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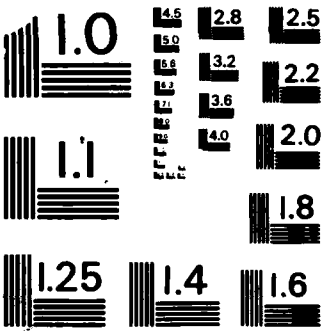

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TEMPERATURE AND CONCENTRATION PROFILES IN HYDROGEN-NITROUS OXIDE FLAMES

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July 1986

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Spontaneous Raman spectroscopy has been used to measure temperature and NO, O , and N concentration profiles in lean to stoichiometric premixed laminar hydrogen-nitrous oxide flames. Relative concentration profiles for OH were also obtained for these flames by use of laser induced fluorescence. The present NO concentration results for the stoichiometric flame agree with previous NO measurements obtained by a different optical technique. Profiling these species for various equivalence ratios provides a further

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20. Abstract (Cont'd):

test for flame modelers.

TABLE OF CONTENTS

I. INTRODUCTION

The hydrogen-nitrous oxide flame is of modeling interest as one of the simplest flames which contains nitrogen chemistry; it is also relevant to our area of interest since nitrous oxide is a common propellant oxidizer. Previously, two experimental studies of species and temperature profiles have been reported. Balakhine, et al.,[1] studied a low pressure (0.05 atm.) lean H_2/N_2O flame with a mass spectrometric technique. Cattolica, et al.,[2] studied an atmospheric pressure stoichiometric flame with laser absorption and fluorescence techniques. We report here, an experimental study of atmospheric pressure H_2/N_2O flames where the equivalence ratio was varied from 0.44 to 1.0. Spontaneous Raman spectroscopy was the primary technique. In addition, laser induced fluorescence was used to obtain relative concentration profiles for OH.

II. EXPERIMENTAL

The apparatus used here has been described in detail previously.[3] A nominal 3W Kr^+ laser operating on the 350.742 nm prism selected UV line is used as the excitation source. The laser cavity is extended with curved mirrors providing an intracavity focus where the burner is placed for study. Scattered light from this focused region is imaged on to a 100 μm entrance slit of a 0.25 m monochromator, dispersed and then detected with an intensified Reticon array. The sampled volume of the focal region approximates a cylinder 3 mm long and 100 μm in diameter. To maximize signal the entrance slit is horizontal; that is, its long axis coincides with the laser beam (or long axis of the cylinder). The resolution of the system is about 12 cm^{-1} full width half maximum, which is sufficient to resolve the vibrational structure of the diatomic molecules.

A commercial sintered bronze flat flame burner (McKenna Products) was used for these flame studies. This type of burner is similar to that used by Cattolica, et al.[2] The 6.0 cm diameter burner head is surrounded by a 7.5 cm diameter shroud through which argon was flowed at a rate of 25 ℓ/min. The reactant gas flow was adjusted to be 15 ℓ/min for all of the equivalence ratios studied. These flows to the burner head are regulated with pressure differences across sintered plugs. The burner is cooled by flowing water through the copper coil imbedded in the sintered plug. Heat extraction is determined from the temperature difference and flow rate of the cooling water.

The N_2 and O_2 calibrations for determining absolute concentration are determined from the ambient room air. For the NO calibration, pure NO gas is flowed through the burner head, as well as argon gas through the shroud. This shroud gas is necessary to minimize formation of NO_2 (which absorbs the laser energy) at the perimeter where the laser beam enters and exits the NO calibration gas. Flames supported by this burner geometry can only be probed to a distance of 0.3 mm from the burner head before the index of refraction gradient deflects the laser beam into the burner head. This deflection also influences the calibration factors for the gases. The collected flame signal has been optimized under these calibration conditions, and since beam position and imaging is slightly different in the flame, a constant factor is applied to all the data of a particular run to correct for the difference. This factor, which typically ranged from 1.2 to 1.3, is determined from a

comparison of the measured N_2 value in the burnt gas region (2-10 mm above the surface) with the calculated value from the NASA-Lewis thermochemical equilibrium code (NLC)[4] with the code temperature constrained to the measured value. In this comparison, to be discussed later, one-half of the measured NO is included in the N_2 value.

