Investigation of flame properties and the impact of ammonia addition on soot formation

Varun Viswamithra* and Shyam Menon[†] Louisiana State University, Baton Rouge, LA, 70808

Addition of anhydrous ammonia into a model gas turbine combustor operating on methane fuel is being pursued to understand its impact on soot formation. Soot formation and temperature estimation is done with the use of two-color method.

I. Introduction

Ammonia is being looked at as a energy carrier. It is a carbon free combustion source. It is however essential to note that the heating value of Ammonia (18.6MJ/kg) is about third the heating value of CH4(50.2MJ/kg). Hence it becomes difficult to use this as a complete replacement of carbon based fuels. However it can be used as a fuel additive. It then becomes important to understand what its effects are in terms of NOx emissions. Its implications on the soot formed

Several studies have been conducted to understand the effects of ammonia addition into various types of flames. Montgomery, M. J et al. [2] in a study addressing the chemical influence of ammonia on soot formation noted that NH3 addition reduced the overall CO2 emissions, it also led to reduced soot formation. A. Valera-Medina et al. [3] conducted numerical and experimental studies to understand flame stability and emission patterns in model gas turbine simulating swirling flows, which used methane and ammonia as fuel in varied mixture ratios, he concluded that there needs to be a stratified injection technique to favor production of reactive species. He also notes that a medium swirl number can cause unfavorable stability issues and suggests to further study directed towards flame characteristics for low swirl number cases. Khateeb, A. A., et al. [4] in a study related to stability and exhaust NO performances with ammonia-methane-air-swirl flames concludes that addition of ammonia widens the range of equivalence ratios for which the swirl flame is stable. He also noted that a concentration of NO decreased with increased ammonia addition. Okafor, E. C., et al. [5] in a study of control of NOx and other emissions using micro gas turbine running on mixtures of ammonia and methane concluded that rich-lean combustion of CH4-NH3-air mixtures emitted less NOx than NH3-air mixture because higher flame speed of CH4-NH3-air mixtures ensured lower NOx in secondary combustion zone



Fig. 1 Motivation for lean operation of gas turbine engines [1]

This study aims to understand the effects of addition of ammonia on the temperature. In addition to this CH*

^{*}Graduate Research Assistant, Mechanical and Industrial Engineering Department, vviswa1@lsu.edu

[†]Assistant Professor, Mechanical and Industrial Engineering Department, smenon@lsu.edu

chemiluminescence has been used to study the heat release pattern and intensity fluctuations.

A. Experimental Setup





Fig. 2 Model gas turbine combustor setup to be used in characterizing the fuel injector performance

Fig. 3 Model gas turbine combustor in operation

Development of this combustor 2 was carried out by Giglio [6] to conduct fundamental combustion research on gaseous fuels. The combustor provides optical access, which will be utilized to study combustion and heat release processes. Temperature will be measured using a four point type-B Thermocouple rake which will enable us to obtain temperature distribution of exhaust gases in the combustion chamber. Thermocouples are located in the downstream section of the combustion chamber. Pressure measurements are done using quartz piezoelectric pressure transducers and are mounted on the wall of the combustion chamber. Figure 6 shows the location of pressure and temperature measuring devices. Both temperature and pressure measurements are done in the downstream section of the combustion chamber and hence measure exhaust gas properties. Figure 3 shows the combustor in operation.

The schematic of the test rig is shown in Figure 4. Air at preset pressure is inducted into the combustion chamber. CH4 and NH3 are mixed based on equivalence ratio required using rota-meters and inducted into the combustion chamber. The cooling water cools all the upstream components. The data acquisition system in this case consists of DSLR camera to capture images to carryout the two color pyrometry and high speed camera coupled with intensifier to carryout CH* chemiluminescence. Other important components in the experimental setup include Thermocouples and Ignition system



Fig. 4 Schematic of Experimental Setup

II. Approach

A. Two color Method

This work aims to study the effect of addition of ammonia in varied quantities with air and to investigate its impact on the soot formation. The estimation of soot formation will be done using two-color method [?]. In two-color method, radiation intensity from the soot particles is detected using two selected wavelengths. Temperature is then determined using the ratio of two wavelengths by elimination of an unknown factor.

