Final Report

Plasma-Enhanced Combustion of Hydrocarbon Fuels and Fuel Blends Using Nanosecond Pulsed Discharges

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Professor Mark Cappelli and Professor M. Godfrey Mungal recently completed a project funded through the Department of Energy titled "Plasma Enhanced Combustion of Hydrocarbon Fuels and Fuel Blends Using Nanosecond Pulsed Discharges". This project had as its goals the study of fundamental physical and chemical processes relevant to the sustained premixed and non-premixed jet ignition/combustion of low grade fuels or fuels under adverse flow conditions using non-equilibrium pulsed nanosecond discharges. A summary of the research findings is presented below. The research resulted in the following publications and presentations:

1. M. S. Bak, S.K. Im, and M. A. Cappelli, *Successive laser-induced breakdowns in atmospheric pressure air and premixed ethane–air mixtures*," Combustion and Flame **161**, 1744–1751, 2014.
2. M.S Bak and M.A. Cappelli, *Simulations of Nanosecond Pulsed Discharges in Atmospheric Pressure Air*," J. Appl. Phys. **113**, 113301, 2013.
3. M. S. Bak, S.K. Im, M. G. Mungal, and M. A. Cappelli, *Studies on the stability limit extension of premixed and jet diffusion flames of methane, ethane, and propane using nanosecond repetitive pulsed discharge plasmas*," Combustion and Flame **60**, 2396–2403, 2013.
4. S.K. Im, M. S. Bak, M. G. Mungal, and M. A. Cappelli, *Plasma Actuator Control of a Lifted Ethane Turbulent Jet Diffusion Flame*," IEEE Trans. Plasma Sci. **41**, 3293 – 3298, 2013.
5. M.S. Bak, H. Do, M.G. Mungal, and M.A. Cappelli, *Plasma-Assisted Stabilization of Laminar Premixed Methane/Air Flames around the Lean Flammability Limit*," Combustion and Flame **159**, 3128–3137, 2012.
6. M. Bak and M. Cappelli, "*A simulation of the effects of varying repetition rate and pulse width of nanosecond discharges on premixed lean methane-air combustion*," Journal of Combustion, **137653**, 2012.
7. N. Hwang, S. Im, M. Bak, and M. Cappelli, "*Characteristics of low frequency air glow dielectric barrier discharges at atmospheric pressure*," Bulletin of the American Physical Society **57**, 7, PR1.00055, presented at the 65th Annual Gaseous Electronics Conference, October 22–26, 2012; Austin, Texas.
8. M. Bak, and M. Cappelli, "*Laser-induced breakdown stabilization of non-premixed jet diffusion flames*" Bulletin of the American Physical Society **57**, 7, NW1.00041,

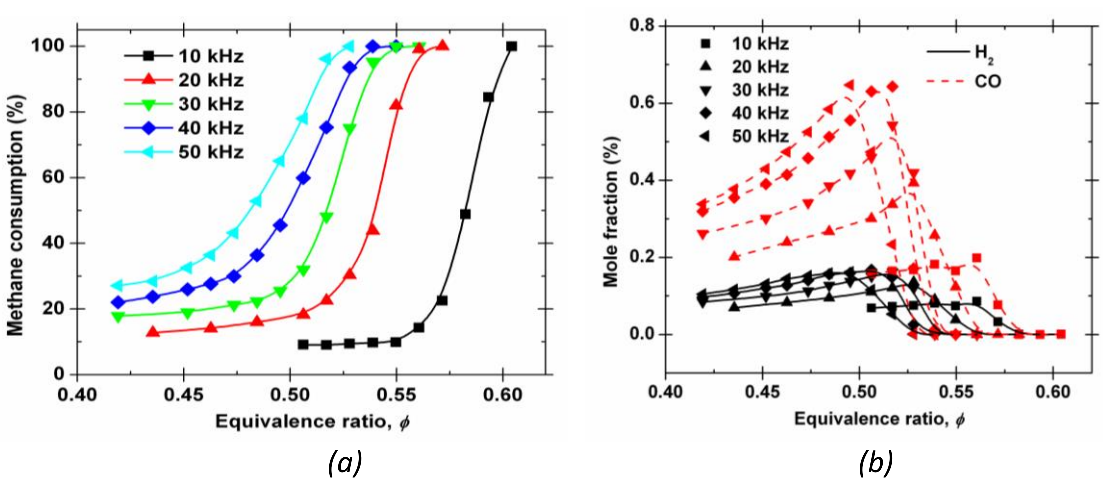
presented at the 65th Annual Gaseous Electronics Conference, October 22–26, 2012; Austin, Texas.

