DOE Low Temperature Plasma
Centers and User Facilities
Annual Meeting

Center for Low Temperature Plasma Interactions with Complex Surfaces (PICI)

Sandia Plasma Research Facility (SPRF)

Princeton Collaborative Low Temperature Research Facility (PCRF)

Center for Studies of Plasma-Assisted Combustion and Plasma Catalysis (PACC)

October 28-29, 2022
Hilton Garden Inn Reagan National Airport
Arlington, VA

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Fusion Energy Sciences Program
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U.S. DEPARTMENT OF ENERGY | Office of Science

Schedule

Zoom password: LTP2022

** indicates virtual talks*

Friday, October 28, 2022			
	Time (Eastern)	**Speaker**	**Title**
8:15 – 8:30 am		***Welcome*** *(Salon 1 & 2)*	
	8:15 – 8:30	Mark Kushner	Introduction to Annual Meeting
8:30 – 9:50 am		***Oral Session I: PICI*** *(Salon 1 & 2). Moderator: Aditya Lele*	
1	8:30 – 8:50	Aditya Bhan*	Pathways and Timescales Involving Radical and Vibrationally Excited Species for NH_3 Formation by Low Temperature, Atmospheric Pressure Plasma Catalysis
2	8:50 – 9:10	Selma Mededovic Thagard	Electrical Discharges in a Bubble Column Reactor: A Novel High Throughput Reactor Design for Water Treatment
3	9:10 – 9:30	Ali Mesbach	Towards Manufacture of Green Fertilizer via Low-Temperature Plasmas
4	9:30 – 9:50	Peter Bruggeman	Advancing Diagnostics for In Situ Measurements of Plasma Interactions with Complex Interfaces
9:50 – 10:10 am		***Break***	
10:10 am – 12:10 pm		***Oral Session II: PCRF*** *(Salon 1 & 2). Moderator: Brian Bayer*	
1	10:10 – 10:30	Yevgeny Raitses	Princeton Collaborative Low Temperature Plasma Research Facility (PCRF): Status Update and New Solicitation of User Proposals
2	10:30 – 10:50	Sophia Gershman	Initiation of a Nanosecond Discharge in Water with Gas Bubbles
3	10:50 – 11:10	Willca Villafana*	EDIPIC-2D, an Open-source Versatile and Comprehensive Particle-In-Cell code for Low-Temperature Plasmas Modeling
4	11:10 – 11:30	Mikhail Shneider	On the Role of Osmotic Pressure in the Interaction of Plasma with Cells in a Physiological Saline
5	11:30 – 11:50	Yevgeny Raitses	Electron Beam Generated ExB Plasmas and Their Applications
6	11:50 – 12:10	Arthur Dogariu	Advanced Optical Diagnostics for Low Temperature Plasmas; Imaging Solvated Atoms with fs-TALIF
12:10 – 1:40 pm		***Lunch***	

Friday, October 28, 2022 (continued)			
	Time	**Speaker**	**Title**
1:40 – 2:40 pm		***Oral Session III: PACC*** *(Salon 1 & 2). Moderator: Joshua Morsell*	
1	1:40 – 2:00	Igor Adamovich	Hybrid Ns Pulse / Capacitively Coupled RF Discharges for Plasma Chemistry and Plasma Catalysis
2	2:00 – 2:20	Yiguang Ju	Plasma Assisted Combustion and Manufacturing
3	2:20 – 2:40	Bruce Koel	Issues of Effectiveness and Energy Efficiency in Plasma Assisted Catalysis for Ammonia Synthesis
2:40 – 3:50 pm		***Oral Session IV: SPRF*** *(Salon 1 & 2). Moderator: Andrew Powis*	
1	2:40 – 2:50	Christopher Shaddix	Sandia’s Plasma Research Facility (PRF): Status Update and New Solicitation of User Proposals
2	2:50 – 3:10	Christopher Kliewer	Nonlinear Optical Diagnostics Laboratory for Low-Temperature Plasma Assisted Chemistry
3	3:10 – 3:30	Lucas Beving	How Sheath Properties Change with Gas Pressure: Modeling and Simulation
4	3:30 – 3:50	Jonathan Frank	Imaging of Methyl Radical and Hydrogen Peroxide in Pulsed Plasmas by Photofragmentation Laser-Induced Fluorescence
3:50 – 4:00 pm		***Break***	
4:00 – 4:50 pm		***Poster Session I*** *(Salon 3 & 4)*	
4:50 – 5:40 pm		***Poster Session II*** *(Salon 3 & 4)*	

