Femtosecond Laser Electronic Excitation Tagging, FLEET, for Combustion and Flow

Trent J. Murray, Jordan Fisher, Mikhail Slipchenko, and Terrence R. Meyer Purdue University, *IEEE*

Abstract— Femtosecond Laser Electronic Excitation Tagging, known as FLEET, can be used to measure the nitrogen gas content within a gaseous mixture. FLEET does not require any trace particles that could affect the combustion reaction or physical properties of the flow. Another advantage is the simple experimental implementation. In this work a 120-femtosecond laser pulse was focused in to the probe volume to dissociate the nitrogen gas via multiphoton process. The intensity of the light emitted after the recombination is proportional to the nitrogen gas to oxygen mass ratio as the dissociated nitrogen bonds with oxygen to form nitric oxide, which does not emit light. Intensity of the light from FLEET within a methane-air diffusion flame was used to determine fuel/air ratio.. The intensity of FLEET signal was calibrated for different mixtures in test cell in the ranges of pressures to simulate the change of number density due to increase of the temperature in the flame.

I. INTRODUCTION

This experiment utilizes the laser technique FLEET, femtosecond laser electronic excitation tagging, to dissociate nitrogen gas, N_2, with a femtosecond laser pulse and then measuring variables of the emission. FLEET is a nonintrusive laser diagnostic technique that does not rely on any tracer particles; FLEET measures the N_2 gas, a native component of the reaction. The importance of FLEET being the least intrusive measurement technique is greatly valued in areas such as combustion where seed particles may influence measurements.

Planar Laser induced Fluorescence, PLIF, is commonly used as a method of temperature measurements and often used to find equivalence ratio in a plane. While this method is often very useful it often relies on seeded particles which may affect the measurements. The source of the fluorescence in PLIF is different to the source of emission from FLEET. The emission of light in FLEET comes from the nitrogen gas forming covalent bonds after dissociation from a femtosecond laser pulse. The primary benefit of FLEET is its simplicity; FLEET only requires one intensified camera and the emission of FLEET contains multiple wavelengths that decay at different rates. This allows for the use of different filters for different environments such as combustion. While FLEET hasn't been widely utilized in combustion reactions, the FLEET emission contains peak wavelengths from 350 nm to 600 nm, as shown in Figure 1, allowing the use of filters to eliminate noise.

The goal of this study is to link the intensity or time decay of a FLEET of Laser Induced Breakdown Spectroscopy (LIBS) emission signal to equivalence ratio. Equivalence ratio is perhaps one of the most important characteristics of a flame. This ratio is extremely crucial for determining thrust, emissions and efficiency. The fuel air mixture is very difficult to measure while in a combustion reaction except when using a spectrometer. However, a spectrometer is limited to the speed of a camera, Photo Multiplier Tube (PMT) is not limited by frames per second. This allows the user to accurately pinpoint where the fluid in a combustion reaction is expanding the most rapidly or where the fuel and oxidizer is not mixing.

Previous studies, [1-4], have described the emission from the dissociation of N_2 gas and the emission spectrum. While the majority of emission is in 550-600 nm range there are peak intensities at near UV and in the UV spectrum, specifically at 350 nm and around 382 nm. The belief is that the lower wavelength emission could be used for combustion reactions to increase signal to noise ratio. Although according to a previous study [2] 550-600 nm emission has a longer lifetime than any other emission making it more viable for the quenching of the combustion reaction. In a study very similar to this one, an unfiltered signal had a lifetime of 1 μs [1].

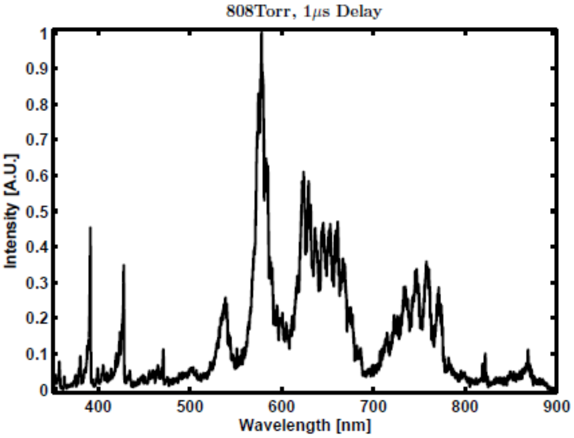


Figure 1. FLEET N_2 Emission Spectrum [2]

In the previously mentioned experiment, involving FLEET in premixed methane pulsed detonation experiment, there was never mention of using a small width bandpass filter to only obtain signal from the dissociation of N_2 gas.