All of the number density profiles have been corrected for the partial masking of the collection optics by the edge of the burner at positions closest to the burner head (0-1.5 mm).

There are several reasons for using the 350.742 nm line of a Kr^+ laser rather than the more standard 514.5 or 488.0 nm lines of an Ar^+ laser for the H_2/N_2O flame studies. First, from previous studies we have found that this line pumps OH and NH and thus relative concentration profiles for these species can be measured using this 350.742 nm line. Second, in the reaction zone vicinity, laser induced fluorescence resulting from either the 514.5 or 488.0 nm pump lines can interfere with the Raman signals.

III. RESULTS

It is possible to observe the Raman spectra simultaneously for four molecules with the experimental detection system. A Raman spectrum indicative of this feature is shown in Figure 1. The Stokes Q-branch rotational-vibrational Raman signals for NO, N_2, and O_2 are individually analyzed using an interactive multivariate least squares fitting procedure[5] which incorporates the necessary molecular and experimentally specific information. An analysis of the N_2O Raman signals has not been done. Temperatures obtained from fitting the Raman data for the various molecules are in good agreement, as seen in Table 1. However, since N_2 is present in the largest abundance it is generally used to determine the temperature. Typical temperature fits result in standard deviations of about 1%. Relative concentrations obtained from the fitting are converted to absolute values by comparison to standards. Estimated absolute errors for the temperature measurements are ±3% and ±10% for the concentration measurements of N_2, O_2, and NO.

Temperature and concentration results determined from Raman signals produced in an H_2/N_2O flame are contained in Table 1. Here the burner position is fixed such that the laser beam remains 3 mm above the burner surface for all the measurements. For each equivalence ratio there is the adiabatic flame temperature and a comparison of the temperature values obtained from fitting the N_2, O_2, and NO signals. As the stoichiometric conditions are approached, the concentrations for O_2 and NO decrease to the point where the fitted temperature has a high degree of uncertainty; however, the temperatures obtained from these different data are generally in good agreement, and much lower than the adiabatic value. Assuming that 3 mm above the burner surface is well into the burnt gas region of the flame a comparison of experimental values with the NLC results can be made. Here, the NLC was constrained to the experimental value of the T_{N_2} for each equivalence ratio in order to account for the substantial heat extraction by the burner.

For most adiabatic flames, the final temperature is the highest at or close to the stoichiometric mixture and falls off with either lean or rich mixtures. The temperature as a function of equivalence ratio (Table 1) does

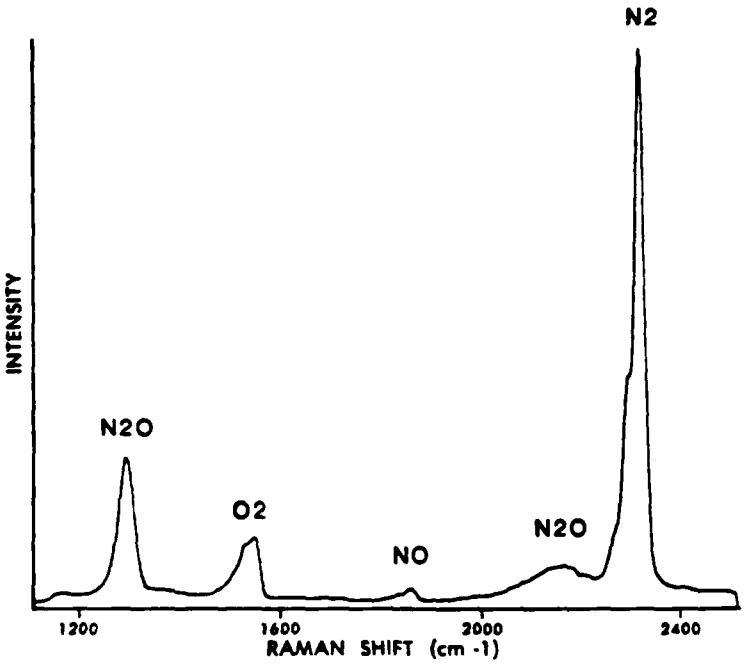

Figure 1. Stokes Q-Branch Raman Spectra of N_2, O_2, NO, and N_2O Occurring in a Hydrogen-Nitrous Oxide Premixed Flame. Laser excitation wavelength is 350.742 nm.