9. M. Bak, and M. Cappelli, "*Nanosecond repetitively pulsed discharge control of premixed lean methane-air combustion*," Bulletin of the American Physical Society **57**, 7, NW1.00042, presented at the 65th Annual Gaseous Electronics Conference, October 22–26, 2012; Austin, Texas.
10. M. S. Bak, H. Do, M. G. Mungal, and M. A. Cappelli, "*Plasma-assisted stabilization of laminar premixed methane/air flames around the lean flammability limit*," Combustion and Flame, April 24, 2012 (available on-line).
11. M.S. Bak, W. Kim, and M.A. Cappelli, "*On the quenching of excited electronic states of molecular nitrogen in nanosecond pulsed discharges in atmospheric pressure air*," Appl. Phys. Lett. **98** Article Number: 011502, January 2011. DOI: 10.1063/1.3535986.
12. M. S. Bak and M.A. Cappelli, "*Quenching of electronically excited states of molecular nitrogen in nanosecond repetitive pulsed discharges in atmospheric pressure air*" Bulletin of the American Physical Society **56**, 15, FTP1.00028, presented at the 64th Annual Gaseous Electronics Conference, November 14–18, Salt Lake City, UT, 2011.
13. M. S. Bak, H. Do, M.G. Mungal, and M.A. Cappelli, "*Laminar lean premixed methane/air combustion near the lean flammability limit using nanosecond repetitive pulsed discharge plasmas*" Bulletin of the American Physical Society **56**, 15, ET2.00002, 2011, presented at the 64th Annual Gaseous Electronics Conference, November 14–18, Salt Lake City, UT, 2011.
14. M. Bak, W. Kim, and M. Cappelli, "*Quenching of Excited Electronic States of Molecular Nitrogen in Nanosecond Pulsed Discharges in Atmospheric Pressure Air*," AIAA-2011-1150, 49th AIAA Aerospace Sciences Meeting including the New Horizons Forum and Aerospace Exposition, Orlando, Florida, Jan. 4-7, 2011.
15. Kim, W., Mungal, M.G., Cappelli, M.A., "*The role of in situ reforming in plasma enhanced ultra lean premixed methane/air flames*," Combustion and Flame **157**, 374-383, 2010.
16. D. Pai, M. Cappelli, C. Laux, "*Nanosecond Repetitively Pulsed Spark Discharges in Ambient Air*," Paper P1.5.48, Proceedings of the 19th International Symposium on Plasma Chemistry (A. von Keudell, J. Winter, K. Boke, P. Schulz von der Gathen), July 26-31, 2009.
17. D.Z Pai, M.A. Cappelli, C.O. Laux, D.A. Lacoste , "*Spark channel dynamics of nanosecond repetitively pulsed discharges in air at atmospheric pressure*," Paper PB10-7, 29th International Conference on Partially-Ionized Gases (ICPIG), July 12-17, 2009, Cancún, México.
18. W. Kim, H. Do, M. A. Cappelli and M. G. Mungal "*Plasma Assisted Flame Holding in Subsonic and Supersonic Flows*," AIAA-2010-0263, AIAA-ASM Weakly Ionized

Gases Workshop, 48th AIAA Aerospace Sciences Meeting, January 4-7, 2010, Orlando, FL.

