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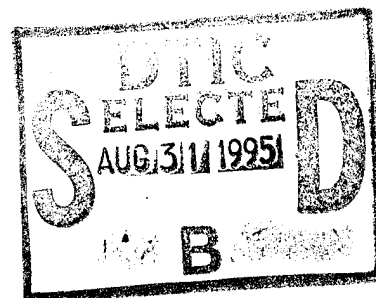


Tomographic Analysis of CO Absorption in Low-Pressure Flame

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1. INTRODUCTION

Absorbances measured using line-of-sight techniques inherently report spectral attenuation integrated along the entire beam path (Limbaugh 1985). At a given frequency, line-of-sight absorbance through a low-pressure combustor system may have contributions from gases at several different densities and temperatures corresponding to the light beam passing through different regions in the low-pressure containment chamber. The degree to which the line-of-sight path is nonhomogeneous is determined by diffusion and flow dynamics within the flame and low-pressure containment chamber. Usually, the region of interest is the flame region where the density of some combustion gases may be less than in cooler portions of the line-of-sight path. This situation is illustrated in Figure 1.

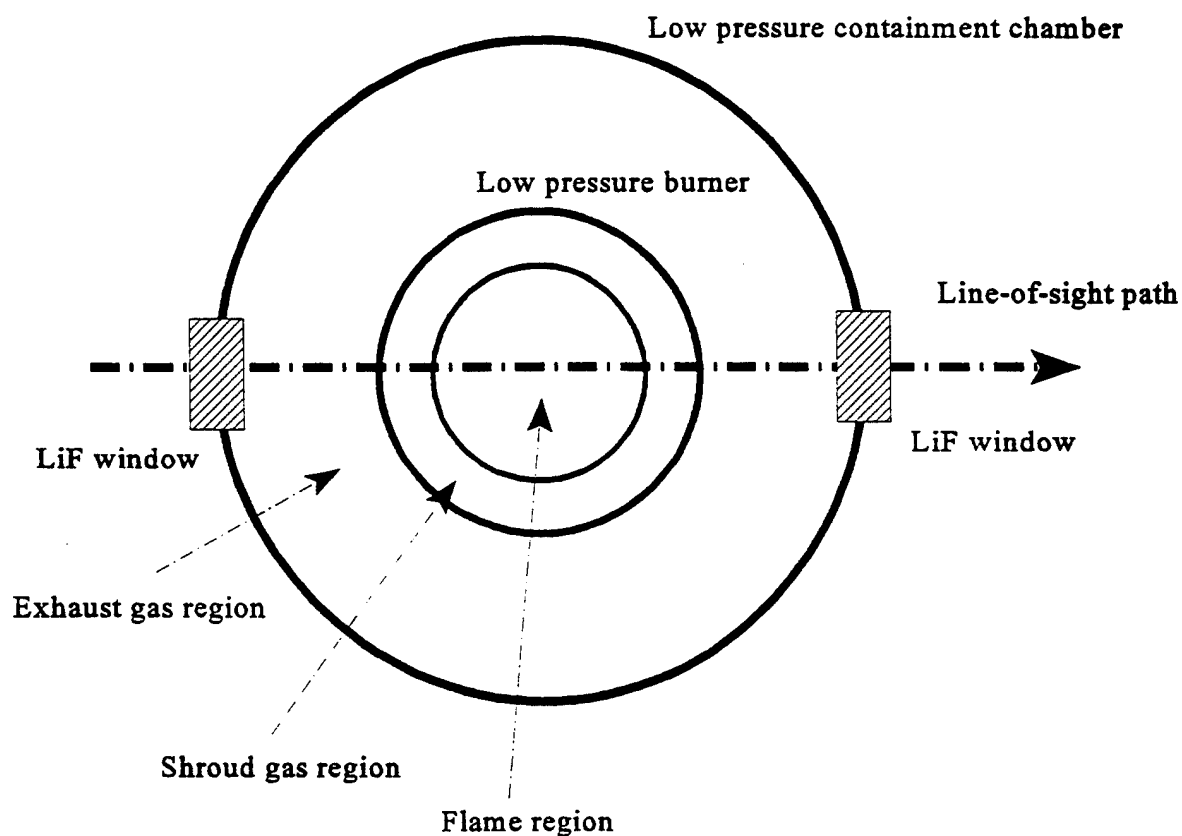


Figure 1. The line-of-sight path through the low-pressure containment chamber as viewed from above the apparatus. Gas densities and temperatures may differ greatly in going from the exhaust gas region into the shroud gas region into the flame region.

Several methods have been developed to minimize contributions to measured absorbance arising from species outside the region of interest of the line of sight. These range from instrumental techniques such as "onion peeling" (Dasch 1992) and tomographic analysis (Cormack 1963), to restricting observation to species or states of species that minimize potential errors or may only exist in the line-of-sight region of interest (Dayton et al. 1993). A common method of measuring temperature using absorbance spectra employs tunable diode laser (TDL)-based, two-line thermometry (Hanson and Falcone 1978; Schoenung and Hanson 1981). In this method, temperature is calculated by fitting the integrated absorbance of two close-lying rovibrational transitions originating from different vibrational states to a Boltzmann distribution.

To minimize errors in temperature measured by two-line thermometry, it is important to minimize the degree of flamespreading, and hence the amount of absorbing gas, beyond the burner surface diameter. This has been shown (Jeffries et al. 1992) to be most easily accomplished by keeping total pressure within the flame chamber above 30 torr. Unfortunately, raising the total pressure causes flame regions to become more compact, limiting spatial resolution. Spectral lines selected for the analysis should have the largest separation of initial energy states possible to maximize temperature sensitivity (Ouyang and Varghese 1990). It is also necessary to maximize the ground state energy of the most intense transition so as to minimize the effect of "cold gas" absorbance.