not behave in this manner. Heat extracted from the burner head by the cooling water was measured to vary from 5.0 Kcal/min for the leanest mixture (0.44) to 11.5 Kcal/min for the stoichiometric mixture. An increase in heat extraction indicates the reaction zone of the flame is moving closer to the burner head surface. Thus two opposing effects occur: approaching a stoichiometric mixture increases the flame temperature, but because of the accompanying increase in flame velocity the reaction zone moves closer to the burner surface increasing the heat extraction. The net observed result is that there are similar experimental flame temperatures for all of the equivalence ratios studied.

By assuming all the heat loss is due to conduction to the burner head, a final flame temperature can be calculated.[6] This has been done for all the equivalence ratios reported here, and the value is constant within the accuracy of the measurement, 2350 ± 100 K. This temperature is 450 K ± 100 K higher than the experimentally measured values. At ϕ=1.0, a comparison of heat loss results with those of Cattolica, et al.,[2] can be made since the burner and experimental conditions are identical. We both determine that about 85% of the heat loss can be accounted for by conduction to the burner head. Cattolica argues that the remaining 15% is due to radiation from the burner surface. At the leanest mixture we find about one-half of the heat loss is due to conduction of the burner head. Now the 450 ± 100 K cannot be due entirely to radiation from the burner surface. Here the burner surface temperature must necessarily be significantly lower since 5.0 rather than 11.5 kcal/min are being extracted. Radiative heat loss from the burnt gases, even

TABLE 1. TEMPERATURE AND SPECIES CONCENTRATIONS IN A H_2/N_2O FLAME FOR VARIOUS EQUIVALENCE RATIOS

ϕ	T_{Ad}(NLC)	T_{N_2}	T_{O_2}	T_{NO}	N_2	N_2(NLC)	O_2	O_2(NLC)	NO	NO(NLC)
Equivalence Ratio			K					Mole Per Cent		
0.44	2745	1945	1965	1970	59.6	57.8	14.8	16.0	3.56	0.52
0.45*	2753	1957			59.2	58.0	14.4	16.0	3.46	0.52
0.52	2806	1923	1948	1966	58.3	56.6	12.8	13.4	3.11	0.44
0.60	2855	1898	1919	1959	58.3	55.4	10.7	10.9	2.73	0.37
0.68	2892	1880	1903	1899	55.7	54.2	8.1	8.6	1.99	0.30
0.70*	2900	1886			53.5	54.0	6.6	7.6	1.65	0.29
0.77	2924	1877	1889	1953	50.5	52.9	5.1	5.9	1.93	0.25
0.88	2949	1889	1914	2009	47.9	51.5	2.7	3.1	1.18	0.18
0.89*	2951	1925			50.3	51.2	2.1	2.6	1.38	0.19
1.00	2964	1925	1914		49.2	49.9	0.58	0.09	0.96	0.04

*These values were extracted from the profile measurements shown on Figures 2, 3, and 4 at a burner position of 3 mm.

assuming it is all water vapor, is about an order of magnitude smaller than required. In summary, the heat losses can be accounted for when the gas mixture is close to stoichiometric, but not for the lean mixtures.

The data for N_2, given in Table 1, show only the excursions of the N_2 concentrations about the NLC values. Experimentally determined concentrations for NO are much larger than those calculated from the NLC and the experimental values for O_2 concentration are generally smaller then those of the NLC. The reason for this is that the experimental flame has not attained complete equilibrium. For a better comparison of the experiment with the NLC values, the NO in excess of the NLC values should be apportioned to the equilibrium products. Concentrations of N_2O at equilibrium are negligible in these flames, thus decreasing NO must increase N_2. The oxygen from NO can be apportioned to O_2 so the experimental values of O_2 in Table 1 are justifiably low. The exception to this trend is for ϕ=1.0 where the experimental value for O_2 is much larger than the NLC value. Here, however, the O_2 concentration is so sensitive to the equivalence ratio that uncertainty in the flow of around 2% can by itself produce this difference. Considering the large difference in the experimental and NLC value for the NO concentration, it is obvious that a code containing some detailed chemistry is required to describe this flame system. A direct comparison of experimental results with those of Cattolica, et al.,[2] can be made at ϕ=1.0. The flame temperature (1925 K versus 1950 K) and the NO concentration (0.96% versus 1.1%) are in excellent agreement, as expected.