Saturday, October 29, 2022			
	Time	**Speaker**	**Title**
8:40 – 10:20 am		***Oral Session V: PICI*** *(Salon 1 & 2). Moderator: Veda Gajula*	
1	8:40 – 9:00	Gottlieb Oehrlein	Study of Surface Interactions for Plasma Catalytic Nitrogen Oxidation
2	9:00 – 9:20	Steven Shannon	Surface Ionization Wave Propagation: Surface Permittivity and Capacitance Effects
3	9:20 – 9:40	Brian Bentz	Tomographic Optical Imaging of a Pulsed Atmospheric Pressure Plasma Jet
4	9:40 – 10:00	Igor Adamovich	Characterization of Plasmas in Contact with a Liquid Water Surface
5	10:00 – 10:20	Mark Kushner	Atmospheric Pressure Plasma Interactions with Channels and Embedded Particles
10:20 – 10:40 am		***Break***	
10:40 – 11:20 am		***Oral Session VI*** *(Salon 1 & 2)*	
	10:40 – 11:20	Mark Kushner	Review of DOE BRN on Plasmas for Microelectronics
11:20 am – 12:20 pm		***Group Discussion*** *(Salon 1 & 2)*	

Poster Session I. Friday, October 28, 4:00 – 4:50 pm		
	Presenter	**Title**
1	Christopher Burger	Plasma Assisted Chemical-Looping Combustion: Low-Temperature Methane and Ethylene Oxidation with NiO
2	Ning Liu	Sensitive and Single-shot OH and Temperature Measurements by Femtosecond Cavity Enhanced Absorption Spectroscopy
3	Keegan Orr	Laser Induced Fluorescence Measurements of Vibrationally Excited Oxygen Produced by Recombination of O Atoms
4	Caleb Richards	CO_2 Dissociation in Ns Pulse and "Hybrid" Ns-RF Discharge Plasmas
5	Andrew Powis	LTP-PIC, an Open-source, Three-dimensional Kinetic Code for Modelling Low-temperature Plasmas on Modern Supercomputing Architectures
6	Veda Prakash Gajula	Enhancing the Perfluorooctanoic acid (PFOA) Degradation through Plasma Reactor Parametric Optimization
7	Michael Hinshelwood	Study of Plasma-catalyst Surface Interactions for Nitrogen Oxidation
8	Victor Miller	Cold Plasma Treatment of Biomass Can Enhance Nitrate Content and Reduce Ammonia Evaporation
9	Sai Raskar	Propagation of Surface Ionization Waves Over Dry and Wet Microchannels Exposed to an Atmospheric Pressure Plasma Jet
10	Tanubhav Srivastava	Falling Liquid Film Plasma Reactor – A New Approach to Study Temporally Resolved Plasma-Liquid Interactions
11	Jianan Wang	OH and Water Vapor Concentration Measurements Surrounding Water Droplets in an Atmospheric Pressure Plasma
12	Brian Bentz	Photoemission Induced Plasma Breakdown in Argon

Poster Session II. Friday, October 28, 4:50 – 5:40 pm		
	Presenter	**Title**
1	Aditya Dilip Lele	Modelling the Effect of Surface Charging on Surface Reactions During Plasma Synthesis of NH_3 Using DFT
2	Xingqian Mao	Effects of Inter-pulse Coupling on Nanosecond Pulsed High Frequency Discharge Ignition in a Flowing Mixture
3	Sai Raskar	Electric Field Distribution in a “Hybrid” RF Discharge with Ionization Generated by Ns Discharge Pulses
4	Madeline Vorenkamp	Plasma Assisted Deflagration to Detonation Transition in a Microchannel
5	Shurik Yatom	Interconnections Among Pulse and Plasma Properties and Chemical Reactions in Gas-liquid Plasma
6	Foluke (Jennifer) Ganzallo	Degradation of Poly- And Perfluoroalkyl Substances (PFAS) in a Plasma Spinning Disk Reactor
7	Kseniia Konina	Low-Temperature Plasma Interaction with Rectangular Microchannels
8	Jordyn Polito	Reaction Mechanisms for Atmospheric Pressure Plasma Treatment of Organic Molecules in Solution
9	Ketong Shao	Density Functional Theory Coupled with Microkinetic Model to Study Plasma-Catalyst Interactions in Ammonia Synthesis
10	Hamzeh Telfah	Kinetics of HO_2 Radical Formation and Decay in Ns Pulse O_2-He Plasmas over a Liquid Water Surface
11	Brian Bayer	Reactor Setup and Experimental Methods to Deduce Reaction Pathways and Timescales in Atmospheric Pressure Plasma Catalysis
12	Joshua Morsell	Propagation of Surface Ionization Waves on Dielectric Substrates with Simple Patterns