2. BACKGROUND

In our experiments, we have measured temperatures and species concentration profiles in a 20-torr premixed CH_4/O_2 flame as a function of height above the surface of a flat flame burner. Species concentrations are measured using line-of-sight TDL infrared absorption spectroscopy. Temperatures are measured using TDL-based, two-line thermometry and a fine-wire Pt/Pt-10%Rh thermocouple. Temperature is measured using two complementary methods because of limitations associated with each technique. For thermocouple measurements, the main limitation is that the thermocouple wire must enter the flame, and this perturbation may cause the measured temperature to be in error. For two-line thermometry, the main limitation is that the technique is a line-of-sight measurement in which absorbance is integrated along the entire light path. Additionally, for compact flame zones, the probe laser beam diameter may be large enough to sample regions of high temperature variance.

As an example of the discrepancy that may exist between results obtained by these complementary methods, Figure 2 shows flame temperatures as a function of height above the burner surface for a 20-torr premixed CH_4/O_2 flame measured by two-line thermometry and by a thermocouple. The thermocouple temperatures have been corrected for radiative emission (Sausa et al. 1990). Figure 2 indicates that the temperature as a function of height above the burner surface in a 20-torr premixed CH_4/O_2 flame rises faster when measured using two-line thermometry than when measured using a thermocouple. Conversely, the overall temperature of the same flame is found to be at a higher temperature and the peak temperature at a distance further from the burner surface when measured by a thermocouple than when measured using two-line thermometry. This report addresses the latter discrepancy by using tomographic analysis to provide a correction to line-of-sight spectra for this combusting system.

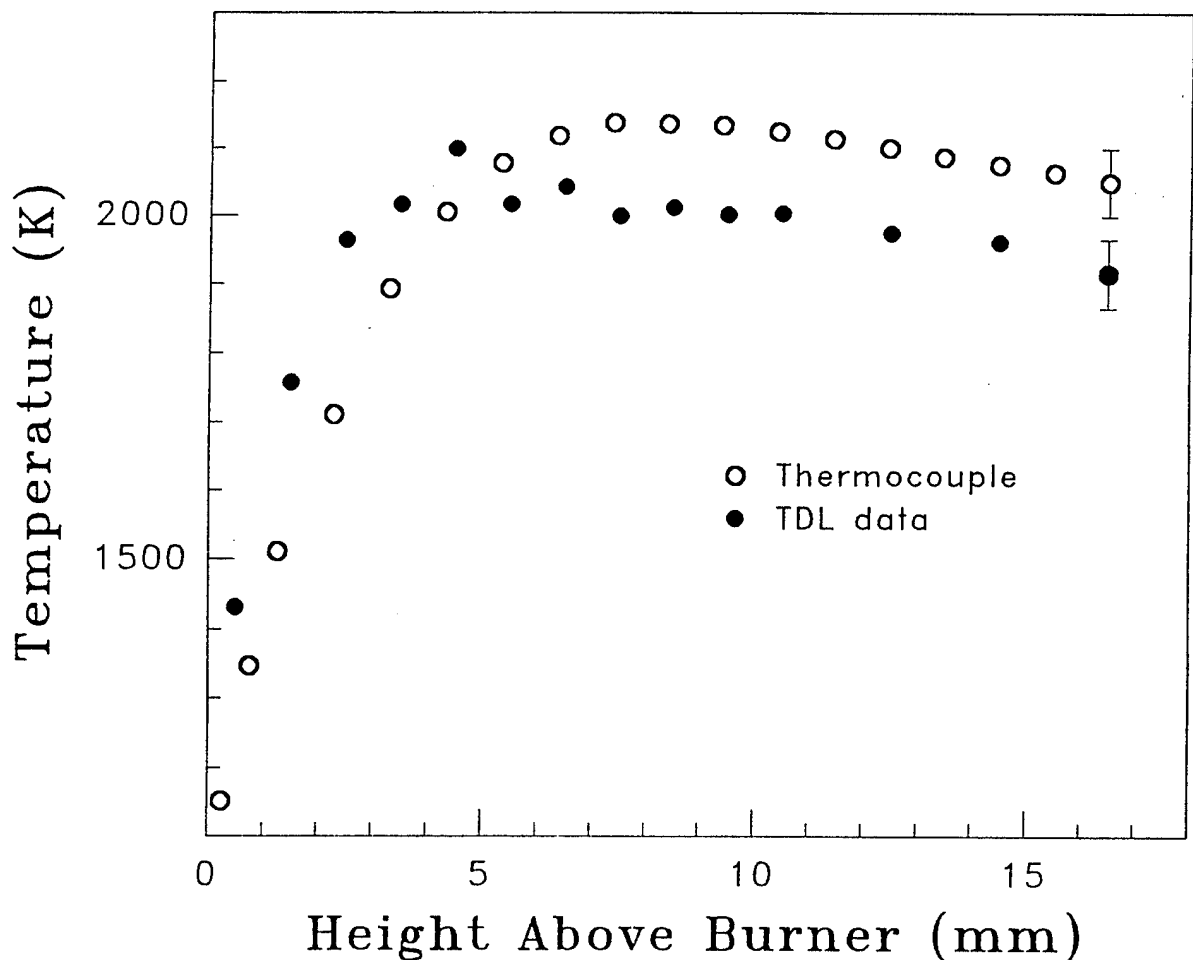


Figure 2. Temperatures measured using two-line thermometry and a Pt/Pt-10%Rh thermocouple show different temperature vs. height above burner surface contours. Thermocouple temperatures have been corrected for emissivity.

3. EXPERIMENTAL

3.1 Apparatus. The experimental apparatus has been extensively described in a previous publication (McNesby and Fifer 1992). Briefly, the apparatus consists of a low-pressure flat flame burner (McKenna Industries, Inc.) mounted on translational stages inside an evacuable chamber. The evacuable chamber is equipped with LiF windows (apertured to 800 μm diameter) to allow passage of infrared laser radiation. Since the infrared laser beam remains fixed in position, different parts of the flame are examined by moving the burner within the chamber relative to the laser beam path.