Profile data for H_2/N_2O flames of equivalence ratios 0.45, 0.70, and 0.89 are displayed on Figures 2, 3, and 4, respectively. In addition to the profiles for T, N_2, O_2, and NO, the OH relative concentration profile has been obtained from laser induced fluorescence measurements using the same laser excitation line, 350.742 nm. The (0,1) $Q_1(19)$ A-X transition of OH is being pumped and the fluorescence emission of the (0,0) band is monitored. The self absorption effects that occur in this experimental arrangement do not affect the OH relative concentration profiles significantly since the distance over which the self absorption occurs is essentially constant both spatially and with respect to OH density for the various burner positions. With a knowledge of the temperature, the fluorescence intensity can be easily converted to relative concentrations through the Boltzmann factor. The only assumption necessary for the validity of the relative concentration profile is a constant quench rate. This assumption is discussed in detail elsewhere.[3]

It was thought that a concentration profile for NH might also be obtained for this flame since we have previously been able to excite NH in a CH_4/N_2O flame operating at 2400K.[3] However, no NH fluorescence was observed. Cattolica, et al.,[2] measure a peak NH concentration of 10^{13} molecules/cm^3 in an H_2/N_2O stoichiometric flame which is also our estimated sensitivity limit for a 2400K flame.[3] The present H_2/N_2O flame temperature is around 500K cooler. Since a high J transition is being pumped, it is not surprising that the NH concentration is below our detection limit. Thus our null result is consistent with the data of Cattolica, et al.[2]

There are some general features that appear in the profile data of Figures 2, 3, and 4. The temperature profiles are much smoother and scatter free than are the concentration profiles, i.e., the temperature parameter which only depends on the spectral shape is substantially more precise than

the concentration profiles which are signal amplitude sensitive and require a calibration. This is especially true for the region which includes part of the reaction zone (0.3 to 2 mm). Here there can be scatter in the data caused by small laser beam deflections. As the laser beam passes from ambient air into the flame zone, a change in the index of refraction occurs due to the temperature change. If this index of refraction gradient is perpendicular to the propagation of the laser beam, no deflection occurs, however, close to the burner surface the flame edge is curved and deflection does occur. This deflection can move the image on the entrance slit of the monochromator, thus reducing the Raman signal. Should the spatial and spectral resolution requirements be relaxed, i.e., larger entrance slits, the steering effect can be made negligible. Line of sight measurements also have compromised spatial resolution because of this effect. At positions closest to the burner surface the flame temperature is in excess of 1700K showing that the early reaction zone is not being probed under these conditions. The flame temperature peaks around 2 mm and the NO, O_2, and N_2 concentrations take on constant values from 2 to 10 mm. Thus, these profiles show that the position 3 mm from the burner surface (data of Table 1) is part of the burnt gas region of the flame where, for the most part, equilibrium conditions exist. The exception is that NO remains above its thermochemical equilibrium value. At positions greater than 4 mm from the burner surface, the rate of decrease in OH concentration slows considerably.

