The Application of Spontaneous Vibrational Raman Scattering for Temperature Measurements in High Pressure Laminar Flames

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Introduction

The application of laser-based non-intrusive diagnostic techniques is playing an increasingly important role for a detailed understanding of chemical kinetics, heat, and mass-transport in flames. Compared to mechanical probing, optical measurement techniques do not perturb the flowfields to be observed and offer the possibility of high temporal and spatial resolutions even in extreme hostile environments where mechanical probes are likely to be damaged (Eckbreth, 1996). Spontaneous vibrational Raman scattering is one of the non-intrusive techniques that can provide quantitative simultaneous measurements of temperature and multi-species concentrations in non-sooting atmospheric turbulent flames (Cheng et al., 1992). The feasibility of applying this technique for temperature and multi-species analysis in spray flame and automobile engine has also been reported Grünefeld et al., 1994). However, no quantitative temperature measurement in high pressure flames has been demonstrated and this motivates the present study of temperature measurement in premixed high pressure methane/air flames.

Experimental Apparatus

The schematic diagram of the UV Raman system is shown in Fig. 1. A narrowband KrF excimer laser (Lambda Physik LPX-250T) produces UV light that is tunable from 247.9 to 248.9 nm with a linewidth of 0.003 nm. The maximum pulse energy is 450 mJ with pulse duration of 20 ns. Light scattered by the 2000 mm focusing lens is measured by a PMT to provide a relative measure of the laser pulse energy. The Stokes Raman signal emanating from the sample volume is collected and focused by two 100 mm diameter plano-convex lenses through a 10 mm thick butyl acetate liquid filter and relayed to the entrance slit of a 0.5 m, f/4 spectrometer (SPEX-500M) with a 3600 grooves/mm grating (0.55 nm/mm dispersion). An intensified CCD camera (Princeton Instruments, 576 x 384 array, 22 x 22 µm pixels) is aligned at the exit plane of the spectrometer for monitoring the Stokes Raman signal. The Raman signal is digitized with a 14-bit A/D card connected to a personal computer for data reduction. The high pressure vessel, which includes a McKenna burner (McKenna Products, Inc.), is illustrated in Fig. 2. Premixed methane/air high pressure flat flames are stabilized on a water-cooled sintered stainless steel plate burner of 30 mm active diameter. An annual N2 shielding gas is provided with matched flow velocities for the flames to minimize mixing of fuel gas with surrounding air. The outer burner diameter including the shroud ring is 60 mm. The fuel-to-air mixture and N₂ shielding gas are controlled by electronic mass flowmeters to achieve good stability and reproducibility of the flames over the entire pressure range. The burner is mounted on a translation stage, which allowed a maximum vertical adjustment of 100 mm with respective to the laser beam.

Results and Discussion

To examine the potential of quantitative Raman temperature measurements in flames at elevated pressures, three different equivalence ratios ($\Phi = 0.7, 1.0, 1.2$) of premixed methane/air laminar flames of 1-5 atm are produced. For each flame condition, the air volumetric flowrate is kept at 30 l/min while the methane flowrate is adjusted to match the required equivalence ratio, so that the pressure effects on temperature distributions along the axial direction can be studied. Temperature measurements in high pressure laminar flames are made by spectral-fit of the theoretical N₂ *Q*-branch spectra to the measured spectra. The resolution of the experimental spectrum

must be adequate to separate the N₂ ground vibrational state transitions from the first vibrationally excited state transitions. A 0.055 nm spectral resolution of the experimental system is sufficient to resolve the vibrational state transitions. The theoretical N₂ Q-branch spectrum is produced by assuming a temperature and calculating \mathcal{E}_{e} for each vibrational-rotational transition with the following equation (Long, 1977):

$$S(V,J) \propto \frac{g(2J+1)(V+1)\epsilon_{S}^{4}A_{\Delta J=0,J}}{Q_{ml}Q_{vih}} \exp\left\{\frac{[-G_{0}(V) - F_{V}(J)]hc}{kT}\right\}$$
(1)