Laser radiation is provided by a helium-cooled TDL (Laser Photonics, Analytics Division) and detected using liquid-nitrogen-cooled HgCdTe infrared detectors. Laser output is collimated, mode and coarse frequency selected, and chopped prior to entering the low-pressure chamber. Lock-in detection at the chopping frequency effectively discriminates against emission from the flame. Entrance and exit apertures mounted on the evacuable burner chamber restrict the maximum beam diameter through the flame to 800 μm . For concentration and two-line thermometry measurements, the laser was scanned over two pairs of CO transitions (Todd et al. 1976) (P18 $\nu=1\rightarrow 2$ at 2042.9972 cm^{-1} ; P12 $\nu=2\rightarrow 3$ at 2042.8085 cm^{-1} and P13 $\nu=2\rightarrow 3$ at 2038.6249 cm^{-1} ; P19 $\nu=1\rightarrow 2$ at 2038.5822 cm^{-1}). These line pairs were selected because they required the smallest scanning range within the specified output of our laser system ($\sim 2030\text{--}2060 \text{ cm}^{-1}$), because absorption from a vibrationally excited state is minimized in room temperature CO, and because these lines conformed to previously published criteria for two-line thermometry experiments (Ouyang and Varghese 1990). Linearity of the laser frequency versus tuning current was checked using a confocal etalon and also by visual examination of room temperature CO absorption profiles. It was found that for the small tuning range of these experiments ($\sim 0.2 \text{ cm}^{-1}$) no correction was necessary to account for nonlinearity of laser frequency with tuning current. All flames studied were stoichiometric CH_4 (0.95 liters/min)/ O_2 (1.9 liters/min) flames surrounded by an Ar (3 liters/min) shroud. Total pressure within the burner chamber was maintained at 20 torr.

Thermocouple temperature profiles were measured using 100- μm Pt/Pt-10%Rh thermocouples coated with a refractory mixture of yttrium oxide and beryllium oxide (Kent 1970). The refractory coating is necessary primarily to promote radiative cooling of the interior of the thermocouple wire and also to inhibit any catalytic reactions that may occur at the platinum surface. Temperatures were measured with the thermocouple junction at the center of the flame. The thermocouple was withdrawn from the flame

region when line-of-sight spectra were measured. Measured thermocouple temperatures were corrected for radiative heat losses (Sausa et al. 1990).

3.2 Data Analysis. It was assumed that all flames investigated possessed axial symmetry. Scans at equal distance from either side of the flame center showed the flame to be axisymmetric to within the accuracy of the experiment. The data collection scheme was as follows. Starting at a fixed height above the burner surface, a transmission spectrum was measured. The horizontal motion stage was then translated 2 mm, and another scan measured. This process was repeated until 20 parallel transmission spectra had been recorded. The TDL probe beam diameter was 800 μm . The initial scan was through the center of the flame, with each successive scan closer to the perimeter of the flame. Since the burner radius is 30 mm, this data collection scheme provides five scans outside a cylinder defined by the burner circumference. This is necessary to examine contributions to line-of-sight absorbance by CO in regions of the flame that have spread radially beyond the burner surface diameter.

The tomographic analysis of the line-of-sight spectra employed a 3-point Abel inversion that has recently appeared in the literature and is available in spreadsheet form (Dasch 1992), although several other methods were also tried and gave similar results (Deutsch and Beniaminy 1983). Absorbance data were used in all inversion routines. For an axisymmetric flame, it is assumed that at any height above the burner surface, temperatures and species concentrations have only radial dependence. Any species or temperature gradients may then be mapped as a series of concentric circles centered about the burner axis.

Tomographic analysis as employed here uses a series of parallel line-of-sight absorbance spectra at a given height in the flame to obtain the radial dependence of species concentration and temperature at that height. The line-of-sight absorbance at a given frequency for a single absorbing species through an optically thin, axisymmetric medium may be given by (Cormack 1982):

$$g(x) = \frac{\int_0^1 f(r) r dr}{(r^2 - x^2)^{1/2}} \quad (1)$$

where $g(x)$ is the line-of-sight absorbance through the medium at lateral position x , and $f(r)$ is the product of the absorption coefficient and the partial pressure at radial position r within the axisymmetric medium. If more than one species absorbs at this frequency, then $f(r)$ would be a sum of absorption coefficients times partial pressures. The previous equation may be solved for $f(r)$ using the Abel transformation:

$$f(r) = -\frac{1}{\pi} \int_r^1 \frac{g'(x) dx}{(x^2 - r^2)^{1/2}} \quad (2)$$

where $g'(x)$ denotes the derivative with respect to x of the function $g(x)$. Spatial resolution of $f(r)$ is determined by the diameter of the infrared laser beam (800 μm). This method of analysis is shown schematically in Figure 3. Problems and limitations of analyzing line-of-sight data using Abel inversion techniques have been discussed previously in the literature (Best et al. 1991).