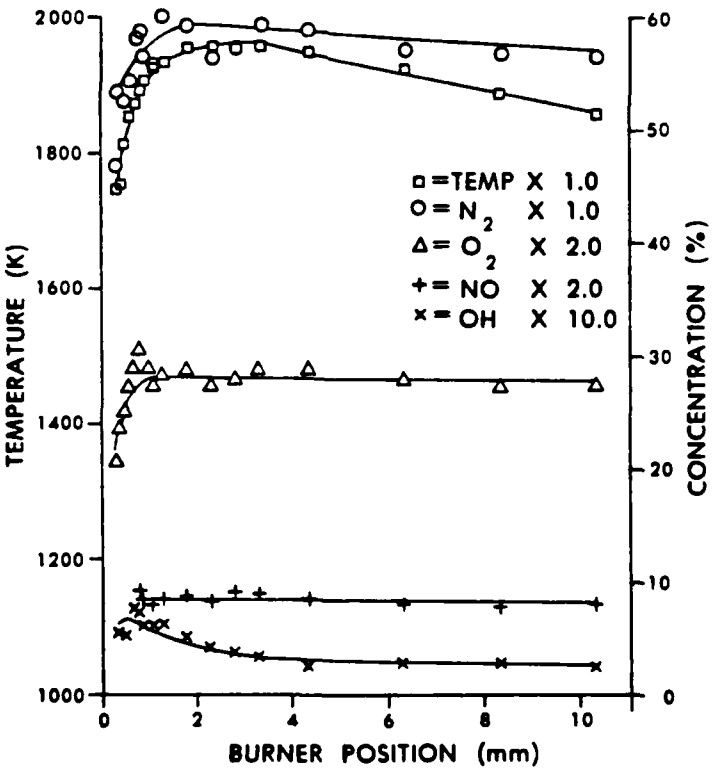

Figure 2. Temperature and Species Concentration Spatial Profiles for N_2, O_2, NO, and OH in Hydrogen-Nitrous Oxide Premixed Flames of Equivalence Ratio of 0.45. The solid lines are drawn in to show general trends and clarify the data points.

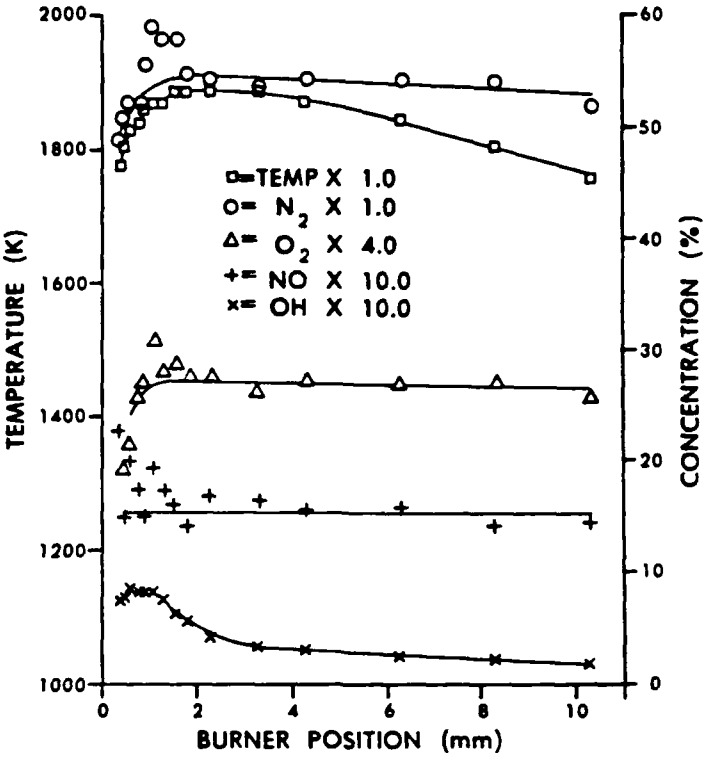

Figure 3. Temperature and Species Concentration Spatial Profiles for N_2, O_2, NO, and OH in Hydrogen-Nitrous Oxide Premixed Flames of Equivalence Ratio of 0.70. The solid lines are drawn in to show general trends and clarify the data points.

Cattolica, et al., find that at positions greater than 6 mm from the burner surface, their measured OH concentrations coincided with the equilibrium values. This information for ϕ=1.0 lends credence to assuming the OH concentration at a distance of 10 mm from the burner surface is in equilibrium (our results). Now the relative OH concentration profiles can be put on an absolute scale by assigning the 10 mm position the equilibrium value (calculated for the measured temperature at this position), and then normalizing the other points. The equilibrium values are 0.15, 0.08, and 0.06 mole percent for ϕ=0.45, 0.70, and 0.89, respectively.

