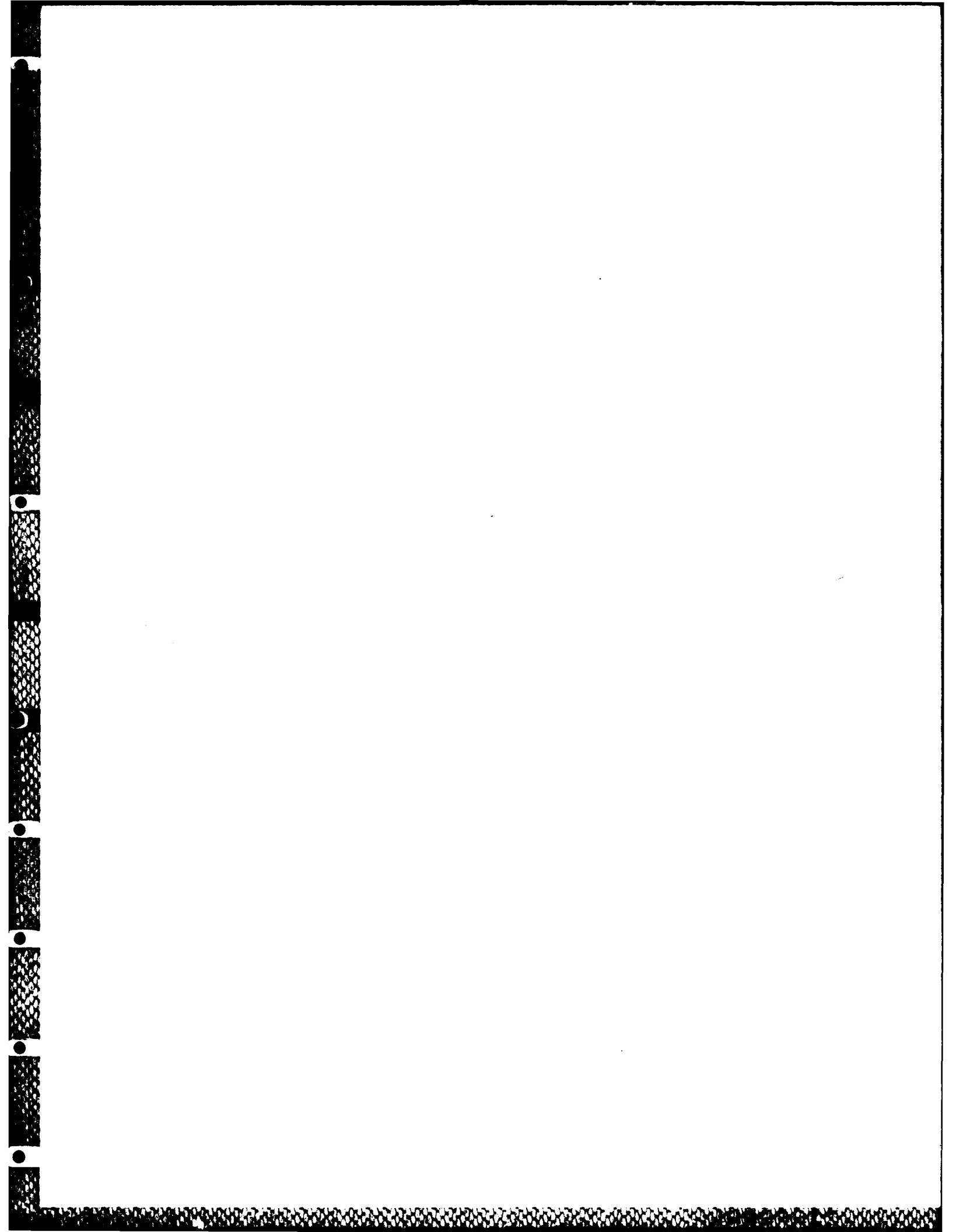
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I. INTRODUCTION

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In the development of liquid propellants for regenerative guns, the physical location of ignition and combustion of a jet or spray of propellant as it enters the combustion chamber can provide insight into the rate of energy release in these systems. Typical traditional methods of studying such phenomena are high-speed photography and shadow or schlieren techniques. While much can be learned with these techniques, they can be limited by their inherent line-of-sight characteristics. An alternative approach is to use a technique which has been recently demonstrated and developed to a practical level in many combustion systems: two-dimensional or planar laser induced fluorescence (2-D LEF).¹