19. Cappelli, M. A., "*Nonequilibrium Plasma Flame Holding in Subsonic and Supersonic Flows*," Aerospace Thematic Workshop: Fundamentals of Aerodynamic Flow and Combustion Control by Plasmas, Les Houches – Mont-Blanc, France, October 11-16, 2009.

During the first year of this research, we investigated both experimentally and by numerical simulations, plasma-chemical processes occurring in pulsed discharges primarily in air in the absence of fuel. We found that dissociative quenching of electronically excited N_2 (N_2 A, B, a′, C) by O_2 produces a significant amount of O radicals (4.4% mole fraction). The O-radical is a key species in combustion as it participates in initiation reactions that form OH radicals as well as other important intermediates. These results are fully documented in publications and the reader is referred to citations. 12, 13, and 14 above. During the second year, we studied the extent of stabilization and its mechanism in lean premixed jet flames with the pulsed discharges. Gas chromatography and thermocouple measurements were conducted along the streamwise direction over the fuel-lean range of equivalence ratio for different discharge repetition rates. Representative results (methane-air) are presented in Fig. 1. The results revealed that the pulsed nonequilibrium discharges can stabilize combustion, but its propagation towards downstream locations is largely limited to the known lean flammability limit. The equivalence ratio for the complete combustion is found to decrease from 0.605 at 10 kHz to 0.53 at 50 kHz. For ethane-air, we found a continuous combustion stream for any of the tested equivalence ratios for 10 kHz and 20 kHz however, the flame was intermittent and the fuel was largely unburnt. The equivalence ratio for complete combustion is found to decrease from 0.544 at 30 kHz to



(a) (b)

Fig. 1. (a) Methane consumption and (b) H_2 and CO mole fractions as a function of discharge repetition rate (constant average power) at an 11 mm fixed downstream location from the discharge.

0.503 at 50 kHz. For propane-air combustion, repetition rates beyond 30 kHz is required for obtaining a stable combustion stream. The equivalence ratio for complete combustion is found to decrease from 0.56 at 40 kHz to 0.54 at 50 kHz. While one general conclusion drawn from our research is that discharges are able to flame-hold under very fuel-lean conditions, we emphasize (citations 3, 5, 6, 10, 11, and 14) that the threshold ratios for complete combustion at locations downstream of the discharges are found to be very close to the known lean flammability limit (0.53, 0.52, and 0.54 for methane, ethane, and propane). The combustion problem, in essence, is limited by flame propagation and not just ignition and flame holding.

During the second year (with carryover into the third year) we also carried out studies of the stabilization of methane, ethane, and propane jet diffusion flames with nanosecond repetitive pulsed discharges (see citation 3). The fuel jet nozzle speeds and were varied (typically between 40 and 100 m/s) while co-flowing air is maintained constant (typically 1 m/s). The discharge is placed within a few jet nozzle diameters downstream of the nozzle. The discharge location could be moved along the radial direction to identify the locations were optimum stabilization is achieved. Our interests included the study of jet diffusion flame blow-off under these conditions. A methane jet, for example, is not stable in the absence of a discharge under these range of conditions (i.e., always experiences blow-off) whereas ethane and propane jet flames experience blow-off at nozzle speeds of 38.5 m/s and 32.0 m/s, respectively. The tested nozzle speeds for these studies are in the range above the minimum observed blow-off speeds.

A photograph of a typical discharge-stabilized propane flame is shown in Fig. 2 for operation well beyond the flame blow-off limit. A representative measurement of the combustion efficiency (at axial locations of a few tens of nozzle diameters downstream) and its variation with discharge radial placement is shown in Fig. 3 (a) also for propane jet-flames. Results are qualitatively similar for other fuels. We generally find that the discharge must be placed in a location where there is a certain range in fuel-air local equivalence ratio, with a weak sensitivity to flow conditions. The local equivalence ratio is determined by laser-induced breakdown spectroscopy (LIBS), the details of which are described in citations 8 and 9. The LIBS measurements allow us to characterize the local equivalence ratio, ϕ, across the jet