Cattolica, et al., measure a peak OH concentration of 0.26 mole percent for an H_2/N_2O stoichiometric flame at T=1950K. We have determined a peak OH concentration of 0.6 mole percent for an H_2/N_2O flame where ϕ=0.89 and T=1925K. From NLC computations at T=1950K, a change of ϕ from 1.0 to 0.89 increases the OH equilibrium concentration by a factor of 2.3. Use of this correction places our result very close to that of Cattolica, et al. Balakhine, et al., measure a peak OH concentration of 0.06 mole percent for a lean H_2/N_2O flame where ϕ=0.46, T=1930K and P=0.05 atm. This result is about a factor of 7 below the NLC equilibrium value. The NLC OH equilibrium concentrations for a ϕ=0.45 and T=1925K H_2/N_2O flame at atmospheric pressure is 0.24 mole percent, and 0.45 mole percent at p=0.05 atm. Our measured relative OH concentrations for the 0.45 and 0.89 equivalence ratios are

similar (within 20% of each other) and the normalized peak concentration is 0.5 mole percent, close to the 0.89 case. Hence, from our method of normalization, there is an order of magnitude difference in the present results and that for the low pressure flame. Although reasons for this difference are not obvious, possible recombination of OH in the sampling nozzle of the low pressure mass spectrometric experiment would result in low OH values.

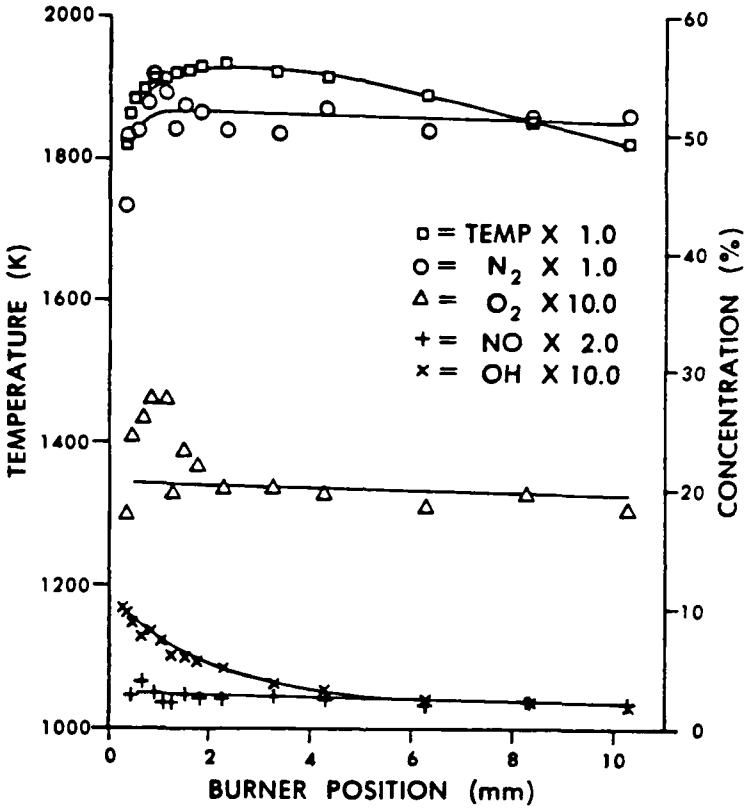

Figure 4. Temperature and Species Concentration Spatial Profiles for N_2, O_2, NO, and OH in Hydrogen-Nitrous Oxide Premixed Flames of Equivalence Ratio of 0.89. The solid lines are drawn in to show general trends and clarify the data points.

IV. SUMMARY

Measured NO concentrations are much higher than the equilibrium values for all the H_2/N_2O flames of the various stoichiometries studied here. These results provide further evidence that detailed chemistry is required to describe this flame system. Both line of sight and spatially resolved optical techniques give similar results for the flame temperature and NO concentration. When comparing the OH concentration of a lean atmospheric pressure H_2/N_2O flame with the same flame operating at low pressure, large differences are observed. Possible pressure effects or the inherent differences of the optical technique with that of mass spectrometric sampling may be responsible.

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