In two-color method a temperature called apparent temperature is defined for a black-body which will emit radiation at an equivalent intensity as that of a grey body at the same temperature. From T_a , it follows that

$$I_{b,\lambda}(T_a) = I_{\lambda}(T) \tag{1}$$

We know that monochromatic emissivity ϵ_{λ} is defined for a temperature T_a is given by

$$\epsilon_{\lambda} = \frac{I_{b,\lambda}(T_a)}{I_{b,\lambda}(T)} \tag{2}$$

Using the above equations with the Planck's equation, we get:

$$\epsilon_{\lambda} = \frac{\exp^{C_2/\lambda T} - 1}{\exp^{C_2/\lambda T_a} - 1}$$
(3)

 ϵ_{λ} is estimated for soot particles by the widely used Hottel and Broughton correlation [?], given below:

$$\epsilon_{\lambda} = 1 - \exp^{(-KL/\lambda^{\alpha})} \tag{4}$$

where, K is absorption coefficient per unit flame thickness, which is directly proportional to the number density of soot particles. L is the thickness of the flame along LOS of flame detection system. Combining the 3 and 4 we get

$$KL = -\lambda^{\alpha} \ln \left[1 - \left(\frac{\exp^{C_2/\lambda T} - 1}{\exp^{C_2/\lambda T_a} - 1} \right) \right]$$
(5)

The KL value which is unknown is eliminated by using Equation 6 for two different wavelengths.

$$\left[1 - \left(\frac{\exp^{(C_2/\lambda_1 T)} - 1}{\exp^{(C_2/\lambda_1 T_{a_1})} - 1}\right)\right]^{\lambda_1^{a_1}} = \left[1 - \left(\frac{\exp^{(C_2/\lambda_2 T)} - 1}{\exp^{(C_2/\lambda_2 T_{a_2})} - 1}\right)\right]^{\lambda_2^{a_1}}$$
(6)

In equation 6, T is the flame temperature. Temperatures T_{a1} and T_{a2} are apparent flame temperatures at wavelengths λ_1 and λ_2 respectively. By rearranging terms we arrive at an equation which is an implicit equation in temperature as shown below:

$$T = \frac{\frac{hc}{k} \left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1} \right)}{ln \left[\frac{1}{C_{\lambda_1 \lambda_2}} \frac{\epsilon_{\lambda_2}}{\epsilon_{\lambda_1}} \frac{S_{\lambda_1}}{\lambda_2} \left(\frac{1 - \exp(-hc/k\lambda_1 T)}{1 - \exp(-hc/k\lambda_2 T)} \right) \right]}$$
(7)

where, $C_{\lambda_1\lambda_2}$ is the calibration constant given by

$$C_{\lambda_1 \lambda_2} = \left(\frac{\eta_1}{\eta_2}\right) \left(\frac{\eta_{\rm D1}}{\eta_{\rm D2}}\right) \left(\frac{\lambda_2}{\lambda_1}\right)^5 \left(\frac{\Delta \lambda_1}{\Delta \lambda_2}\right) \tag{8}$$

Once the temperature is obtained we can estimate the soot volume fraction using the equation below:

$$f_{\nu} = -\frac{\lambda_S}{K_{ext} * L} ln \left\{ 1 - \epsilon_L(\lambda, T_L) \frac{\tau_S}{\tau_L} \frac{S_{S\lambda}}{S_{L\lambda}} \exp\left[-\frac{hc}{k\lambda_S} \left(\frac{1}{T_L} - \frac{1}{T_S} \right) \right] \right\}$$
(9)

III. Chemiluminescence

Chemiluminescence, is the emission of light by a particular chemical reaction where certain types of radicals emit light at a particular wavelength. CH* filter is made use of in this study to understand the heat release patterns and intensity fluctuations. Setup used for the study uses a Photron SA3 high-speed camera along with lenses and an intensifier (Model: UVI1850) to amplify the flame image obtained through CH* filter which operates at 430 ± 5 . The image 5 below shows the setup for chemiluminescence:



Fig. 5 Chemiluminescence Setup

IV. Results and Discussion

The major component of work that has been completed relates to the setup of the model combustor. In addition to the mechanical setup, a data acquisition and control system has been setup to run the combustor setup as per required procedure which records temperature data as discussed in I.A. Figure 7 shows the combustor in operation. The fuel used for this test run is methane and ammonia.

A. Temperature

The images shown below shows the effect of addition of ammonia, there is significant color change due to light emission happening at different wavelengths.







70 % CH4- 30% NH3

Equivalence Ratio $\phi = 1.1$, Effect of addition of Ammonia in varied percentages

85 % CH4- 15% NH3



100 % CH4- 0% NH3



70 % CH4- 30% NH3

Images shown below shows a direct comparison of the image obtained from the DSLR camera (left) and the contour plot of the normalized temperature field obtained by processing the same image. The central and side wall re-circulation zones in a swirl combustor can be clearly seen in the contour plot. Highest temperatures are attained in these regions and the normalized plots predict the right locations as well.



Fig. 6 Flame



Fig. 7 Corresponding Temperature field

B. CH* Chemiluminescence

Figures below shows time averaged images of CH* Chemiluminescence. There are cases for different values of equivalence ratios ϕ from 1.2-0.8 varied in steps of 0.1. Each equivalence ratio has three cases for different amount of ammonia added. The case for equivalence ratio of 0.8 shows that the flame is stable and with the addition of 15% ammonia the CH* radicals are displaced and hence we see the image to have a dispersed flame structure, with the addition of 30% ammonia the flame is further more dispersed. This trend is seen in cases with all equivalence ratios. With increasing equivalence ratio the flame becomes more and more unstable and we see larger displacements of flame and It is interesting to note that for equivalence ratio of $\phi = 1.2$ and 30% ammonia, We capture flame with largely dispersed CH* radicals and we see the same in time averaged image for this case.