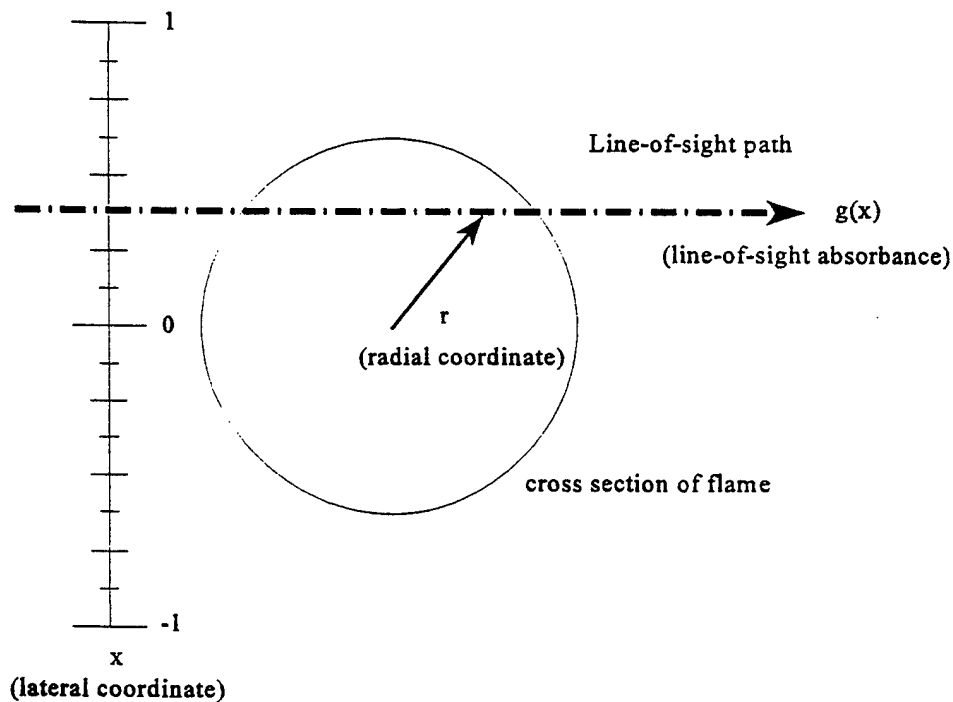


Figure 3. Moving the line-of-sight path to different values of the lateral coordinate, x , allows different line-of-sight absorbances, $g(x)$ to be measured (see text).

The error in the tomographic analysis employed here was estimated by generating synthetic line-of-sight data for a "perfect" flat flame (which would have a uniform radial CO distribution across the burner face and no CO beyond the burner face radius) and then using the 3-point Abel inversion to retrieve the parameters used to originally generate the synthetic data. Tomographic analysis of these synthetic data sets showed a small systematic error for values of localized absorbance at the edge of the flame, near the region of the largest change in localized CO absorbance with radial position. The nature of the error was

to overestimate the species distribution at the flame edge by approximately 5%. However, the Abel inversion technique did not report localized absorbance outside of the synthetic flame region. Also, the effect of absorption of light by CO outside of the optically accessible 40-mm radius was found to have a significant effect on reported localized absorbance within the optically accessible area. Since the absorbance from the hot band transitions used here was vanishingly small outside of the optically accessible region, we do not believe this to be an important source of error. A discussion of errors in tomographic reconstruction is beyond the scope of this report and has been discussed elsewhere for the case of one-dimensional systems (Best et al. 1991). We do believe that it is necessary to examine the effect of the inversion technique on synthetic data that approximates each system to be examined.

For the data analysis employed here, raw transmittance spectra for each scan were rationed to an artificial baseline constructed from nonabsorbing portions of each spectrum. A new baseline was constructed for each individual scan. This procedure was necessary because of the continual drift of the TDL output with time. Normally, compensation for this drift is accomplished by sampling a portion of the laser output with a pellicle beamsplitter and simultaneously collecting a background scan. However, for our apertured line of sight, it was found that the slight beam walk (caused by the grating used to coarse select the laser frequency) across the low-pressure chamber entrance aperture during laser frequency scanning made obtaining a simultaneous output matched background spectrum difficult.

The transmittance spectrum obtained using the synthetic baseline was then converted to absorbance, and each spectral feature fit to a Voigt line shape (which just happens to describe the data well) to obtain an integrable function. Since we are assuming variance of CO temperature and density along the line of sight, no inferences in this are drawn from the values of the Voigt fit parameters. An alternative method was to collect a background spectrum with the flame off at the end of a set of parallel scans. When using this method, it was necessary to employ a baseline correction following conversion to absorbance prior to fitting a Voigt function to the data. Both methods of obtaining absorbance spectra gave similar results. The fitted spectrum was then integrated and the results used as input to the Abel inversion routine. Again, it should be emphasized that for the tomographic analysis, the Voigt function was used only to obtain an integrable function that described the data. An additional reason for using the Voigt function to approximate the observed data is that a fit of both peaks in the measured spectrum results in deconvolution of the line shapes, minimizing integration errors caused by line overlap.

For the two-line thermometry experiments where uniform conditions along the line of sight were implicitly assumed (the "corrected" line-of-sight measurements), CO temperature and concentration were varied in an expression for the Voigt line shape until a best fit to the observed data for the two lines observed in the spectrum was obtained. The areas underneath the individual peaks of the fitted spectrum were used for the final temperature calculation. In this way, both rovibrational and translational manifestations of the temperature were used in the determination (Varghese 1980). Typically, 2,048 data points were collected over a scan range of 0.1 cm^{-1} for an effective resolution (Rayleigh criterion) of approximately 0.0005 cm^{-1} . At 2,000 K, the line width (Guelachvili et al. 1992) (FWHM) for CO rovibrational transitions originating from an excited vibrational state is approximately 0.05 cm^{-1} .

4. RESULTS AND DISCUSSIONS

Figure 4 shows an absorbance spectrum of CO through the center of a 20-torr CH_4/O_2 flame. The line of sight is 5.2 mm above the burner surface using an 800- μm -diameter infrared laser beam. Figure 5 shows the line-of-sight integrated absorbance for CO P19 $v=1 \rightarrow 2$ (the large peak in Figure 4) in the 20-torr CH_4/O_2 flame as a function of the radial distance from the burner center to the line of sight. Spectra were measured at a constant height of 5.2 mm above the burner surface. This figure is a graphical representation of the data input into the Abel inversion routine. Since the edge of the burner surface lies at -30 and $+30$ mm, CO absorbance outside of this region is assumed to be from gas outside of the cylinder defined by the burner circumference. From Figure 5, it is clear that there is significant line-of-sight absorbance for the CO (P19 $v=1 \rightarrow 2$) transition from hot CO gas outside of the region defined by the burner circumference.