Abstracts - Oral Presentations

[I-1] Pathways and Timescales Involving Radical and Vibrationally Excited Species for NH_3 Formation by Low Temperature, Atmospheric Pressure Plasma Catalysis

Brian Bayer [a], Peter Bruggeman [b] and Aditya Bhan [a]

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Collisions between gas molecules and high-energy electrons produce an array of excited and radical species that can create new pathways for molecular conversion by heterogeneous catalysis. Here, we enumerate radical and excited species produced in a radiofrequency-driven atmospheric pressure plasma jet and determine their reaction pathways in the context of NH_3 synthesis by plasma catalysis. The plasma jet produces measurable, controllable fluxes of N, H, and $N_2(v)$ to the inlet of a catalyst bed placed downstream of the jet. We demonstrate that reactions involving N are responsible for NH_3 formation over Fe, Ni, and Ag metal wools through correlation between the quantity (Figure 1a) and timescale (Figure 1b) for consumption of N and formation of NH_3. For conditions with sufficient H_2 and H, ~100% of N reacts to produce NH_3 over Fe, Ni, and Ag (Figure 1a). NH_3 yield from N drops by at least 50% in the absence of H, indicating that H is necessary to selectively produce NH_3 from N. Though gas-phase NH_3 formation also occurs via radical pathways, we show that NH_3 formation is both faster and more selective when catalyst is present for identical fluxes of reactive species.

$N_2(v)$ is more abundantly produced than N by at least an order of magnitude and is consumed in the packed bed, but we do not observe NH_3 formation from $N_2(v)$. We show that loss of $N_2(v)$ in the packed bed corresponds well with a model that describes loss of $N_2(v)$ by surface-mediated vibrational quenching. We compare rate constants for vibrational quenching that fit our data with rate constants for dissociative adsorption from literature [1] and observe that rate constants for quenching are larger than rate constants for dissociative adsorption at T = 400 K below v = 10 for Fe, below v = 19 for Ni, and below v = 36 for Ag. For the densities investigated in this study, vibrational levels greater than or equal to 10 account for only ~10% of $N_2(v>0)$, indicating that the majority of $N_2(v)$ that is produced by the plasma is unreactive for catalytic reactions.

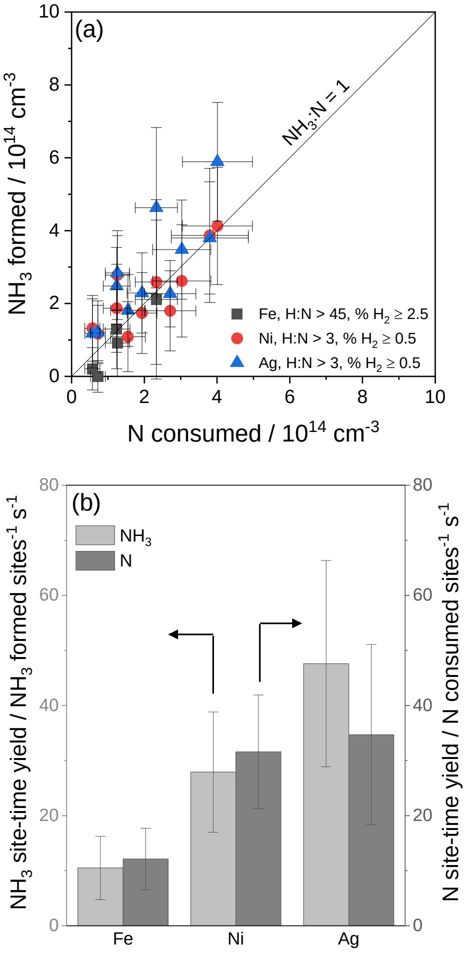

Figure 1 – (a) correlation between quantities of NH_3 formed and N consumed over Fe, Ni, and Ag for a range of process conditions. (b) comparison between site-time yields for NH_3 formation and N consumption over Fe, Ni, and Ag.