In order to use the 2-D LEF technique, a molecule must be selected which is both reasonably abundant and characteristic of the pyrolysis or combustion process of interest. These species are typically diatomic or triatomic radicals which are created and rapidly consumed in the process. Typical species might be NO₂ for a pyrolysis experiment or OH for combustion. The beam from a laser tuned for resonant absorption by the probed species is expanded in one dimension to form a sheet of light. An optical system with its axis normal to the plane of the laser beam is used to image the resulting fluorescence onto the detector. The result is an image of the concentration of the probed species in a slice through the combustion event. Thus one may obtain information which is free of boundary layer effects and which can be complimentary to other diagnostic observations. Key parameters are the selection of the radical to be probed and choice of detector with a major trade-off being between framing rate and sensitivity. In the present study the radicals present in the ignition and combustion of liquid propellants have been characterized. Our goal is to develop this technique with specific application to liquid propellant materials and pursue its application under high pressure conditions.

II. OBSERVATIONS

A. Emission Spectra

The liquids of interest here are composed of hydroxylammonium nitrate (HAN), an aliphatic ammonium nitrate (e.g., isopropylammonium nitrate, IPAN), and water. A brief study of the structures of these molecules suggests that during reaction a number of diatomic radicals might be formed which would be suitable for identifying regions of reaction, including NH, NH₂, CN, and OH. One would also expect NO₂ to be produced during some stages of reaction. The most direct way to obtain evidence of the presence of such species under flame conditions is to introduce small quantities of the appropriate liquids into a flame, in a manner similar to conventional analytical chemistry techniques. This was done by using small custom made burners of various geometries which

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- (a) R.K. Hanson, G. Kychakoff, E.C. Rea, Jr., B. Hiller, R.D. Howe, and M.A. Kimball-Linne, Proceedings of the 20th JANNAF Combustion Meeting, CPIA Pub. No. 383, p. 43, 1983. (b) M.J. Dyer and D.R. Crosley, Opt. Lett. Vol. 7, p. 328, 1982. (c) R.J. Cattolica and S.R. Vosen, Sandia Report SAND84-8640, 1984.

contained separate flow channels for hydrogen and an oxidizer (either air or nitrous oxide) which burned as a highly turbulent diffusion flame. A few hundred microliters or less of the liquid was introduced into the flow of air. The air flow tube was approximately 0.8 mm in diameter; the resulting high Reynolds number flow caused the atomization of the liquid into small droplet sizes which were rapidly heated and consumed by the flame. The emissions from the flame were dispersed by a 0.25 m spectrograph and detected by a silicon intensified target (SIT) vidicon tube with an ultraviolet scintillator.

Spectra have been observed during the injection of both aqueous HAN and HAN/IPAN/water mixtures. In all cases the emission was spectrally typical of that from gaseous hydrocarbon flames. Preliminary measurements using a hydrogen/nitrous oxide flame showed that the emission from the radicals OH and NH, which are already present in abundance in the flame, was decreased when the liquid was added. This effect was probably due to cooling of the flame by the addition of the liquid. Subsequently all spectra shown were taken with air as the oxidizer. With HAN injection into the air flame, NH emission ($A^3\Pi-X^3\Sigma^-$) was positively identified, as shown in Figure 1. Although the SIT tube sensitivity near 335 nm is down about a factor of seven compared to 385 nm, the NH signals were substantially weaker than expected, based on the observations with the hydrogen/nitrous oxide flames. Also observed was broadband emission centered at about 500 nm which was probably due to NO_2 . An additional peak in the region of NH_2 emission near 547 nm was observed; positive identification was not made, however. With the injection of HAN/IPAN/water mixes into the flame, NH signals remained weak, CN ($B^2\Sigma^+-X^2\Sigma^+$) Figure 2, and CH ($A^2\Delta-X^2\Pi$) Figure 3, were strong. In this case C_2 ($d^3\Pi_g-a^3\Pi_u$) was clearly present as shown in Figure 4. However, the C_2 intensity is about a factor of ten below that of CN. The unidentified emission was again observed near 547 nm, although with a broader peak than with HAN. Again, the broadband emission characteristic of NO_2 was observed, as seen underlying the C_2 spectrum in Figure 4. From our past experience with similar flames,² it is also very likely that NCO is present; its emission would be masked by that of CH in Figure 3 [see Figure 5 of Reference 2].