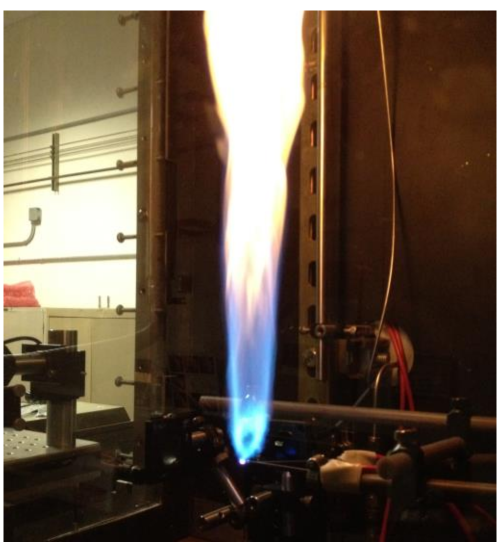
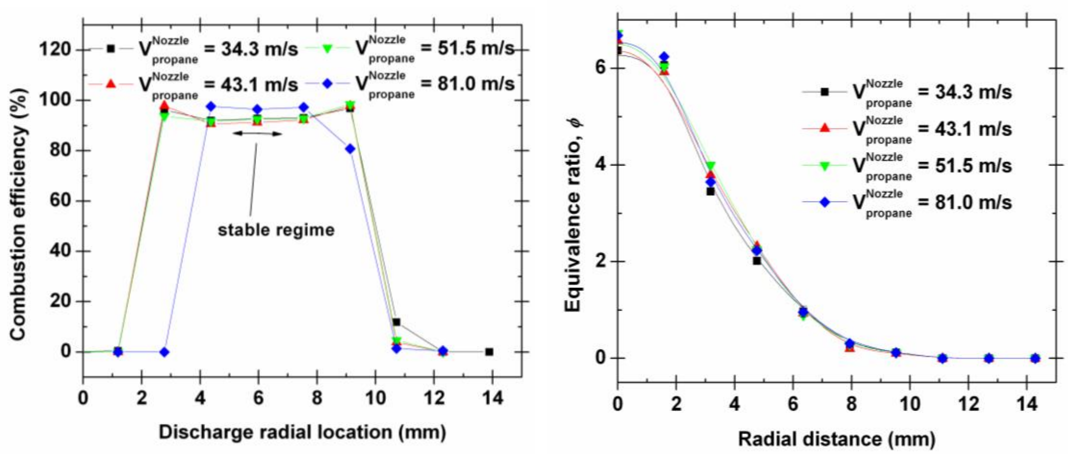


Fig. 2. Photograph of discharge-stabilized propane-jet flame. The discharge is located where $\phi \approx 1.27$, the jet speed is 43.1 m/s, and air coflow speed is 0.8 m/s.

mixing layer as shown in the right panel Fig. 3 (b). In general, no combustion (stabilization) is seen when the discharge is located where the fuel concentration is too rich (jet axis) or too lean (large radial locations). The flame is found to be anchored best to the discharge is placed where the local equivalence ratio, ϕ is between 0.8 and 1.9, i.e.,



(a) *(b)*

Fig. 3. (a) Propane jet combustion efficiencies at a fixed axial downstream location given radial discharge locations and (b) the corresponding equivalence ratios at the discharge locations.

where the mixture is close to a reasonably combustible condition (this regime is marked in the figures.). Outside of this range, the flame base begins to pulsate with diminishing frequency the further from the optimum location. In spite of this pulsation, complete combustion is still achieved close to the stable regime because of its high pulsation frequency. Therefore, the spatial region of complete combustion is found to be broader than that for stable flames, and it becomes more narrow as the jet speed increases for all of the tested fuels.