100 % CH4- 0% NH3



85 % CH4- 15% NH3 Equivalence Ratio $\phi = 0.8$



70 % CH4- 30% NH3



100 % CH4- 0% NH3



85 % CH4- 15% NH3 Equivalence Ratio $\phi = 0.9$



70 % CH4- 30% NH3



100 % CH4- 0% NH3



85 % CH4- 15% NH3 Equivalence Ratio φ = 1.0



70 % CH4- 30% NH3



100 % CH4- 0% NH3



85 % CH4- 15% NH3 Equivalence Ratio *φ* = 1.1



70 % CH4- 30% NH3



C. CH* Chemiluminescence- Intensity Fluctuations

The plots below show intensity fluctuations as a function of equivalence ratio. We see that with decreasing value of equivalence ratio the intensity of fluctuation decreases. This is due to the fact with reduced equivalence ratio the energy of the flame reduces and hence we see oscillations with smaller amplitudes.



Intensity fluctuations as a function of Equivalence ratio

The images below show the intensity fluctuations over 1000 frames for varying equivalence ratios and percentage ammonia additions. It is seen that for the case with $\phi = 1.0$ and pure ammonia case the intensity fluctuations are minimal when compared to the cases with 15%. The is because the ammonia addition lowers the adiabatic flame temperature and leads to a weaker flame and hence larger oscillations. In case of 30% ammonia addition the mean intensity of the flame goes down and hence the fluctuations are seen with lower mean value of the flame intensity. Flame intensity plot with equivalence ratio $\phi = 0.9$ and pure methane case is seen to have lower mean value when compared to the case with pure methane at $\phi = 1.0$ this is due to the lower amount of fuel inducted into the combustion chamber. The Flame intensity plot with equivalence ratio $\phi = 0.9$ and 15% ammonia addition has larger oscillations when compared to the case with pure methane and same ϕ . The case with 30% ammonia has lower oscillations, as more ammonia is inducted into the combustion chamber the flame so weak so to not be able to oscillate or show any signs of instability.



V. Conclusion

Re-purposing of model gas turbine combustor to operate with ammonia-methane-air mixtures has been completed It was seen that Temperature estimation using two-color pyrometry showed reduction in flame temperature with increasing NH3 addition consistent with previous observations. CH* chemiluminescence imaging showed expected trends: increase in reaction zone size and intensity with increasing phi and decreasing intensity with increasing NH3 content. CH* intensity showed increasing fluctuations at rich mixtures consistent with higher heat release oscillations. NH3 addition appeared to cause increase in heat release oscillations

VI. Future Work

The following work will be taken up in the near future

- Study the flame characteristics as a function of back pressure
- Conduct thorough temperature and pressure measurements of exhaust gases
- Conduct similar studies using with the ammonia addition using liquid fuel
- Add Laser-induced incandescence (LII) and compare the efficacy of two color pyrometry against LII

Acknowledgments

This work is supported by NASA EPSCoR and the Board of Regents of the state of Louisiana.

References

[1] Dunn-Rankin, D., Lean combustion: technology and control, Academic Press, 2011.

- [2] Montgomery, M. J., Kwon, H., Xuan, Y., McEnally, C. S., and Pfefferle, L. D., "Chemical influence of ammonia on suppressing soot formation pathways," Tech. rep., Yale Univ., New Haven, CT (United States), 2020.
- [3] Valera-Medina, A., Marsh, R., Runyon, J., Pugh, D., Beasley, P., Hughes, T., and Bowen, P., "Ammonia–methane combustion in tangential swirl burners for gas turbine power generation," *Applied Energy*, Vol. 185, 2017, pp. 1362–1371.
- [4] Khateeb, A. A., Guiberti, T. F., Zhu, X., Younes, M., Jamal, A., and Roberts, W. L., "Stability limits and exhaust NO performances of ammonia-methane-air swirl flames," *Experimental Thermal and Fluid Science*, Vol. 114, 2020, p. 110058.
- [5] Okafor, E. C., Somarathne, K. K. A., Ratthanan, R., Hayakawa, A., Kudo, T., Kurata, O., Iki, N., Tsujimura, T., Furutani, H., and Kobayashi, H., "Control of NOx and other emissions in micro gas turbine combustors fuelled with mixtures of methane and ammonia," *Combustion and Flame*, Vol. 211, 2020, pp. 406–416.
- [6] Giglio, A. L., "Design, Fabrication, and Testing of a Micro Fuel Injection Swirler for Lean Premixed Combustion in Gas Turbine Engines," 2008.