B. Two-Dimensional Fluorescence Emission

As a first step prior to two-dimensional laser probing of radical concentrations, the spatial distribution of the spontaneously emitting radicals was studied when a droplet was introduced into a flame. The experiment consisted of a hydrogen/air flame stabilized on a 3/4 mm diameter sintered bronze burner. A droplet of approximately 1 to 2 mm³ was introduced into the hot gas region immediately above the flame by holding it on a piece of number 18 nickel wire. Under these conditions a droplet typically survived several seconds including a heating phase, sputtering and fizzing, and finally being consumed, usually with a bright flash of light. The emission from the droplet was imaged with an f/10 quartz optic onto a silicon intensified target (SIT) vidicon tube, part of a PARC, Inc. OMA-I system. For the work reported

2. W.R. Anderson, J.A. Vanderhoff, A.J. Kotlar, M.A. DeWilde, and R.A. Beyer, J. Chem. Phys., Vol. 77, p. 1677, 1982.

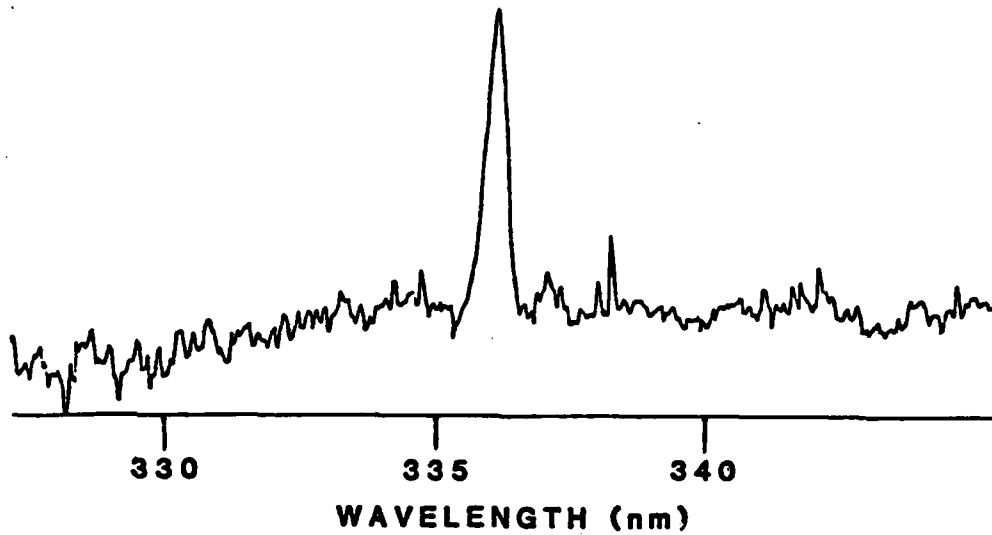


Figure 1. NH ($A^3\Pi-X^3\Sigma^-$) Emission Resulting from the Injection of HAN Into the Hydrogen/Air Flame. Note the lower signal level compared to the background noise levels in Figures 2-4.