Reference

[1] P. Mehta et al., Nat. Catal., **1**, 269-275 (2018)

[I-2] Electrical Discharges in a Bubble Column Reactor: A Novel High Throughput Reactor Design for Water Treatment

Mikhail Vasilev, Jennifer Ganzallo and Selma Mededovic Thagard

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(vasilem@clarkson.edu, ganzalfj@clarkson.edu, smededov@clarkson.edu)

Plasma-based water treatment is a potential alternative for conventional wastewater purification techniques. However, an efficient implementation of the plasma technology on an industrial scale is challenging due to inherent multiphase nature of the process. In general, contaminant removal efficiency strongly depends on the plasma-liquid contact area and the highest treatment efficiencies are observed in reactors with high area-to-volume ratios, e.g., designs utilizing liquid atomization or thin water films. Overall performance of such designs, however, is limited by low liquid throughput.

This work investigates contaminant removal performance of a tubular bubble column reactor with a concentric rod-cylinder electrode configuration. A surfactant, rhodamine B dye, and a non-surfactant, caffeine, were used as model contaminants (Figure 1). The effect of hydrodynamic regimes on degradation was investigated by varying the gas flow rate, the initial bubble size, the characteristic reactor dimension, and surface tension of the treated solution. The effect of discharge properties on degradation was assessed by varying the input energy and the operating gas composition. Ultimately, we show that continuous sparging of gas is a superior method for contact area enhancement for the applied electrode configuration that results in competitively high removal rates and yields high degradation energy efficiencies. Future directions for the reactor scale up are also discussed.

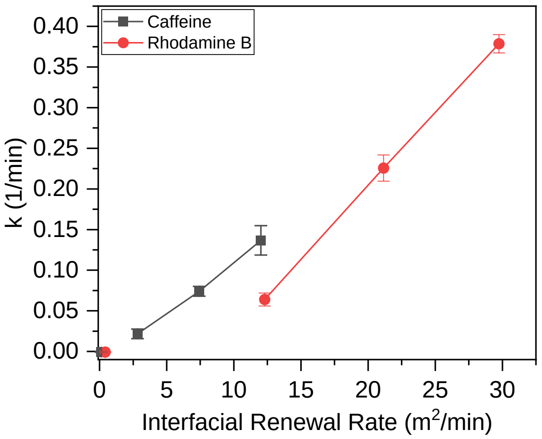

Figure 1 – Removal of caffeine and rhodamine B dye in the bubbling reactor as a function of the liquid interfacial (surface) renewal rate.

[I-3] Towards Manufacture of Green Fertilizer via Low-Temperature Plasmas

Ali Mesbah

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Nitrogen (N) fixation, essential to manufacturing fertilizers, plastics and pharmaceuticals, is almost exclusively performed via the Haber-Bosch (HB) process for ammonia (NH_3) synthesis. The HB process requires high temperatures and pressures, and utilizes hydrogen produced from methane steam reforming, which is extremely energy intensive. Over 80% of NH_3 produced is used for fertilizing agricultural crops, which sustain food demands of nearly 50% of the world's population. Despite dramatic advances in HB energy efficiency, the HB technology is inherently centralized and capital intensive, which makes it ill-posed to take advantage of the distributed nature of alternative feedstocks and renewable energy for N-based fertilizer manufacture.

Our research group is investigating the development of a new modular process that is intended to take advantage of the geographically distributed production of biowaste (e.g., manure) on dairy farms and the availability of renewable electricity from solar for distributed manufacturing of organic fertilizer at the point-of-use in remote locations. Our research is focused on exploring intensification of bioprocessing and non-thermal plasma processing in one modular unit to enrich biowaste at the point-of-production via capturing and recycling NH_3 emitted from biowaste. In this talk, we will discuss our recent experimental results on plasma processing of biowaste and present our vision for developing a bio-process that can be deployed on dairy farms for renewable fertilizer manufacture.

[I-4] Advancing Diagnostics for *In Situ* Measurements of Plasma Interactions with Complex Interfaces

Jingkai Jiang, Tanubhav Srivastava, Jianan Wang and Peter Bruggeman

Department of Mechanical Engineering, University of Minnesota, 111 Church St SE, Minneapolis,
Minnesota, 55455, USA (pbruggem@umn.edu)

The unique non-equilibrium conditions of low temperature plasmas enable the delivery of highly reactive species to substrates. Advances in the generation and control of atmospheric pressure plasmas led to the emergence of many new applications but also to several new scientific questions and major challenges for diagnostics. While plasma-surface interactions at low pressure are often due to energetic ion etching, at atmospheric pressure, the ion mean free path is much smaller than the sheath thickness resulting in insufficient ion energies. Hence, at atmospheric pressure, neutral reactive species often play an important role in plasma-surface interactions. In addition, sheaths and boundary layers at atmospheric pressure occur on length scales of 100 µm or less. We will discuss these unique challenges in the context of plasma catalysis and plasma-liquid interactions.