During the third year of the program, 2-D plasma kinetic and flame simulations (citation 6) were performed to investigate the mechanisms responsible for premixed jet-flame combustion enhancement and the effects seen of repetition rate and pulse width. While these simulations are primarily for premixed combustion, they do provide insight on the kinetics and chemistry of discharge-assisted non-premixed jet flames, as combustion is largely stabilized in regions where there is a partial premixing of the surrounding air-fuel streams. A representative result of the simulations is shown in Figs. 4 and 5 for premixed methane-air at an equivalence ratio of ϕ= 0.45, well below the lean flammability limit. More details can be found in citations 1 and 5. These images are frames taken from movies that describe the evolution of the simulations in time following several hundreds of laser pulses, which eventually achieves a quasi-steady state. The reader can find a representative movie of the simulations for methane-air flames in http://www.stanford.edu/group/pdl/news.html.

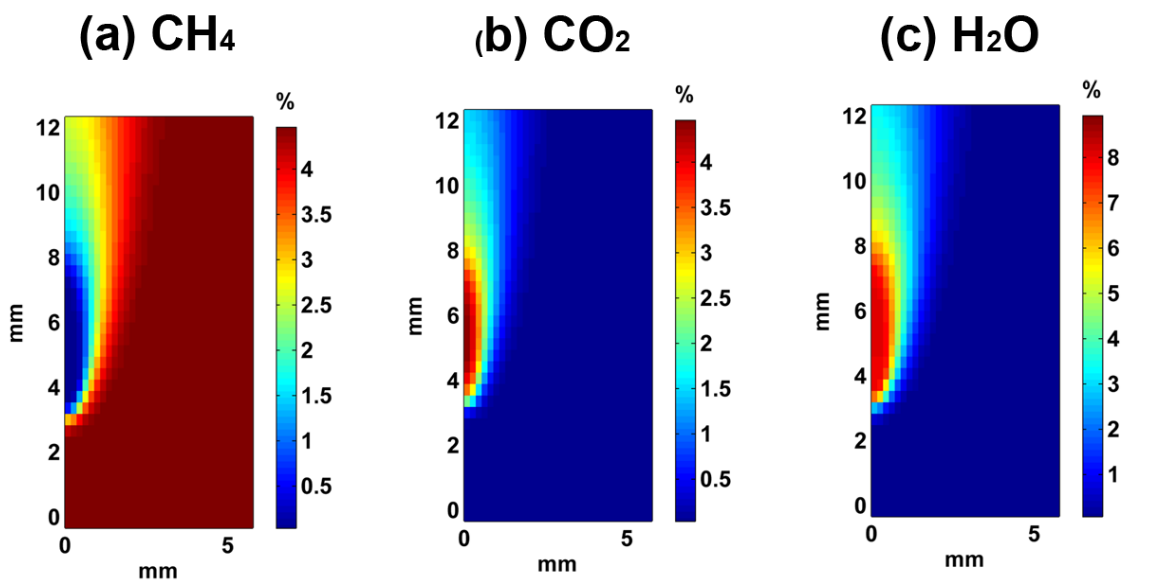


Fig. 4. Quasi-steady state spatial distribution of (a) CH_4, (b) CO_2, and (c) H_2O for a discharge repetition rate of 10 kHz and a 9 ns pulse width. The discharge is located between 3 and 4 mm in axial location and is 1 mm in radial dimension.

Simulations such as those depicted in Figs. 4 and 5 were carried out for a wide range of repetition rates and pulse widths under conditions of constant average power (typically several watts). The air and fuel flow direction is from bottom to top in these images. The discharge kernel is a region of uniform field located between 3.5 and 4.5 mm and of radius 0.7 mm. The effective reduced electric fields studied are in the range