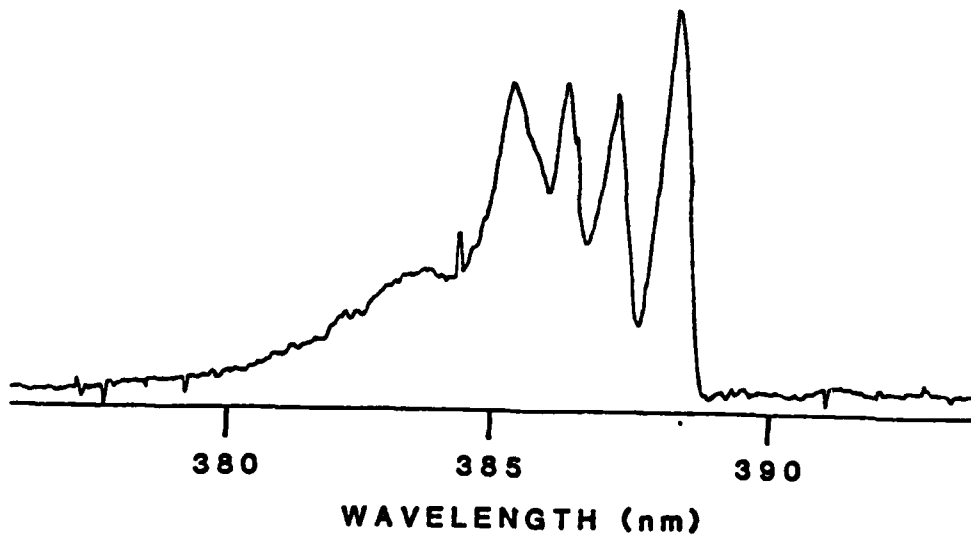


Figure 2. CN ($B^2\Sigma^+-X^2\Sigma^+$) Emission Resulting from the Combustion of a Typical HAN/IPAN/Water Propellant in a Hydrogen/Air Flame

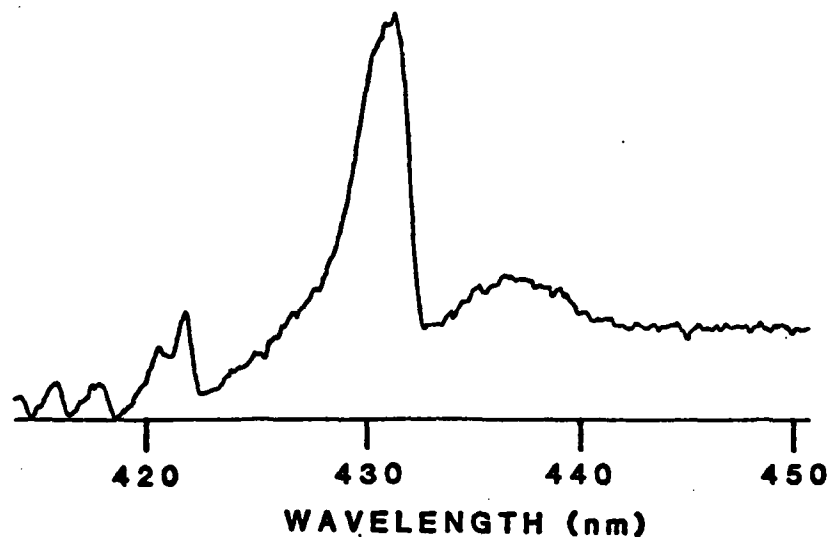


Figure 3. CH ($A^2A-X^2\pi$) Emission from the Combustion of the Same Liquid as Figure 2. Note that CN emission is also present at the shorter wavelengths shown.

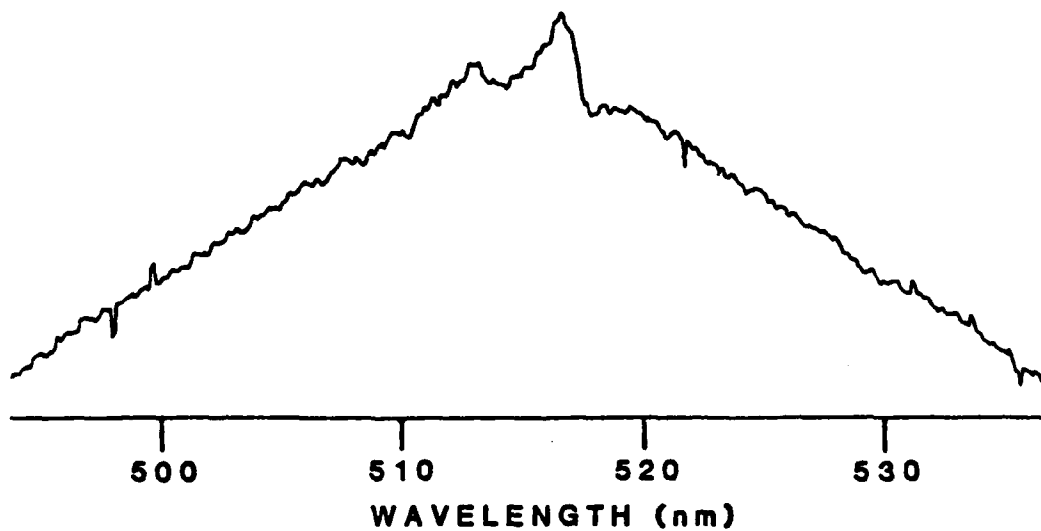


Figure 4. C_2 ($d^3\pi_g-a^3\pi_u$) Emission (The Two Distinct Peaks in the Center) Atop a Broad Pedestal of Emission from an Unidentified Species, Probably NO_2