A previous study utilized the CN vibration when dissociating a methane air flame via multi-photon dissociation from a femtosecond laser [7]. The main finding of this paper is the linear correlation of the CN vibration to the equivalence ratio. This study was the basis for the CN tracking method, while not considered FLEET it is still Laser Induced Breakdown Spectroscopy. This paper showed that the cyanide (CN) formation from dissociated atoms of methane and nitrogen can be tagged in order to find equivalence ratio.

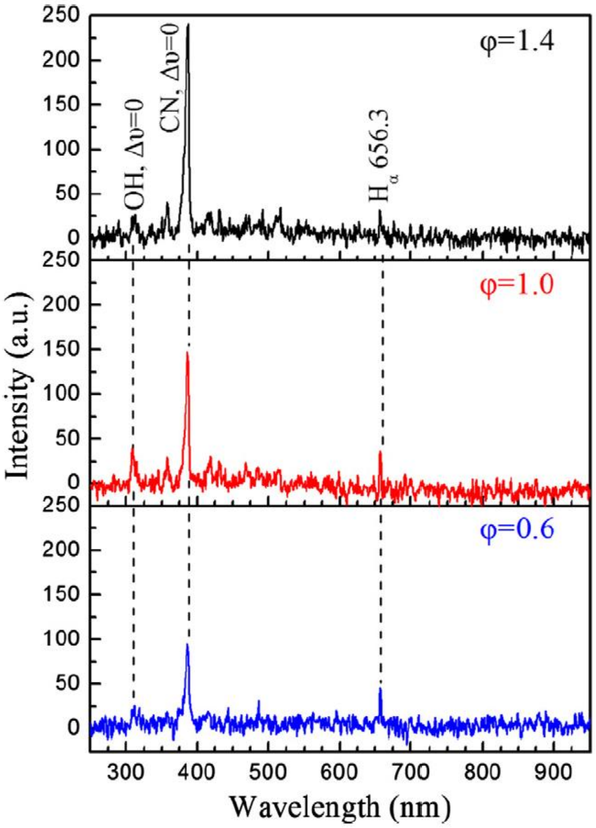


Figure 2. CN vibrational intensity [5]

II. METHOD

The experiment was run using a coherent Ti:Sapphire laser with wavelength of 800 nm at 120 fs pulses exciting the N_2 gas with focusing lens of +50 mm to provide the highest intensity to noise ratio in the combustion process. To prevent the camera from sensing emissions from other gases, mainly methane, filters were used; mainly a 550-600 nm [2]. It is known that the higher the concentration of O_2 gas, the lower the intensity of the FLEET emission, due to the formation of nitric oxide (NO). Using an intensified camera with a gate of 200 ns and a range of delays from 200 ns to 500 ns, the FLEET emission was captured with minimal noise from the flame. The hypothesis is that the signal intensity would mimic the N_2:O_2 ratio, shown in Figure 2, therefore mimic the equivalence ratio while moving laterally along the flame. The method of creating baseline measurements was to measure intensities of known O_2 counts such as in atmospheric air and the center of the jet of CH_4.

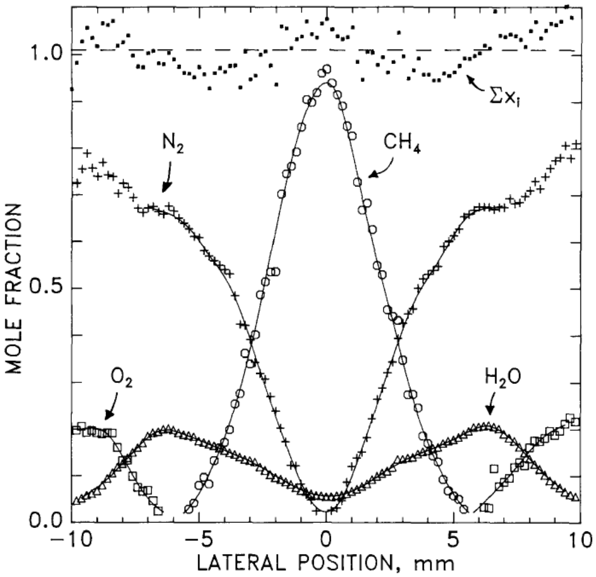


Figure 2. Gas concentrations in methane air diffusion flame [5]

To control the effect of change in temperature on the signal intensity a shorter width bandpass filter was used, as temperature has little effect on wavelengths with peak intensities such as $\lambda=382$ nm [3] and with the gate at 200ns the smaller increase of non-peak intensities would be null. To take the temperature into effect, Coherent Anti-Stokes Raman Spectroscopy (CARS) could be used alongside FLEET. A methane cylinder with a needle valve, a check valve, and a rotameter attached to ¼" tubing was used to create a laminar flame with a Reynolds number below 4000. Previous experiments including the N_2 concentration used mass flow rate of around 1.1 mg/s, this was around the mass flow rate for this experiment to compare FLEET results to another already published model of gas concentrations in a methane air diffusion flame at atmospheric pressure [5-6] shown in Figure 2.