Figure 6 shows the results of tomographic analysis of line-of-sight absorbance spectra through a 20-torr CH_4/O_2 flame, with the line of sight contained within a plane 800 μm thick and 4.19 mm above the burner surface. The tomographic analysis indicates that the distribution of population in the first and second excited vibrational states (assuming rotational equilibrium) within the line-of-sight plane is not uniform. From reconstructions using synthetic line-of-sight data, we estimate the uncertainty in the localized CO distribution to be less than 5%. The increase in localized CO density at the edges of the flame was observed for all heights studied.

The ratio of localized integrated absorbance of P13 ($v=2 \rightarrow 3$) to P19 ($v=1 \rightarrow 2$) is also shown in Figure 6. This ratio is nonlinearly proportional to the rovibrational flame temperature at this position in

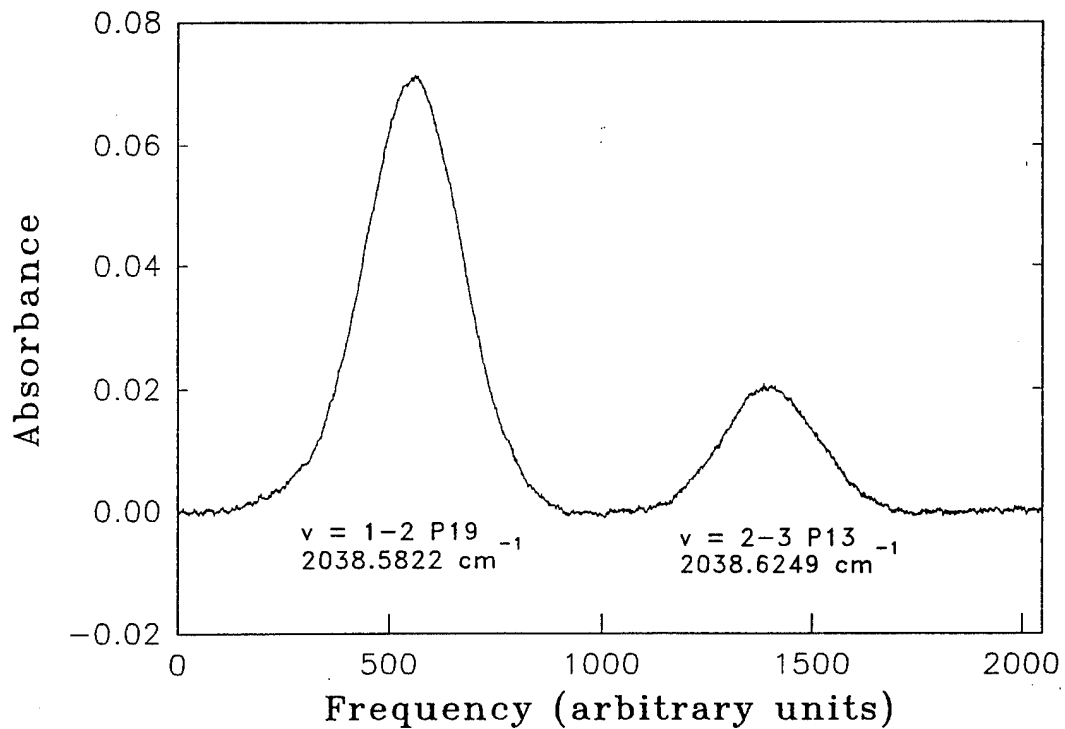


Figure 4. CO hot band absorbance in a 20-torr stoichiometric methane/oxygen flame.

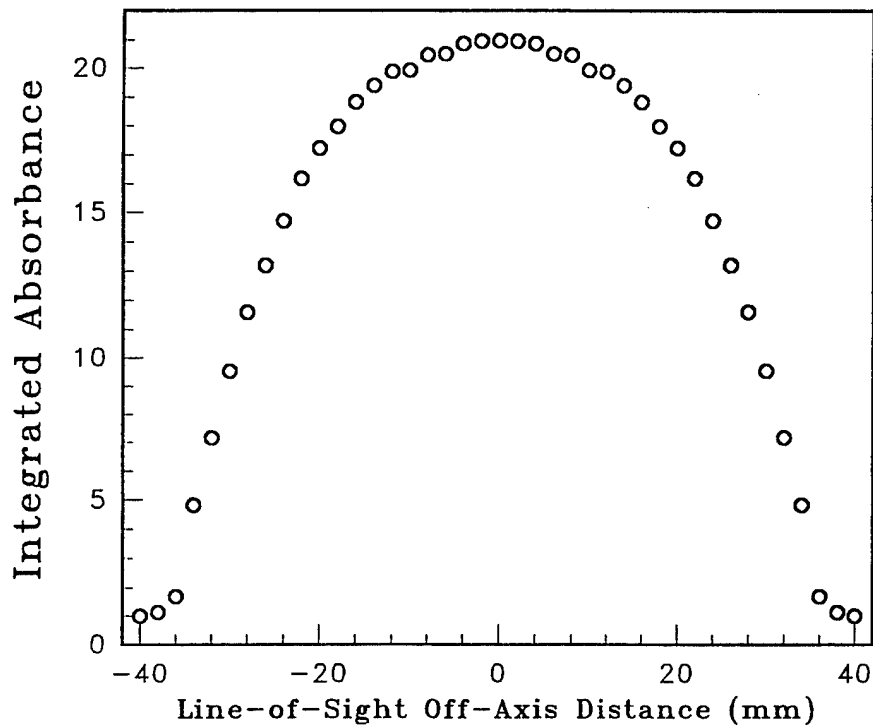


Figure 5. Line-of-sight integrated absorbance for CO P19 ($\nu=1\rightarrow 2$) in a 20-torr stoichiometric methane/oxygen flame as a function of off-axis distance using a laser beam diameter of 0.8 mm. All spectra were measured through a plane lying 5.2 mm above the burner surface. The edge of the burner is at -30 and 30 mm. Data have been reflected about the burner axis.