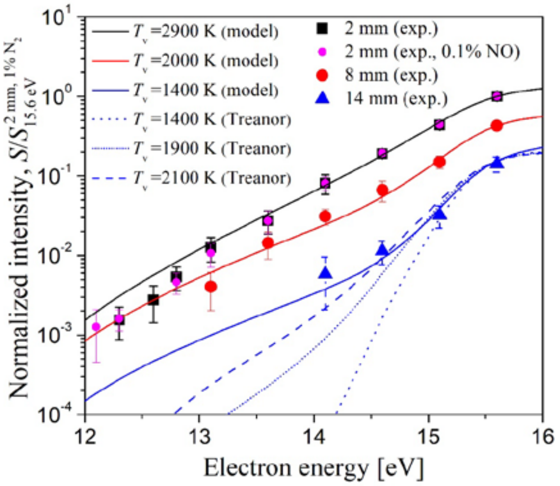

Figure 1 – MS signals of N_2^+ normalized by the measured signal for 2 mm at an ionizer electron energy of 15.6 eV in an Ar + 1 % N_2 RF plasma jet and electron impact ionization cross-sections of $N_2(v)$ as a function of the electron energy in the ionizer. The plasma conditions are: feed gas – Ar (1 slm) + 1% N_2. The indicated distance in the figure represents the distance from the nozzle of the jet to the substrate. Fitting of the MS spectra with the Treanor distribution up to its minimum and the calculated VDF`s by the 0-D kinetics model are included. In all cases, the vibrational temperatures correspond to the slope of the VDF at v=0.

Molecular beam mass spectrometry enables directly linking the obtained fluxes with plasma-surface interaction studies, owing to its ability to detect a large range of different species and to measure species fluxes or densities at a substrate. We have recently extended the capability of MBMS for measuring vibrationally excited nitrogen $N_2(v)$ which is suggested to play a major role in plasma catalysis of ammonia synthesis. We showed the capability of MBMS to measure vibrationally excited species of N_2 near interfaces when the vibrational distribution function is known or assumed. The experimental result shows also excellent agreement with state-to-state kinetic models of $N_2(v)$ developed by Prof. Adamovich’s group as shown in Figure 1 [1].

While MBMS is an excellent tool to study plasma-surface interactions for solids, it cannot be readily applied to the study of interaction of plasmas with liquids. We will show examples of spatially resolved OH radical density measurements near liquid droplets by laser induced fluorescence enabling the quantitative study of plasma-liquid interactions. Furthermore, we extended *in situ* diagnostics into the liquid phase through the adaptation of a falling liquid film plasma reactor developed by the group of Prof. Frontiera at the University of Minnesota enabling the *in-situ* measurement of the temporally resolved conversion of various compounds using optical absorption spectroscopy. This system enables for the first time the investigation of near interfacial reactions in the liquid phase uniquely allowing us to link liquid phase with gas phase plasma processes while providing new insights in plasma-liquid interactions.

Reference

[1] J. Jiang, C. Richards, I. Adamovich and P. J. Bruggeman, Plasma Sources Sci. Technol. (in review).

[II-1] Princeton Collaborative Low Temperature Plasma Research Facility (PCRF): Status Update and New Solicitation of User Proposals

Yevgeny Raitses [a], Igor Kaganovich [a], Mikhail Shneider [b], Sophia Gershman [a], Arthur Dogariu [b], Shurik Yatom [a], Anatoli Morozov [b], Nirbhav S. Chopra [a] and Willca Villafana [a]

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(b) PCRF, MAE Department, Princeton University (adogariu@princeton.edu)

The Princeton Collaborative Low Temperature Plasma Research Facility (PCRF) [1] is a collaborative research facility providing expertise and instrumentation for comprehensive characterization of low temperature plasmas (LTPs) with the goal of advancing methods of predictive control of LTP. PCRF collaborative users have access to the state-of-the-art research capabilities, including advanced plasma diagnostics (e.g. Laser-Induced Fluorescence (LIF), Two-Photon Absorption LIF, Thomson scattering, Electric Field-Induced Second Harmonic Generation, Hybrid Coherent Anti-Stokes Raman Scattering), a variety of plasma sources, computational codes (e.g. 2-D and 3-D Particle-in-Cell codes and fluid codes), and theory support. Since its launch in 2019, about 70 users from the plasma and a broader scientific community have been awarded runtime at the PCRF. The projects cover: i) plasma-liquid and plasma-solid interactions, ii) plasma transport, iii) collective phenomena in LTP, iv) use of LTP in modern applications, including plasma processing and synthesis of materials, plasma medicine, plasma-assisted catalysis, sustainability, and aerospace. In this presentation, we will discuss PCRF research capabilities, recent user projects and opportunities for collaboration.