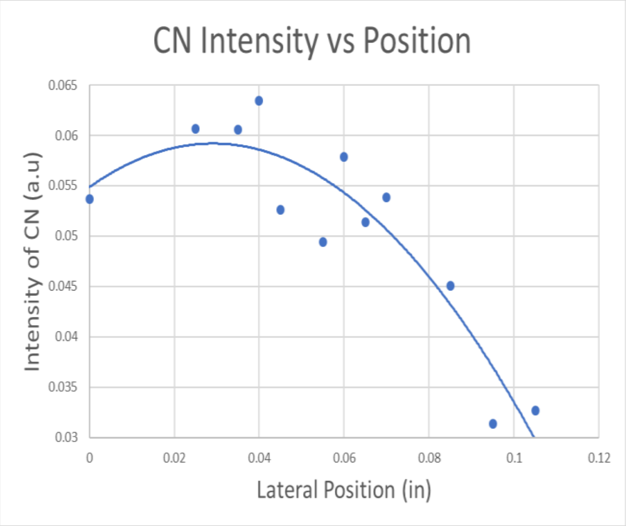
Overall, if averaging along lateral points, the equivalence ratio of a diffusion flame, φ, should be equal to one. This is a simple check to see if FLEET can be used to make accurate local φ measurements. There are other controlled data points of known O_2 concentration to create an exponential decay to fit signal intensity as a function of N_2:O_2 ratio. If this proves to be inaccurate FLEET is also a velocimetry technique, meaning the shear layer or the highest mixing region, φ=1, could be found using velocimetry techniques.

According to a previous FLEET study, the laser interacts with methane gas [1], CH_4, emitting light therefore a set of filters ranging from 340-360 nm, 370-390 nm and 550-600 nm were used to mitigate the emission of FLEET from CH_4. Since there are no prior studies of FLEET emissions from CH_4 the emission spectrum is unknown. The filters were chosen based on the N_2 dissociation emission spectrum [2], and the width of the bandpass was to minimize both the effect of temperature on intensity as well as the CH_4 emission. The alternative to this method would be to use the time constant of the FLEET emission to differentiate the signal from N_2 gas from CH_4 signal.

The method that showed the most promising results was by tagging the CN emission with a Photo Multiplier Tube (PMT) with a 390 nm ±10 nm bandpass filter. The flame was not entirely enclosed causing instabilities with increasing radial distance. The intensity of the CN vibration was recorded via voltage output with an oscilloscope with a voltage gain of 200 V in a low light setting.

III. RESULTS

The CN emission proved the most promising as it is already linked to a correlation with equivalence ratio. While the data is not yet calibrated, the results are promising in that the CN signal is at the level of noise while still within the, meaning there is no remaining methane at that point. This is promising and a good proof of concept that the CN vibration can be tagged within a diffusion flame. It is still unknown if the near UV N_2 emission can be used to measure equivalence ratio or just the CN vibration. Shown in Figure 4, the methane jet's width can be calculated by the maximum steady state value, and the point where there is no methane remaining can also be found by finding the point where the minimum steady state begins, in this case 0.095". The results are not verified, and the accuracy is unknown. The use of a spectrometer to measure concentrations of each fluid would provide the accuracy to the results obtained from this study.



The data collected in Figure 4 does not include the entire gaussian peak that is due to the noise level steady state value at the .095" point. This point is specifically where there is no methane hence there is no formation of CN just the noise from creating plasmas and some near UV N_2 emissions. The standard deviation of peaks obtained increases with respect to lateral position. This is due to a decrease of pressure at the fringes of the burner entrapping air causing the flame to flicker. This experiment would be greatly improved with a mixing chamber that is not affected by surroundings.

While the N_2 reformation emissions are not mentioned, it is still the belief that they could be used to find equivalence ratio but are not the best method. The CN emission is clearly the better option as it has a linear correlation between equivalence ratio and intensity. Due to a minor problem with the Ti:Sapphire laser the quantity of data is rather small but it proves that the relationship between CN vibrational intensity and equivalence ratio.