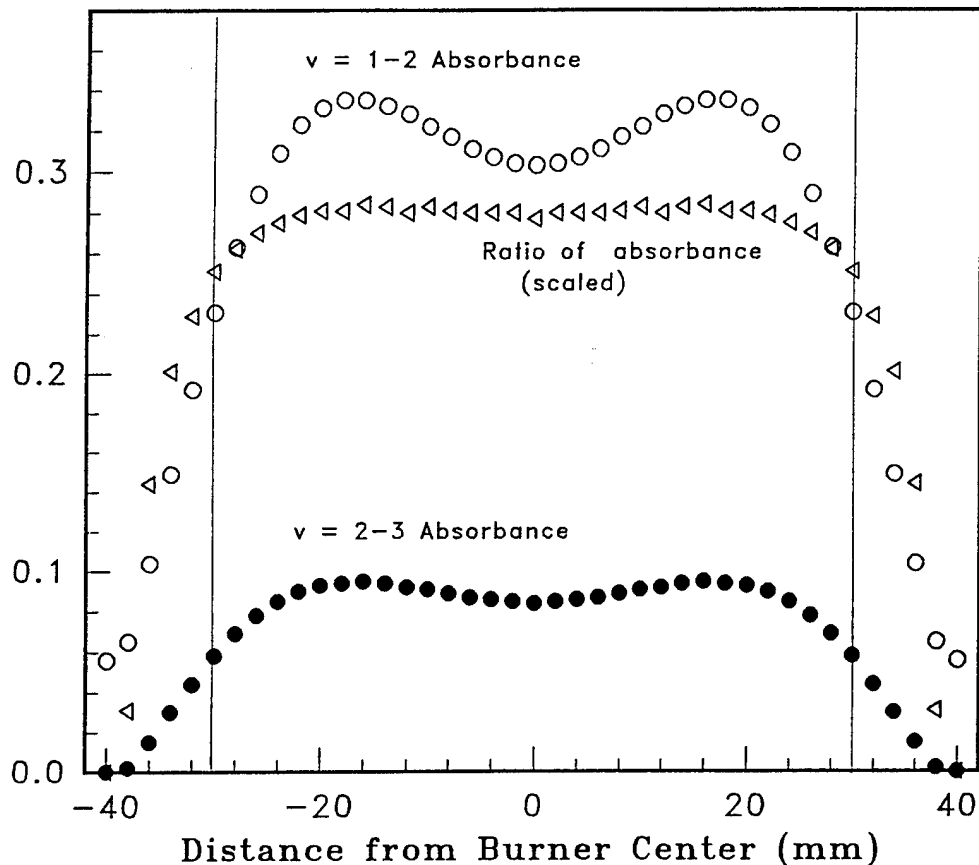


Figure 6. Results of tomographic analysis on line-of-sight CO absorbance spectra lying in a plane 0.8 mm thick and 4.19 mm above the burner surface for a 20-torr stoichiometric methane/oxygen flame. Note that a constant temperature is seen across the burner face even though the populations of P19 ($v=1 \rightarrow 2$) and P13 ($v=2 \rightarrow 3$) are not constant. Data have been reflected about the burner axis.

the flame. It is observed that the temperature in the flame within the line-of-sight plane is fairly constant even though the population of excited states varies with radial position. A conclusion from this figure is that the assumption of uniform concentration or population distribution at a given height in the flame based upon uniform temperature at that height is questionable. Although nonuniformity at a given height across a "flat" flame has been observed previously (Jeffries et al. 1992) for species other than CO, to our knowledge this is the first observation of this phenomenon using a line-of-sight technique. Calculations are currently underway to determine if radial variations in temperatures or radial species are responsible for the radial variation in CO excited state populations.

Finally, from Figure 6, it may be seen that population in the first excited vibrational state of CO extends further outside the flame region than does population in the second excited vibrational state. The

fraction of the area under the curves for localized $\nu=1\rightarrow 2$ and $\nu=2\rightarrow 3$ absorbance in Figure 6 that lies within the flame zone cylinder defined by the burner circumference is 0.88 (± 0.01) and 0.92 (± 0.02), respectively. Figure 7 shows the fraction of line-of-sight $\nu=1\rightarrow 2$ absorbance in a 20-torr CH_4/O_2 flame that lies within the cylinder defined by the burner circumference as a function of height above the burner surface and the best linear fit to the data. The best linear fit within the region defined by the burner diameter (60 mm) is given by

$$y = -0.012x + 0.94 \quad (3)$$

where y is the fraction of $\nu=1\rightarrow 2$ absorbance within the flame region and x is the height in millimeters above the burner surface of the line of sight. The uncertainty in the slope in Equation 3 is ± 0.001 . Figure 8 shows the fraction of line-of-sight $\nu=2\rightarrow 3$ absorbance within the 20-torr premixed CH_4/O_2 flame as a function of height above the burner surface along with the best linear fit to the data. The analogous equation to Equation 3 is