New Soliciation of Collaborative Research Proposals

Call for proposals opens: October 10th, 2022

Call for proposals closes: December 16th, 2022

External Review: ~1 month

Notification of Principal Investigators: by February 3,

This work is supported by the US Department of Energy through contract DE-AC02-09CH11466.

Reference

[1] http://pcrf.pppl.gov

[II-2] Initiation of a Nanosecond Discharge in Water with Gas Bubbles

Sophia Gershman [a], Nicholas Sponsel [b] and Katharina Stapelmann [b]

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(b) North Carolina State University

Intentionally introduced or naturally occurring, bubbles are ubiquitously present in liquids sustaining electrical discharge such as for example, in plasma machining, failing dielectrics, water purification, or testing the compositions of liquids. The discharge in these complex systems can initiate at the electrodes, in bubbles attached to the electrodes, or in bubbles floating in the liquid depending on the rate of rise and the duration of the applied voltage.[1]

Fast imaging and precise timing[2] allows us to investigate the initiation and development of a nanosecond pulse discharge in deionized water with a quasi-stationary gas bubble between two electrodes submerged in deionized water in a pin-to-pin and pin-to-plane configurations (Figure 1). The applied voltage pulse is an unmatched 20 kV with a 7 ns rise time and ~30 ns duration of the first pulse followed by decaying oscillations lasting approximately 500 ns. When observed on a time scale of hundreds of nanoseconds or longer, the discharge appears to bridge the gap between the electrodes, while imaging on a nanosecond scale time-resolved from the applied voltage rise time (7 ns), through the end of the first pulse (~30 ns), and longer (>50ns) reveal a different story.

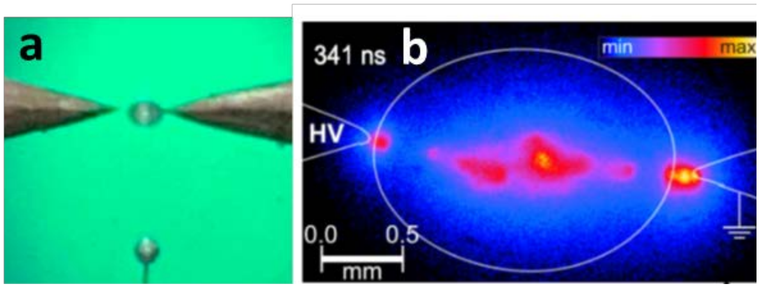

Figure 1 – (a) A backlit “quazi-stationary” bubble between two sharp electrodes; (b) image of light emitted at the electrode tips and in the bubble . ICCD gate of 150 ns captures repeated applied pulses..

We found that in positive and negative applied voltage polarities, discharge initiates in the water at the tip of the anode. The discharge in the water rapidly extends (~ 10^4m/s) toward the apex of the bubble and glowing regions begin to form inside the bubble. The steep rate of rise of the applied voltage (dV /dt < 4 kV/ns) and the short pulse duration for the development of discharge in the water suggest that cavitation is a likely mechanism for discharge initiation and propagation in water.1 In addition, the short duration of the applied voltage pulse results in only a partial Townsend discharge inside the bubble. The timing of the appearance of the discharge in the bubble depends on the distance between the bubble apex and the electrode tip. Imaging with delay times of 10 - 100 ns extending to the subsequent repeated voltage pulses captures repeated partial discharges in the same quasi stationary bubble (Figure 1 b).

This work was performed at the Princeton Collaborative Research Facility (PCRF) supported by the US Department of Energy through contract DE-AC02-09CH11466.

References

[1] M. N. Shneider and M. Pekker, Liquid Dielectrics in an Inhomogeneous Pulsed Electric Field (Second Edition), 2053-2563 (IOP Publishing, 2019).
[2] N. Pillai, N. L. Sponsel, K. Stapelmann, and I. A. Bolotnov, J. of Fluids Eng. 144 (2022).
[3] Nicholas L. Sponsel, Sophia Gershman, Maria J. Herrera Quesada, Jacob T. Mast, and Katharina Stapelmann, J. Vac. Sci. Technol. A 40 … (2022).