IV. CONCLUSION

FLEET is generally considered a Molecular Tagging Velocimetry (MTV) method, however, the goal of this study was to utilize the effect oxygen has on the signal given from FLEET to find local equivalence ratios, φ. Using the signal strength as a variable with the velocimetry technique already utilized by FLEET studies, areas of mixing can be found. This is relevant as other laser diagnostic techniques used for measuring φ are extremely complex or time consuming such as CARS, or rely on seeded particles such as PLIF. While both measurement techniques are used to measure temperature, FLEET is used to measure the N_2: O_2 ratio.

While this study did not obtain a wide variety of quantitative data, it however proves that a femtosecond laser can be focused onto the flame to track the methane fuel content and therefor the CN formation. There is another method that wasn't used due to limitations, that does seem promising as well but would require either two filtered cameras/PMT's or limit the speed of gathering data with a spectrometer. This method would be to evaluate the ratio of a constant or near constant emission with that of the CN emission. This method would be very useful and could provide the user with an already calibrated data table to see the most reactive positions. Then knowing this, other properties of the combustion reaction could be known by using computational fluid dynamics (CFD).

The method is also very useful for making simultaneous velocity and equivalence ratio measurements within a flame. This makes it very useful for highspeed flow systems such as in pulse detonations or rotating detonation engines, though a longer distance focal lens would be needed. The need for this method in rotating and pulse detonation engines is due to the formation of shockwaves which make conventional velocimetry diagnostics such as seeded PIV not usable whereas with FLEET it is possible to find remaining fuel and the velocity of the exhaust fluid without any seeded particles or intrusive instruments.

Further studies will be conducted to establish a calibration and then estimates of error before moving onto turbulent systems; as well as using a two camera/PMT system to monitor the relative intensity of the CN vibration with respect to a more stable or constant emission from the laser dissociating fluids within the system. While the PMT is very useful for making very rapid measurements it cannot be used for velocimetry techniques. Meanwhile, due to a limited lifetime of the CN vibration it is unknown how accurate the experimental results from using this method would be.

ACKNOWLEDGMENT

This paper would not have been completed without the tremendous generosity of Zucrow Laboratories of Purdue West Lafayette. It was truly a fantastic experience as an undergraduate to conduct research with some of the most talented individuals working there. Jordan Fisher was a tremendous help and mentor throughout this study and it would not have been completed without him. I would like to thank my faculty advisor Professor Mikhail Slipchenko who pushed me to complete my work to the best of my ability. I would like to thank Spectral Energies for lending the photo multiplier tube for this study.

REFERENCES

[1] N. DeLuca, R. Miles, N. Jiang, W. Kulatilaka, A. Patnaik, and J. Gord, "FLEET velocimetry for combustion and flow diagnostics," Appl. Opt. 56, 8632-8638 (2017).

[2] N. J. Deluca, R. B. Miles, W. D. Kulatilaka, N. Jiang, and J. R. Gord, “Femtosecond Laser Electronic Excitation Tagging (FLEET) Fundamental Pulse Energy and Spectral Response,” *30th AIAA Aerodynamic Measurement Technology and Ground Testing Conference*, 2014.

[3] B. R. Halls, N. Jiang, J. R. Gord, P. M. Danehy, and S. Roy, “Mixture-fraction measurements with femtosecond-laser electronic-excitation tagging,” *Applied Optics*, vol. 56, no. 11, 2017.

[4] N. Jiang, B. R. Halls, H. U. Stauffer, P. M. Danehy, J. R. Gord, and S. Roy, “Selective Two-Photon Absorptive Resonance Femtosecond-Laser Electronic-Excitation Tagging (STARFLEET) Velocimetry in Flow and Combustion Diagnostics,” *32nd AIAA Aerodynamic Measurement Technology and Ground Testing Conference*, Oct. 2016.

[5] K. C. Smyth, J. Miller, R. C. Dorfman, W. Mallard, and R. J. Santoro, “Soot inception in a methane/air diffusion flame as characterized by detailed species profiles,” *Combustion and Flame*, vol. 62, no. 2, pp. 157–181, 1985.

[6] P. H. Joo, M. R. Charest, C. P. Groth, and Ö. L. Gülder, “Comparison of structures of laminar methane–oxygen and methane–air diffusion flames from atmospheric to 60atm,” *Combustion and Flame*, vol. 160, no. 10, pp. 1990–1998, 2013.

[7] M. Kotzagianni and S. Couris, “Femtosecond laser induced breakdown spectroscopy of air–methane mixtures,” Chemical Physics Letters, vol. 561-562, pp. 36–41, 2013.