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This research is concerned with the development of a technique for quantitative, minor-species concentration, time-series measurements in turbulent flames. Past work has demonstrated the capability of picosecond time-resolved laser-induced fluorescence (PITLIF) to obtain sufficient signal with present laser technology to achieve this goal. The specific aim of this AASERT project is to refine the capabilities of the PITLIF instrument. The last progress report detailed the numerical modeling of the PITLIF instrument and feasibility studies for a new data-collection system. During the past year, this new instrumentation, termed LIFTIME, has been installed and tested. As demonstrated here, LIFTIME is capable of making rapid and continuous fluorescence-lifetime measurements as required for quantitative measurements of concentration. The effectiveness of the technique was verified by applying LIFTIME to two known-fluorescence liquids for which the fluorescence lifetime was monitored up to a sampling rate of 500 Hz with less than 10 percent uncertainty. LIFTIME was also tested by measuring hydroxyl concentrations in nine laminar flames. The results are shown to agree well with both previous measurements and predictions.

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(AASERT96) RESPONSE ENHANCEMENT FOR PITLIF INSTRUMENT

AFOSR Grant Number F49620-95-1-0272

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SUMMARY/OVERVIEW

This research is concerned with the development of a technique for quantitative, minor-species concentration, time-series measurements in turbulent flames. Past work has demonstrated the capability of picosecond time-resolved laser-induced fluorescence (PITLIF) to obtain sufficient signal with present laser technology to achieve this goal. The specific aim of this AASERT project is to refine the capabilities of the PITLIF instrument. The last progress report detailed the numerical modeling of the PITLIF instrument and feasibility studies for a new data-collection system. During the past year, this new instrumentation, termed LIFTIME, has been installed and tested. As demonstrated here, LIFTIME is capable of making rapid and continuous fluorescence-lifetime measurements as required for quantitative measurements of concentration. The effectiveness of the technique was verified by applying LIFTIME to two known-fluorescence liquids for which the fluorescence lifetime was monitored up to a sampling rate of 500 Hz with less than 10 percent uncertainty. LIFTIME was also tested by measuring hydroxyl concentrations in nine laminar flames. The results are shown to agree well with both previous measurements and predictions.

TECHNICAL DISCUSSION

Quantitative measurements of radical concentrations in turbulent flames are required for an understanding of important interactions between fluid mixing and chemical reactions. Laser-induced fluorescence (LIF) possesses the spatial and temporal resolution necessary for monitoring radical concentrations in reacting flows; however, LIF suffers from an inverse dependence on the local quenching rate coefficient. Although laser-saturated fluorescence (LSF) permits measurements of species concentrations without recourse to quenching calculations[1], the LSF technique requires large laser powers which forces the laser repetition rate to be on the order of 10 Hz. This repetition rate can provide mean concentrations and higher moments but is too slow to resolve frequency information about the species fluctuations in turbulent flames. High-repetition-rate lasers can be utilized for measuring time series but the quenching rate coefficient must be obtained simultaneously.

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[1] J. R. Reisel, C. D. Carter, and N.M. Laurendeau, *Energy & Fuels* **11**,1092 (1997).

Picosecond time-resolved laser-induced fluorescence (PITLIF) has previously demonstrated the capability to monitor fluorescence signal fluctuations on the time scale of turbulence[2]. PITLIF is a linear LIF technique which uses a high-repetition rate laser (80 MHz) to enable rapid sampling of the resulting fluorescence signal. The short duration laser pulse (1.5-18 ps) also permits direct measurement of the quenching rate coefficient. However, these measurements have not yet been performed simultaneously. An improvement to PITLIF labeled the laser-induced fluorescence triple-integration method (LIFTIME) has been recently developed. LIFTIME permits calculation of the electronic quenching rate coefficient at speeds approaching that needed for turbulence and allows on-the-fly quenching corrections and thus quantitative measurements of species number densities.

LIFTIME was modeled after the rapid lifetime determination (RLD) method[3] which determines the fluorescence lifetime from the ratio of two integrated areas under the fluorescence decay. This method requires only simple algebraic equations and has the advantage of being very fast. However, the RLD method cannot be used without significant errors given a unknown signal background. For many applications (such as liquid lifetime studies) the background may be negligible. However, a large flame emission background is often present in flame environments.

LIFTIME improves upon the RLD method by adding a third integrated bin capable of directly monitoring the background. Figure 1 depicts the three-bin arrangement for LIFTIME for which the fluorescence lifetime can be computed by

$$\tau = \frac{\Delta t}{\ln\left[(D_2 - D_3)/(D_3 - D_4)\right]} \qquad (1)$$

where τ is the lifetime and Δt is the width of each bin (D_2,D_3, and D_4 as defined in Fig. 1).