[II-3] EDIPIC-2D, an Open-source Versatile and Comprehensive Particle-In-Cell Code for Low-Temperature Plasmas Modeling

Willca Villafana [a], Dmytro Sydorenko [b], Alexander V. Khrabrov [a], Igor D. Kaganovich [a], Sierra Jubin [a], Andrew Tasman Powis [a], Stéphane Ethier [a], Svetlana Selezneva [c], David Smith [c], Shahid Rauf [d], Haomin Sun[a,e], Jian Chen[f]

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(b) University of Alberta
(c) General Electric Global Research Center
(d) Applied Materials Inc
(e) University of Science and Technology of China, Hefei, Anhui, China
(f) Sun Yat-sen University, China

Low-temperature plasmas are used in a wide range of industrial applications and are often subject to kinetic effects. A careful study of these non-equilibrium phenomena can be addressed with a Particle-In-Cell (PIC) technique. Here, we present recent applications of such an approach with the open-source 2D PIC code, EDIPIC [1].

Successor of a previous 1D version [2], EDIPIC-2D is first used to model a plasma-generated electron beam system for plasma processing applications. Based on a 2D axisymmetric geometry, the model examines the creation and evolution of a partially magnetized plasma in low-pressure (10 – 40 mTorr) Ar gas on injection of an energetic electron beam (2 keV). The scaling of the transport of charged particles is assessed with a large parametric study with different values of the neutral gas density, beam current, and magnetic field strength. The overall plasma parameters are found consistent with theoretical results.

In a second example, EDIPIC-2D is used to model a hollow cathode. Hollow cathodes are efficient plasma sources that are present in a wide variety of applications including electric propulsion, and surface processing studies. Current studies of hollow cathodes, which are often based on a fluid or hybrid approach, rely on semi-empirical data and so need to be adjusted for each new configuration. Besides, the large mean free path downstream of the orifice continues to challenge any fluid model. To improve our understanding of the plume, EDIPIC-2D models the near cathode exit with a 2D axisymmetric geometry and investigates the impact of the orifice size on the plasma dynamics.

In a third example, an in-depth study of beam-plasma interaction was performed [3,4].

Acknowledgments

Theoretical work on beam-plasma interaction [3,4] was performed at the Princeton Collaborative Research Facility (PCRF) supported by the US Department of Energy through contract DE-AC02-09CH11466. The code development was funded by the PPPL LDRD program and collaborations with Applied Materials by CRADA. The work presented here was funded in part by the Advanced Research Projects Agency (ARPA-e), US Department of Energy, under Award Number DE-AR0001107.

References

[1] https://github.com/PrincetonUniversity/EDIPIC-2D

[2] D. Sydorenko, "Particle-in-Cell Simulations of Electron Dynamics in Low Pressure Discharges with Magnetic Fields." University of Saskatchewan Ph. D. Thesis. 2006.

[3] H. M. Sun, J. Chen, I. D. Kaganovich, A. Khrabrov, and D. Sydorenko, Phys. Rev. Lett. **129**, 125001 (2022).

[4] H. M. Sun, J. Chen, I. D. Kaganovich, A. Khrabrov, and D. Sydorenko, Phys. Rev. E **106**, 035203 (2022).

[II-4] On the Role of Osmotic Pressure in the Interaction of Plasma with Cells in a Physiological Saline

Mikhail N. Shneider

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(m.n.shneider@gmail.com)

When non-equilibrium plasma generated by various sources interacts with a physiological solution in a Petri dish with cell cultures, visible changes in cell morphology and dielectric properties of cell membranes occur. All these phenomena can be unambiguously explained by the change in osmotic pressure [1,2]. Since cell membranes are permeable to water molecules and poorly permeable to solvated ions, a change in the composition of ions in a physiological solution leads to a change in the osmotic pressure on the cell membrane. This, in turn, leads to a stretching or contraction of the membrane, depending on the ratio of total external and internal pressures. As a result of change in the osmotic pressure in the cells in the solution, different regimes are possible: hypotonic, leading to an increase in the observed size of cells (swelling) or hypertonic, in which the cell shrinks and deforms.