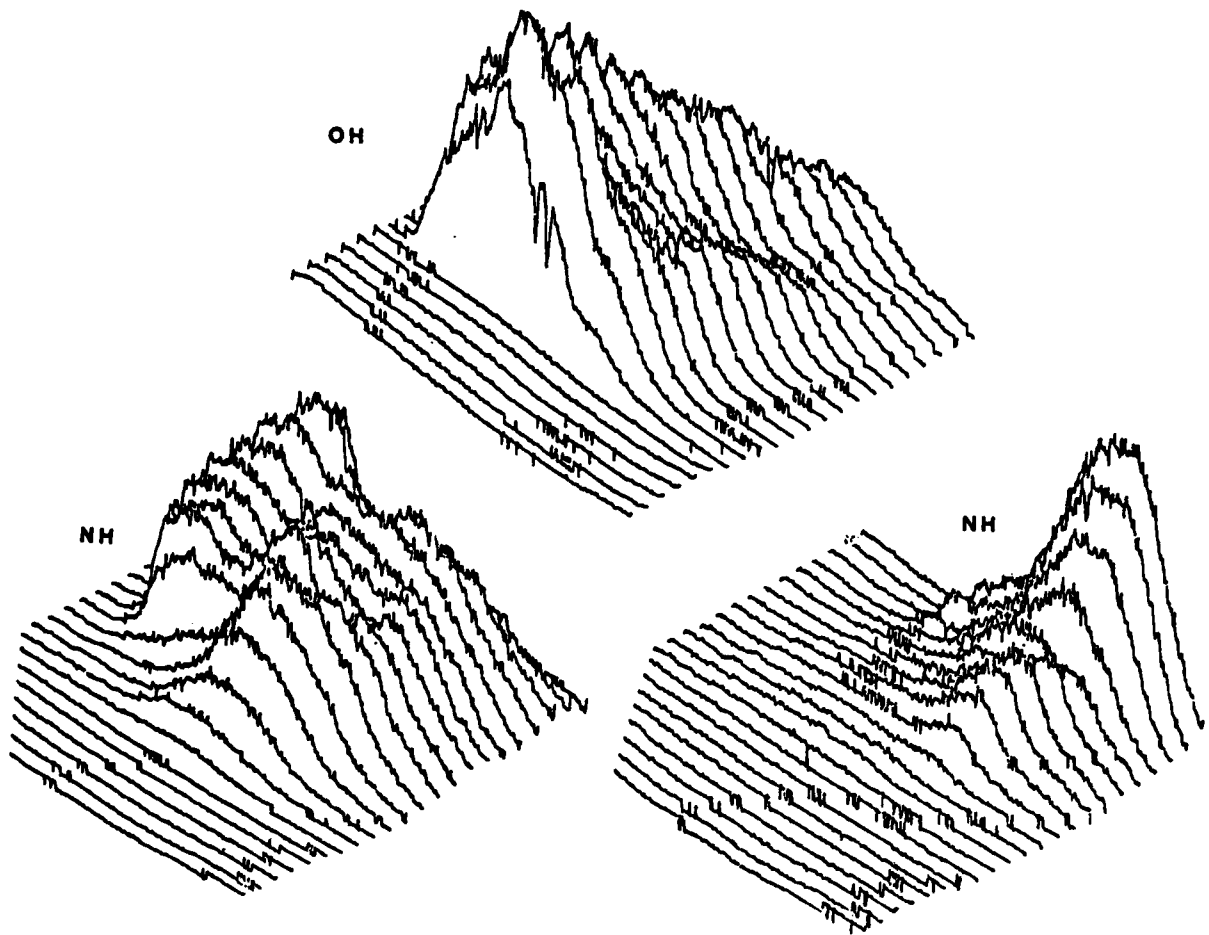


Figure 5. Spatially Resolved Fluorescence from a Liquid Drop Above a Sintered Bronze Burner, Showing OH, and NH from Two Different Drops. The burner surface is between traces 6 and 7 of the plot; the flow of hot gases is up and to the right of the figure.

here, the vidicon was set up with a 33 line scan of 500 points each. Although this resolution is somewhat limited, it is adequate for preliminary studies. The video signal was digitized and stored by a dedicated PDP 11/34 laboratory computer. The data were then plotted in a hidden-line manner to show the variations across the imaged area. Various narrow bandpass filters were used on the detector to isolate the emission from individual species.