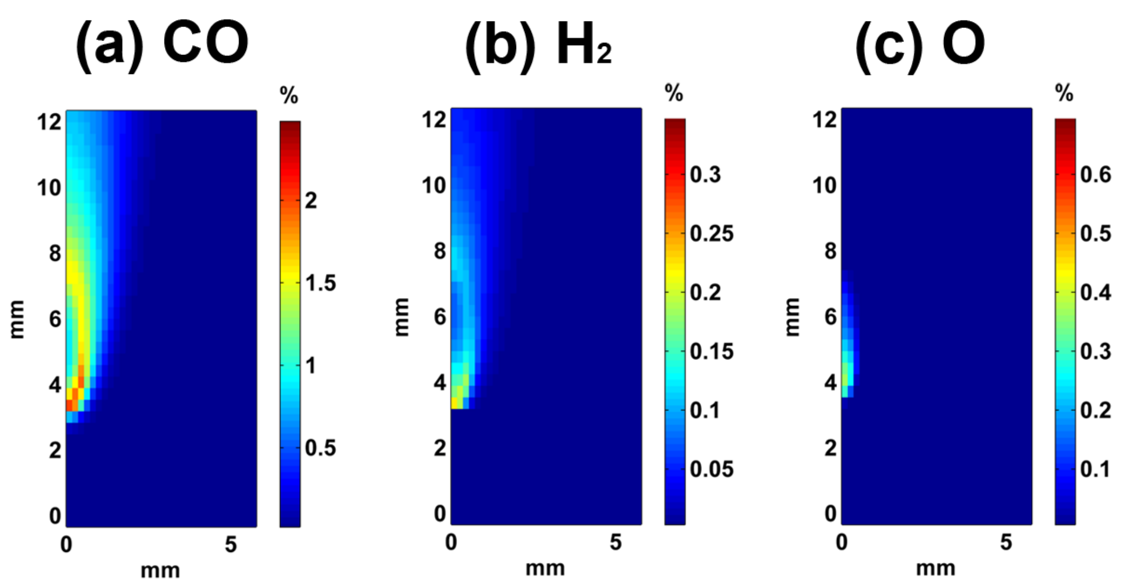


Fig. 5. Quasi-steady state spatial distribution of (a) CO, (b) H_2, and (c) O, for a discharge repetition rate of 10 kHz and a 9 ns pulse width.

of 315 – 350 Td, selected to best match the measured temperatures seen in the discharge. The simulations are able to predict the time-histories of the species within the discharge kernel, as well as the flame propagation characteristics. It is noteworthy that the methane concentration remains low within the discharge (and immediate downstream) region. The converse is true for the stable combustion products. Although combustion is initiated in the discharge kernel, it is apparent that it does not propagate, as the flame (as evidenced from the axial variations in the figure) is diluted by the surrounding unburnt gas stream. Conditions of slightly higher equivalence ratio would show the typical characteristics of a V-flame, indicating flame propagation, consistent with what is seen experimentally. The lack of flame propagation is also apparent upon close inspection of the intermediate species (Fig. 5). For example, oxygen atoms are seen to be produced in large quantities in the discharge kernel, but they recombine and/or are immediately diluted in the downstream flow. In a no-cost extension period, we were able to complete a study of the detailed simulations and kinetics of pulsed nanosecond discharges (citation 2), and examine a new means of using nonequilibrium plasma discharges in stabilizing combustion (citation 4). Finally, as it seems that use of an electrode discharge to stabilize combustion may be too intrusive in some applications, we have studied and compared to nanosecond pulsed discharges, the use of laser spark discharges in stabilizing premixed ethane-air flames (citation 1).

Student Training and Acknowledgements

This DOE-funded work provided support for Mr. Moonsoo Bak, who also leveraged funding from Stanford's Graduate Student Fellowships. It also provided partial support for Post-doctoral researcher Seong-kyun Im. For almost two years, the program benefited from the participation of independently-funded post-doctoral visitor (N. Hwang). Since the project completion, we have continued, with the external support of M. Bak, studies of plasma-assisted spray-flame combustion. We are in the process of negotiating a grant from the Air Force Office of Scientific Research, leveraging the knowledge gained from these studies, to expand our research to understand nonequilibrium plasma-stabilization of liquid fuel spray flames. Note that this DOE program was combined with NSF support through the NSF-DOE Program in Basic Plasma Sciences. A total of ten (10) peer-reviewed publications that have already been extensively cited are directly traceable to the support provided by the DOE/NSF program. The Principle Investigators are very grateful for the support.