Based on the past year's success with numerical simulations of LIFTIME, a photon counting system was built to implement LIFTIME experimentally. Briefly, a photomultiplier tube (PMT) monitors the fluorescence signal and a discriminator converts analog, single-photon pulses from the PMT to logic pulses. A second discriminator gates these logic pulses to achieve the three temporal bins as depicted in Fig. 1. A series of parallel pulse counters then integrate the total counts in each bin (D_1-D_4) for a specified period of time controlled by software. The wire schematic for the photon counting system and complete experimental details were presented in detail recently[4].

Two fluorescence mediums, diphenyloxazole (PPO) and quinine sulfate monohydrate (QSM), were employed to calibrate and test LIFTIME. PPO has a fluorescence lifetime of 1.28 ns and the QSM lifetime can be varied via diluted salt solutions. Seven solutions of QSM were created with lifetimes ranging from 1.0 to 3.0 ns. The LIFTIME measurements were compared to independent measurements using a convolute-and-compare technique previously used with PITLIF[5]. The excitation wavelength was 309 nm and the detection bandwidth was 435-445 nm. Figure 2 is the resulting calibration plot for LIFTIME as expressed with respect to the convolute-and-compare measurements. As predicted by simulations, the instrumentation causes the measured decay constant to be slightly larger than the actual decay constant (~8%). Nevertheless, a linear fit can be constructed which effectively corrects for the slight instrumentation bias.

An important and certainly expected feature of LIFTIME is that its uncertainty rises with an increase in sampling rate. However, fluorescence lifetimes can still be determined with less

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[2] M. W. Renfro, S. D. Pack, G. B. King, N. M. Laurendeau, *Combust. Flame* **115**, 443 (1998).

[3] R. M. Ballew and J. N. Demas, *Anal. Chem.* **61**, 30 (1989).

[4] S. D. Pack, M. W. Renfro, G. B. King, N. M. Laurendeau, *Opt. Let.* **23**, 1215 (1998).

[5] T. A. Reichardt, M. S. Klassen, G. B. King, and N. M. Laurendeau, *App. Opt.* **35**, 2125 (1996).

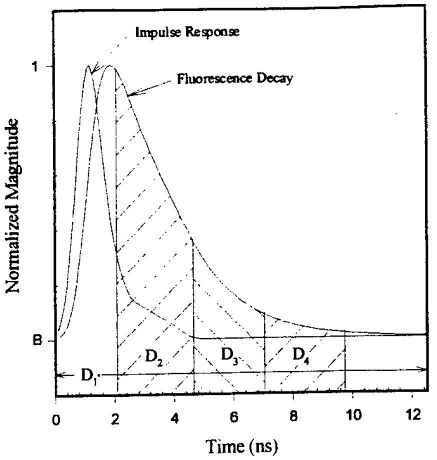

Fig. 1. Graphical representation of the LIFTIME method. D_1 is 12.5 ns wide, whereas D_2, D_3, and D_4 are fixed at 3.5 ns. The dashed line is the impulse response of the PITLIF system.

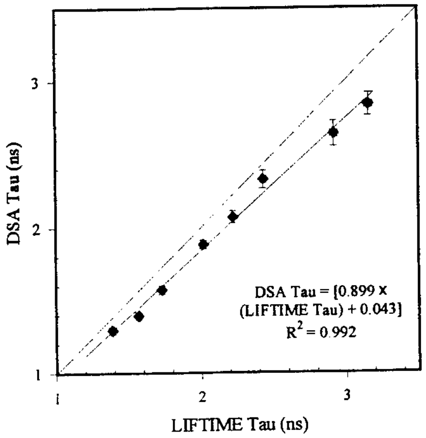

Fig. 2. Calibration of LIFTIME with independent convolute-and-compare measurements. The error bars represent the 95 percent confidence interval for repeatability.

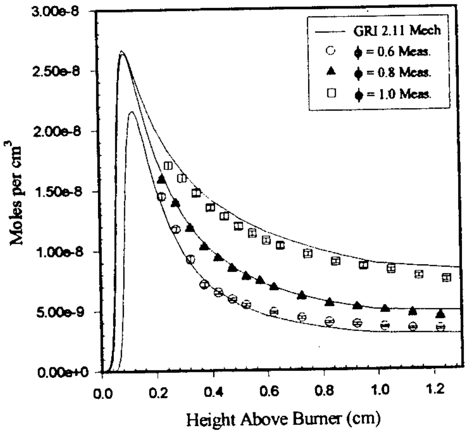

Fig. 3. Measured OH concentrations for three laminar premixed methane flames. Also shown are computations using the GRI 2.11 mechanism[6].

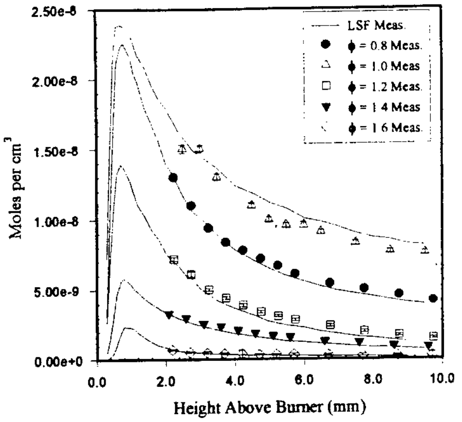

Fig. 4. Measured OH concentrations for five laminar premixed ethane flames. Also shown are previous LSF measurements[1].