The expression for intensity given by Eq. 1 is obtained from Placzek's polarizability theory, which assumes that the laser frequency is far from a molecular resonant frequency. This is not strictly valid for N₂ at 248 nm excitation, with a term value of 69283 cm⁻¹ for the $a^{1}\Pi_{g}$ electronic state. To account for the difference in resonant enhancement of the various energy levels a single resonance theory is used whose intermediate state frequency is modified by the initial energy level of each Raman transition. The A parameter used by this theory is assumed to be independent of the modified intermediate state frequency. The calculated resonant enhancement difference is slight (~15%) between the ground vibrational state and the first excited state transitions but can significantly alter temperature measurements. The calculated N_2 Raman lines are then convoluted by a theoretical slit function to mimic the experimental spectra. Comparison of the experimental and theoretical N₂ *O*-branch spectra is shown in Fig. 3 for stoichiometric flames operated at pressures of 1-5 atm. Measurements are made at 10 mm above the burner surface. Whereas the peaks of vibrational transitions are quite well distinctive at 1 atm, they start to overlap as pressure increases. To simulate the N_2 spectrum at 1 atm, the slit function that is used is symmetrically trapezoidal in shape with a top of 0.01 nm and a base of 0.13 nm. Several different line shapes such as Voigt (Woyde and Stricker, 1990), Gaussian, and combined Gaussian (Attal-Trétout et al., 1990; Kohse-Höinghaus et al., 1990) profiles are tried, but the trapezoidal shape slit function gave best match with the experimental spectrum. The best-fit temperature is 1870 K with an uncertainty of \pm 60 K. For higher pressure flames the slit function used for 1 atm is no longer valid due to coupled effects of pressure and temperature broadening. The top/base widths of the trapezoidal slit functions are 0.03/0.15, 0.04/0.16, 0.05/0.17, and 0.07/0.19 nm and the resulting temperatures are 1760±50, 1680±40, 1650±40, and 1650±40 K for 2-5 atm, respectively. Figure 3 indicates that at a fixed stoichiometry ($\Phi = 1.0$) and exit velocity the flame is pushed toward the burner surface as pressure increases. The flame temperature decreases to about 220 K from 1 atm to 5 atm at 10 mm above the burner surface. The effects of pressure on temperature distribution for $\Phi = 0.7, 1.0, \text{ and } 1.2$ flames are shown in Fig. 4. The symbols represent the Raman measured temperatures and lines for thermocouple temperatures. Thermocouple measurements indicate that temperature decreases gradually with increasing the distance above the burner surface for three flames and all pressure conditions. Raman scattering temperature measurements show the similar trends except for the 1 atm flames, which flat-flame conditions are preserved for the height up to 20 mm. Good agreement is achieved between the thermocouple- and Raman-measured temperatures with a discrepancy of less than 3% of that measured temperatures. At 1 atm, the measured maximum temperatures are 1770, 1870, and 1870 K for $\Phi =$ 0.7, 1.0, and 1.2 flames, respectively. These temperatures are about 50, 340, and 245 K below their respective adiabatic flame temperatures due to insufficient cold gas velocities. The decrease of temperature with increasing pressure is due to higher pressure pushed the flame closer to the burner surface and subsequently cooled the flame temperature by heat loss. For spectral-fit temperature measurement, the trapezoidal slit function is assumed to calculate the synthetic spectrum and to match the measured N₂ spectrum for each pressure condition. The linewidth of the $N_2 Q$ -branch spectrum in flames at a fixed pressure is then determined by the full linewidth at half maximum (FWHM) of the slit function. The results of the line shape analysis as a function of pressure are shown in Fig. 5. The solid line is the corresponding linear fit and it is used to extrapolate the linewidth for higher pressures. The calculated N₂ *Q*-branch spectra for high temperature and high pressure conditions are shown in Fig. 6. Figure 6a simulates the N_2 spectra in flames at T = 2300 K and pressures of 10, 15, 20, 25, and 30 atm. At 10 atm, collision-induced population changes are responsible for the broadening of the whole Q-branch. This leads to a collapse of the spectrum into one broad line. In addition to collisional broadening an appreciable spectral shift of the intensity maximum is observed. The potential of applying spectral-fit method for temperature measurement at pressure up to 30 atm is examined in The calculated spectra indicate that an apparent spectrum difference is observed for flame Fig. 6b. temperatures of 2200, 2300, and 2400 K. This means that the spectral-fit method can be used for measuring a mean flame temperature of 2300 K with an uncertainty of $\pm 4.5\%$ at 30 atm. The accuracy of temperature measurements would increase with decreasing flame temperature and pressure.

Conclusions

Time-averaged vibrational Raman spectra are measured in premixed methane/air flames at pressures up to 5 atm. The objective of this work is to demonstrate the applicability of the spontaneous vibrational Raman scattering technique for temperature measurements in high pressure flames. Spectral-fit of the theoretical N₂ Q-branch spectra to the measured spectra are used to determine the mean flame temperature. For pressure range up to 5 atm, an accuracy of $\pm 3.5\%$ is achieved within the measured flame temperatures. The spectral-fit Raman-measured temperature agrees well with the thermocouple-measured temperature to within $\pm 3\%$ for $\Phi = 0.7$, 1.0, and 1.2 flames at pressures up to 5 atm. Further analysis indicates that the spectral-fit temperature measurement technique can be applied to combustion systems with pressure and temperature as high as 30 atm and 2300 K, which has an uncertainty of $\pm 4.5\%$.

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Fig. 1 Schematic diagram of the UV Raman system.



Fig. 2 High pressure burner.



Fig. 3 Spectral-fit temperature measurements in stoichiometric CH_4/air flames at 1-5 atm and H = 10 mm.



Fig. 4 Temperature distributions along the axial direction of the burner for $\Phi = 0.7$, 1.0, and 1.2

flames. Symbols represent Raman-measured temperatures and lines for thermocouple-measured temperatures.



Fig. 5 Measured linewidths of the N_2 Stokes Q-branch in flames at 1-5 atm.



Fig. 6 Theoretical N_2 Q-branch spectra. (a) For P = 10, 15, 20, 25, and 30 atm at T = 2300 K, (b) For T = 2200, 2300, and 2400 K at P = 30 atm.