Spatially resolved emission was recorded for OH, CN, and NH with droplets of propellant "1846." This propellant's composition is 60.8 weight percent HAN, 19.2 weight percent triethanolammonium nitrate (TEAN), and 20% water. Typical plots made with fixed image area are shown in Figure 5 for OH from the flame and NH from two different droplets. Thus the OH plot in Figure 5 can be used to define the position of the burner and flow of hot gases. The two NH plots in Figure 5 show the variability of combustion behavior of two similar drops. The drop shown on the left ignited in place and was swept away with the hot flow. The second NH plot, from a more violent event, shows an

apparent bifurcation and two regions of combustion. The NH emission in this latter plot extends down to the burner surface. The spatial separation of the NH and OH is clear in this figure. Signal strength in emission is certainly more than adequate; in many cases, especially with CN, the detector was saturated in the region of droplet combustion. The limited resolution of the detector (33 lines used, 22 lines shown in the plots) is clearly adequate for many studies. It is also very apparent that reproducibility from droplet to droplet is not good under these conditions, as might be expected. It was not clear when performing the observations that apparent energy release during the event and signal level were correlated well with changes in droplet size.

III. PROGNOSIS FOR PLANAR LASER FLUORESCENCE PROBING

Several factors must be considered in choosing a candidate molecule for probing in these systems. Possible choices of interest here are: OH, NH, CN, C₂, NCO, CH, NO₂, and NH₂. The anticipated behavior would be that NH, NH₂, and possibly NO₂ would be produced in pyrolysis or earlier in the flame, and be rapidly consumed in the flame reaction zone. CN, C₂, CH, and NCO are both created and consumed in the narrow region of combustion. OH is typically much longer lived and can survive well beyond the actual flame region. Spectroscopically, NH₂ and NO₂ are different from the diatomics in that their emission tends to be broad and spread over wider regions. This same property of NO₂ also gives it good absorption of most visible wavelengths, allowing the use of fixed frequency lasers. However, preliminary attempts with laser fluorescence of the pyrolysis gases of these liquids has shown that under at least some conditions almost no NO₂ is formed. Thus care must be taken in choosing it as a target molecule. Of the diatomic species, NH would chemically be the species of choice for studies of chemical reaction early in the combustion process. Unfortunately, the NH optical transitions are in a region where relatively little laser power is available; these emission studies also suggest that the NH which is present does not survive the flame long enough to build to appreciable concentrations in the hot regions. OH has been used in previous 2-D LEF studies of flames, but is of lesser interest here because it survives long after most of the heat release is accomplished; thus it is not a good indicator of narrow regions of flame reaction zone. However, OH has the substantial advantage of being extremely well characterized spectroscopically, and is not easily dismissed as a possible probe species. As mentioned above, CN is generally confined to the reaction zone of a flame. In addition, because of transition strength values, CN might provide signal levels approximately two orders of magnitude above those of OH under similar conditions. The additional attraction of good laser power availability at the right wavelengths also makes CN attractive. C₂ has favorable lifetime and transition strength values, as well as having transitions at wavelengths which are accessible. However, it is not a strong emitter in our studies and may not be present in sufficient abundance to be a good probe species. Other possibilities, NCO and CH, have optical transitions in unfavorable regions from the consideration of laser power for pulsed, tunable lasers.

The above discussion is centered heavily on the concept of one diagnostic pulse per event. Other possibilities are also being considered. These include the use of a high repetition rate copper vapor laser which could excite NO₂ and possibly C₂, and cw argon or krypton lasers, which can excite

NO₂, C₂, and NCO, with detection by high speed camera in both cases. Possible improvements in electronic cameras or other techniques are also being considered to allow higher framing rates.

IV. SUMMARY

Flame emission studies have been performed to identify and to show qualitative relative abundances of radicals present in the combustion of typical liquid propellants. From these observations and spectroscopic considerations, it has been concluded that CN has several advantages as a target molecule for a single pulse experiment. Other approaches which will allow observations which are closer to a high speed movie of radical spatial distributions are also possible and will be pursued.

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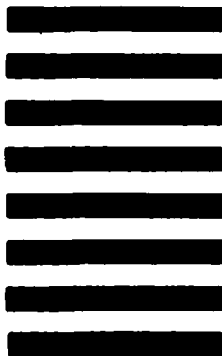
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