$$y = -0.0085x + 0.97. \quad (4)$$

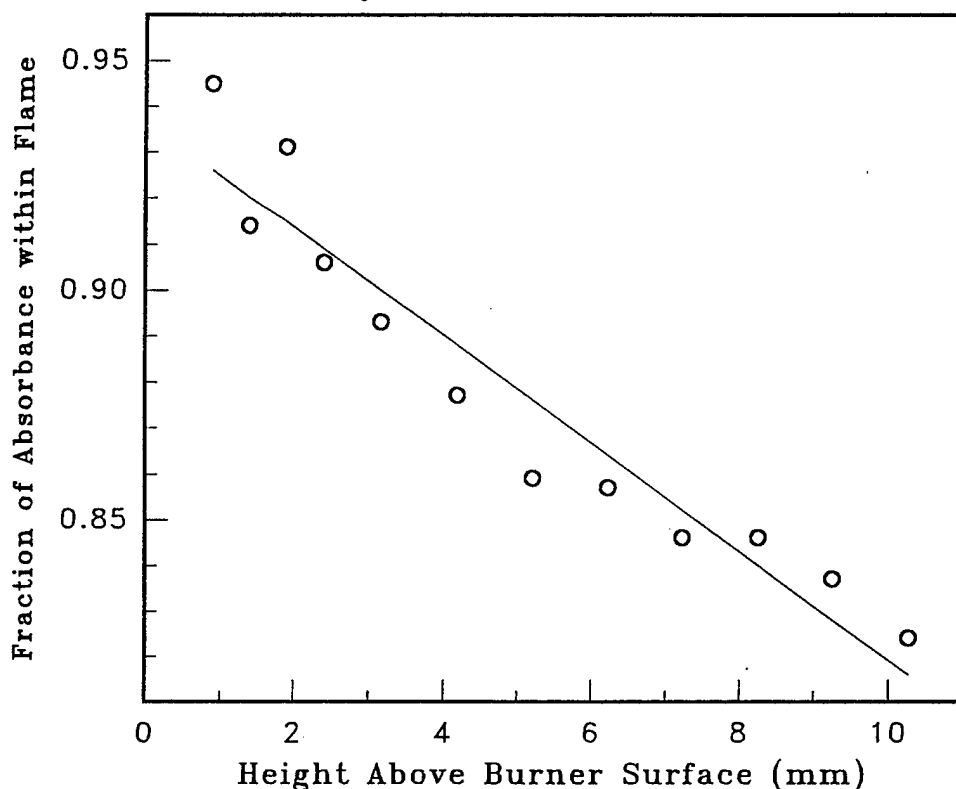


Figure 7. The fraction of CO $\nu=1\rightarrow 2$ absorbance that lies within the flame region as a function of height above the burner surface for a 20-torr stoichiometric methane/oxygen flame.

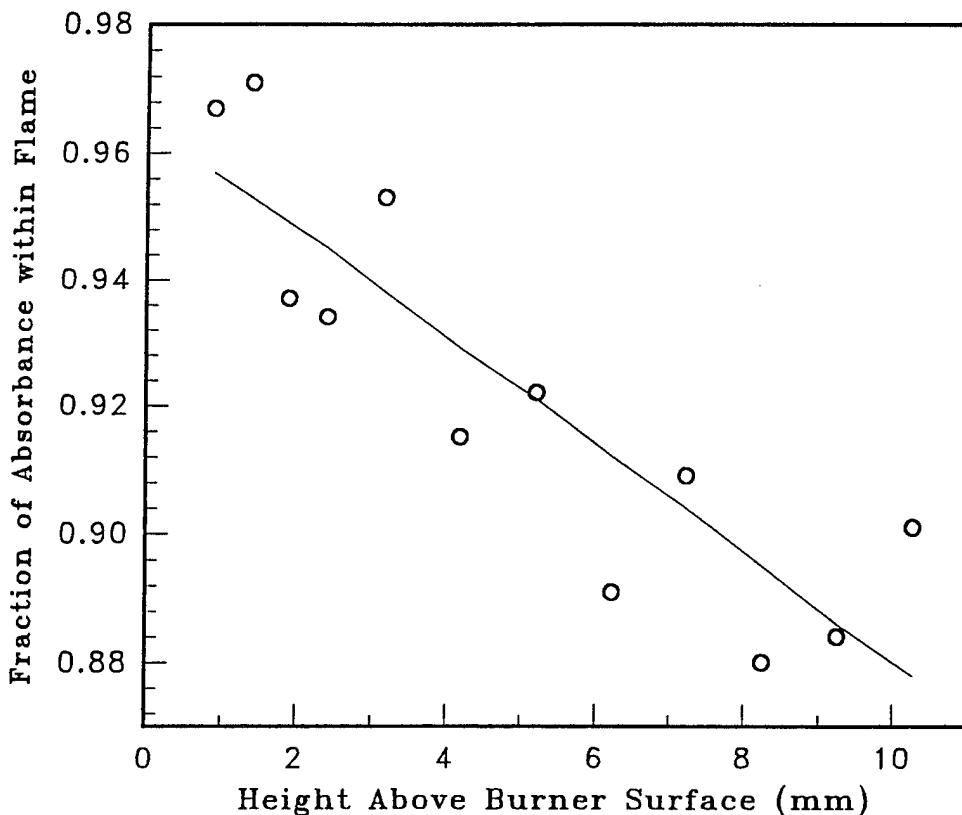


Figure 8. The fraction of CO $v=2 \rightarrow 3$ absorbance that lies within the flame region as a function of height above the burner surface for a 20-torr stoichiometric methane/oxygen flame.

The uncertainty in the slope in Equation 4 is ± 0.0014 . Again, it should be emphasized that this correction applies only to the flame region defined by the burner diameter (60 mm). For line-of-sight spectra over the region studied here (0–10 mm above the burner surface), Equations 3 and 4 indicate that as distance from the burner surface increases, the amount of $v=1 \rightarrow 2$ absorbance that lies outside the flame increases at a faster rate than does the amount of $v=2 \rightarrow 3$ absorbance that lies outside the flame. The effect is that at any height above the burner surface, the observed line-of-sight $v=1 \rightarrow 2$ integrated absorbance is in error to a greater degree than the observed line-of-sight $v=2 \rightarrow 3$ integrated absorbance. For each transition, an excess of integrated absorbance is always observed. Since the temperature is a nonlinear function of the ratio of the integrated absorbance of the $v=2 \rightarrow 3$ transition to the $v=1 \rightarrow 2$ transition (Herzberg 1950), the observed temperature is always too low and the correction to the observed temperature is not directly proportional to the height above the burner surface. This effect has been predicted (Ouyang and Varghese 1989) but has not been demonstrated previously by experiment.

