QUANTITATIVE SPECIES MEASUREMENTS IN MICROGRAVITY COMBUSTION FLAMES

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INTRODUCTION

The capability of models and theories to accurately predict and describe the behavior of low gravity flames can only be verified by quantitative measurements. Although video imaging, simple temperature measurements, and velocimetry methods have provided useful information in many cases, there is still a need for quantitative species measurements. Over the past decade, we have been developing high sensitivity optical absorption techniques [1-3] to permit in situ, non-intrusive, absolute concentration measurements for both major and minor flames species using diode lasers [4, 5]. This work has helped to establish wavelength modulation spectroscopy (WMS) as an important method for species detection within the restrictions of microgravity-based measurements.

More recently, in collaboration with Prof. Dahm at the University of Michigan, a new methodology combining computed flame libraries with a single experimental measurement has allowed us to determine the concentration profiles for all species in a flame. This method, termed ITAC (Iterative Temperature with Assumed Chemistry) was demonstrated for a simple laminar non-premixed methane-air flame at both 1-g [6] and at 0-g in a vortex ring flame [7]. In this paper, we report additional normal and microgravity experiments which further confirm the usefulness of this approach.

We also present the development of a new type of laser. This is an external cavity diode laser (ECDL) which has the unique capability of high frequency modulation as well as a very wide tuning range. This will permit the detection of multiple species with one laser while using WMS detection.

EXPERIMENTAL

The drop tower apparatus has been described previously [7, 8]. In brief, a diode laser beam is repetitively rastered across a flame region of interest at 10-20 Hz, with a data collected at a spatial resolution of 0.5 – 1.0 mm. Second harmonic (2f) WMS spectra are recorded and analyzed to obtain temporal and spatial species profiles. In this work, an 1854 nm diode laser was used to detect water vapor using both direct and WMS absorption. The results described here are based primarily on the direct measurements, since the absorbances are sufficiently high (up to 8%) and the more sensitive (but more complicated) WMS analysis was not required. The microgravity measurements were made using the NASA GRC 2.2-sec drop tower. The only difference, from an experimental standpoint, from earlier work is that the Wolfhart-Parker (one dimensional, non-premixed) burner
was modified so as to make the flame twice as long (8-cm) and half as wide so as to maintain the same total gas flow, but improve the absorption path. The spatial resolution was narrowed from 1.0 to 0.5 mm to accommodate this change.

ECDL

Diode lasers have become increasingly important for optical detection of gases. However, a drawback of using diode lasers for gas sensing applications is that they operate over a very limited wavelength range. Typically, only one species can be detected with a given laser. One way in which to expand the output wavelength range of a diode laser is to use an external cavity configuration. A Fabry-Perot (multi-mode) diode laser is combined with a grating and return mirror to create a resonator which, through feedback, couples with the gain region of the diode. The properties of these lasers are that they typically exhibit wider coarse tuning ranges than a simple diode laser, but still show single mode (i.e., single frequency) output. With such a configuration, multiple species detection is possible. However, external cavity diode lasers (ECDLs), in general, cannot be wavelength modulated more than a few kHz (limited by mechanical tuning of an optical element to vary wavelength; diode current tuning no longer is effective). This inability to provide rapid wavelength modulation at higher frequencies results in only limited achievable sensitivity.

The ECDL developed in this research relies on a novel cavity design (Patent Pending) that overcomes the low modulation frequency limitations of present external cavity lasers. This ECDL is wavelength modulated with diode injection current, just like a regular diode laser. Furthermore, in contrast to present external cavity laser designs, the design is simple, inexpensive to implement and rugged. Our design combines the stability and tunability of an ECDL with the wavelength agility of a diode laser. The wide availability of Fabry-Perot diode lasers permits the construction of an ECDL at almost any wavelength from 680 nm to 2.3 μm, with a tuning range exceeding 25 nm (50 times that of a diode laser). As a result, strong absorption bands of virtually all major and minor flame species of interest are now accessible.

RESULTS

Flame Species Measurements – 1g

In a previous paper [6], we presented a comparison of ITAC-derived methane profiles in the non-premixed flame compared with directly measured profiles (using the observed methane absorbance coupled with thermocouple readings). In Fig. 1, we add the ITAC-derived methane profile using only the thermocouple measurements as input to the ITAC computation. While the center concentrations is in good agreement with the other approaches, the wings are wider. The discrepancy may be due to how the separation between the noisy baselines in the spectra and

Figure 1 - Methane profile in non-premixed flame.
methane absorption spectra are defined, as well as the sensitive dependence of the absorption cross section on temperature for the specific absorption line used. We do not believe that radiative heat loss effects are the source, since preliminary calculations for this flame show them to be relatively weak [10]. The estimated accuracy of the thermocouple measurements in the region of interest is ±50°C. We are currently analyzing the water vapor measurements to clarify this issue and expand the results to both the rich and lean sides of the flame.

**Flame Species Measurements – 0g**

The use of the Wolfhard-Parker burner for microgravity studies is relatively new. As shown in Fig. 2, the transition from normal to zero gravity is striking. The flame front (dark regions in the photos) expands and greatly diminishes in height as gravity is reduced. As a result, this should make the observation of intermediates such as OH and C₂H₂ easier to observe. Direct absorption measurements of water vapor across this flame were made at zero gravity. At this time, the results are still undergoing analysis and will be presented at the Workshop.

**External Cavity Diode Laser Development**

Broadband optical sources are useful for performing survey spectroscopy of unique samples and combinations of samples. This type of survey is indispensable when determining interferences between a gas to be detected and other contaminant gases present in a sample. These surveys are typically done with an FTIR or equivalent direct absorption-based instruments. However, these instruments are bulky and prohibitively expensive.

An additional feature of our ECDL laser is that it can be configured to simultaneously put out wavelengths over a wide spectral band, where direct absorption survey spectra can be obtained with a scanning monochromator. The ECDL causes the longitudinal mode spacing to narrow such that a typical monochromator cannot resolve them. Thus, to the monochromator, the ECDL appears as a white light source. However, the ECDL retains the excellent spatial propagation properties of a laser. An illustration of this feature is the recording of the entire 1.53 μm band of acetylene in Fig. 3.
For WMS detection, the laser must be capable of high frequency (> 10 kHz) tuning over wavelength ranges commensurate with the absorption line width. DFB lasers tune continuously with injection current; in that manner it could be considered to be an analog tuning device. The ECDL, on the other hand, tunes in discrete jumps. There is a continuing change in the laser power within one jump, but the wavelength is static to first order. This manner of tuning is more digital in nature. It should be emphasized that continuously tunable lasers are often operated with modulation waveforms that, in effect, cause discrete tuning behavior. An example of such a discrete modulation waveform is the square or modified square waveform. Thus, there is no inherent drawback with a digitally tuned laser. An example of a $2f$ (10 kHz)spectrum is shown for HCN in Fig. 4.

With the stand-alone WMS technique, noise equivalent absorbances of $1 \times 10^{-4}$ have been obtained. This is about an order of magnitude lower performance than for typical WMS implementations. An electronic spectrum analyzer was used to determine the limiting noise source in the 1535 nm ECDL. The limiting noise source was not an etalon, as is usually the case, but second harmonic distortion. This is not uncommon with Fabry-Perot diode lasers and is also present in distributed feedback devices. Since second harmonic distortion is a feature of the laser output and not due to the optical set-up, it can be suppressed by incorporation of a ‘noise canceller’ dual beam subtraction after the design of Haller and Hobbs [9].

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REFERENCES