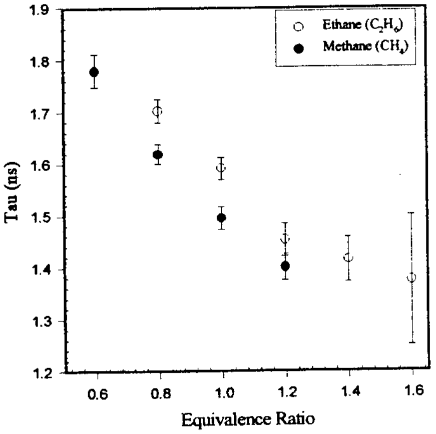

Fig. 5. Fluorescence lifetime versus equivalence ratio in the methane and ethane premixed flames. Error bars represent the 95% confidence interval of the mean.

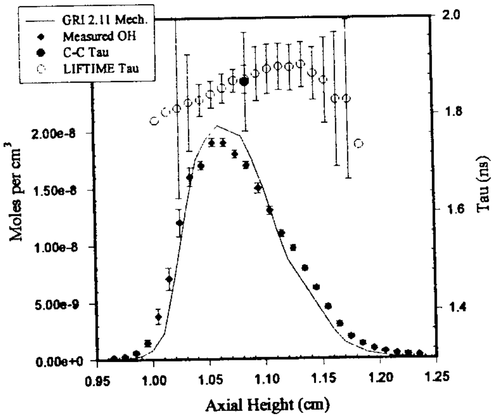

Fig. 6. Measured [OH] and fluorescence lifetime in the counterflow diffusion flame. Error bars represent the 95% confidence interval of the mean. Also shown are predictions using the GRI 2.11 mechanism[6]. C-C is an independently determined convolute-and-compare lifetime to confirm the accuracy of LIFTIME.

Personnel, Publications, and Technology Transfers

Professors Galen B. King and Normand M. Laurendeau are co-principal investigators for this research program. Mr. Michael W. Renfro is a continuing Ph.D. student on the project. Mr. Spencer D. Pack completed the requirements for his M.S. degree in June and is now working at Rolls-Royce Allison Gas Turbine.

Publications and presentations related to the current project are listed below.

(1) M. W. Renfro, S. D. Pack, G. B. King, and N. M. Laurendeau, "Hydroxyl Time-Series Measurements in Laminar and Moderately Turbulent Methane/Air Diffusion Flames," *Combustion and Flame* **115**, 443-455 (1998).

(2) S. D. Pack, M. W. Renfro, G. B. King, and N. M. Laurendeau, "Photon-Counting Technique for Rapid Fluorescence Decay Measurements," *Optics Letters* **23**, 1215-1217 (1998).

(3) M. W. Renfro, Y. R. Sivathanu, J. P. Gore, G. B. King, and N. M. Laurendeau, "Time-Series Analysis and Measurements of Intermediate Species Concentration Spectra in Turbulent Nonpremixed Flames," Central States Section Meeting, The Combustion Institute, Lexington, KY (1998).

(4) S. D. Pack, M. W. Renfro, G. B. King, and N. M. Laurendeau, "Photon-Counting Technique for Rapid Fluorescence Decay Measurements," Central States Section Meeting, The Combustion Institute, Lexington, KY (1998).

(5) M. W. Renfro, S. D. Pack, G. B. King, and N. M. Laurendeau, "Quantitative Hydroxyl-Concentration Time-Series Measurements in Turbulent Nonpremixed Flames," Poster Session, Third International Workshop on Turbulent Nonpremixed Flames, Boulder, CO (1998).

(6) M. W. Renfro, Y. R. Sivathanu, J. P. Gore, G. B. King, and N. M. Laurendeau, "Time-Series Analysis and Measurements of Intermediate Species Concentration Spectra in Turbulent Nonpremixed Flames," Twenty-Seventh International Symposium on Combustion, Boulder, CO (1998).

(7) M. W. Renfro, S. D. Pack, G. B. King, and N. M. Laurendeau, "A New Photon Counting Technique for Rapid, Quantitative Hydroxyl-Concentration Time-Series Measurements," Poster Session, Twenty-Seventh International Symposium on Combustion, Boulder, CO (1998).

We are presently completing an internet web site to make available each of the numerical algorithms developed under this project. The convolute-and-compare algorithm and the pulse pileup correction, which we are presently addressing, are not specific to the PITLIF measurements and have potential applications in other fields. The photon-counting-system design is also applicable to other studies and its design will be made availabe on the same web site.