Figure 9 shows the temperature data presented in Figure 2 together with the line-of-sight temperature data to which corrections according to Equations 3 and 4 have been applied. In general, the primary effect of the line-of-sight correction is to shift the uncorrected temperature to a higher value. The secondary effect of the correction (because of the differing values of the slopes in Equations 3 and 4) is to shift the peak temperature further from the burner surface. Also, for the two-line thermometry experiments, sensitivity decreases as the difference in initial energy states for the two transitions ($E''_2 - E''_1$) approaches kT . For the transitions used in these experiments, this situation exists near the peak measured temperature. It is possible that better data would be obtained by repeating the experiments using transitions for which initial states differ by more than one vibrational quantum number. Another possible reason for the remaining discrepancy between corrected two-line thermometry measurements and thermocouple measurements is that the linear fit to the data in Figures 7 and 8 is inappropriate and instead a nonlinear fit to the data should be used. In this case, it would be necessary to do tomographic analysis of line-of-sight spectra over a greater vertical range than has been done here. Still, the net result of the correction is to bring the line-of-sight based temperature measurements into better agreement with thermocouple measurements. Also, it has been suggested that since the final "burned gas" temperatures are in good agreement, it is possible that the discrepancy between thermocouple and optically measured temperatures in the flame region are due to perturbations of the flame chemistry by the thermocouple. Our experiences with thermocouple measurements indicate that this is probably not the case for the experiments reported here. Finally, it is possible that horizontal movement of the burner within the low pressure chamber may disturb the flow of gas outside of the burner region. We have assumed no appreciable effect of burner translation, but have not been able to verify this assumption.

5. CONCLUSION

Two main conclusions may be drawn from this work. First, it has been shown that when using line-of-sight two-line thermometry to examine low-pressure flames, the observed temperature is always too low and the correction that must be applied to the observed temperature is a nonlinear function of the height above the burner surface. This is because there is a radial spread of the flame beyond the burner radius, imparting a 2-D structure to the flame. It has been shown that constant temperature across a flame at a given height in the flame does not imply constant species concentration across the flame at that height. Finally, for low-pressure work (<20-torr total pressure) where two-line thermometry is employed, it is recommended that some type of tomographic analysis of line-of-sight data be performed to obtain a functional relationship between absorbance outside of the flame region and height above the burner surface

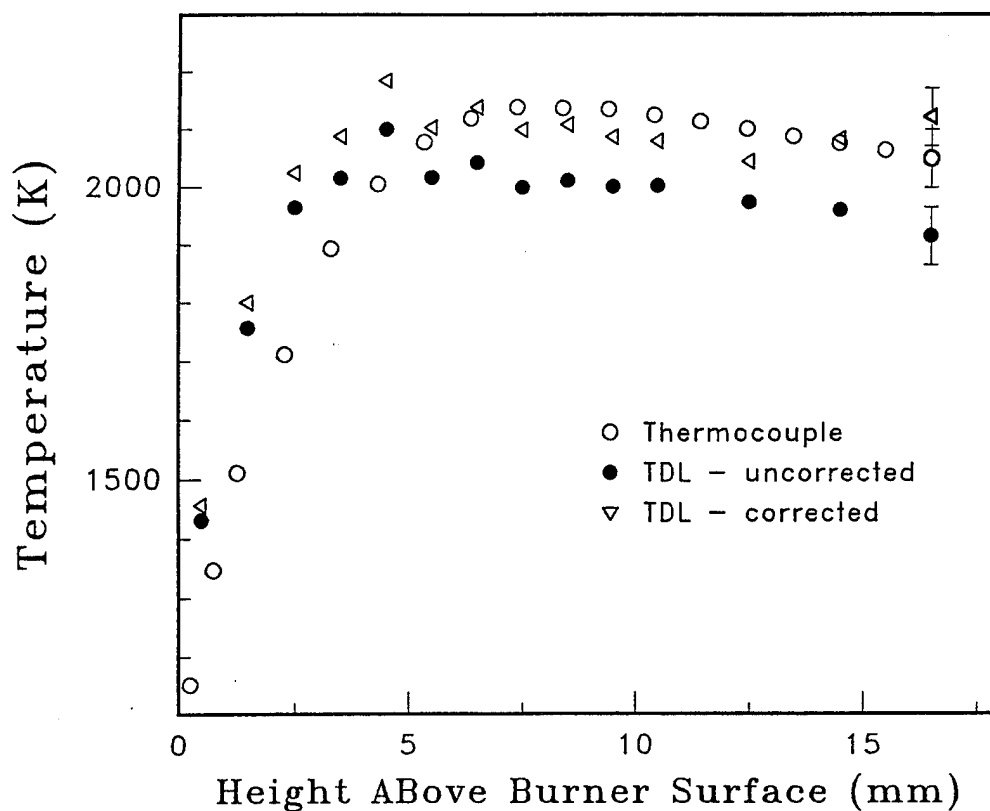


Figure 9. Uncorrected and corrected flame temperatures measured using line-of-sight two-line thermometry and using thermocouples as a function of height above the burner surface for a 20-torr stoichiometric methane/oxygen flame.

for the two transitions involved in the calculation. We believe that as multidimensional detector arrays become more affordable, tomographic analysis will become an important tool for routine investigations of combusting systems.

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