If the mechanical properties of the membranes in deceased (cancerous) and healthy cells are different, then a change in the osmotic pressure drop can lead to different physiological consequences. In experiment it will manifest as a selective action of non-equilibrium plasma on different cell cultures. Since the modulus of compression of membranes in diseased cells exceeds that of membranes in healthy cells, it should be expected that the decrease in the dielectric constant of membranes in sick cells due to water displacement should be greater than the decrease in the dielectric constant in healthy cells.

To confirm the mechanisms of selective action of non-equilibrium plasma on cell cultures, it is very important to measure the dielectric permittivity of the membranes and its variation due to changes in the osmotic pressure in various cells, in particular, healthy and cancerous. This kind of research is being proposed by Prof. Vandana Miller as a future PCRF project for theoretical modeling by the author and experimental support by Dr. Sophia Gershman.

References

[1] M. N. Shneider and M. Pekker, J. Appl. Phys. **123**, 204701 (2018)
[2] M. N. Shneider and M. Pekker, Plasma Res. Express **1**, 045001 (2019)

[II-5] Electron Beam Generated ExB Plasmas and Their Applications

Yevgeny Raitses [a], Nirbhav S. Chopra [a], Shurik Yatom [a], Santosh Kondeti [a], Arthur Dogariu [b],
Ivan Romadanov [a], and Kai-Mei Fu [d]

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Low temperature plasmas generated by electron beams (e-beams) are promising for applications requiring efficient generation of ions and radicals in low pressure environments ranging from hundreds of microTorr to hundreds of mlliTorr [1–4]. For example, it was recently shown that an e-beam generated plasma with applied crossed electric and magnetic fields (ExB) operating with argon-hydrogen gas mixture in the several mTorr pressure range can generate fluxes of hydrogen atoms and ions, enabling low damage hydrogenation on an atomically thin 2D material such as graphene [4] and hydrogen passivation of diamond substrates for quantum sensor applications [5]. In this talk, we will compare plasma properties and stability of e-beam generated ExB plasmas produced by different electron sources, including an ion-induced secondary electron emission (iSEE) cathode, a thermionic cathode and an RF plasma cathode. The focus will be on electron and ion kinetic properties, chemical composition and characteristic instabilities generating dynamic and stationary plasma structures. In the described experiments, electrostatic probes, optical emission spectroscopy, ns and CW Laser-Induced Fluorescence (LIF) and fs-Two-Photon Absorption LIF (TALIF) diagnostics were used to characterize the plasma (e.g. Fig. 1). In addition, ex-situ material evaluation of two-dimensional nanomaterials and diamond substrates were used to characterize plasm-induced effects (e.g. damage) and processing (e.g. hydrogenation, passivation, and cleaning) on these ion-sensitive materials. The e-beam ExB plasma sources with all three cathode types, which span different pressure ranges and are suitable for the generation of various plasma chemical compositions, are now offered to users and collaborators interested in materials processing projects at the Princeton Collaborative Plasma Research Facility (PCRF, http://pcrf.pppl.gov).

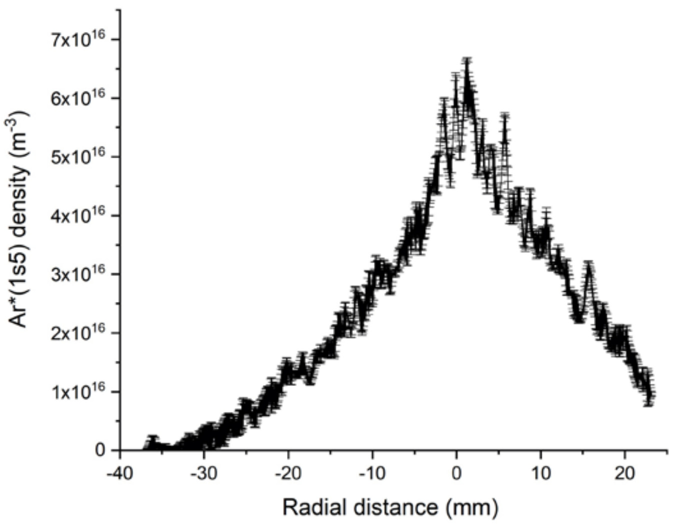

Figure 1 – Radial density of argon metastables (1s$_5$) measured by ns-LIF in an ExB plasma source with iSEE cathode operating at 4 kV, 60 mTorr Ar, 150 Gauss.

This work was performed at the Princeton Collaborative Research Facility (PCRF) supported by the US Department of Energy through contract DE-AC02-09CH11466.

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[II-6] Advanced Optical Diagnostics for Low Temperature Plasmas; Imaging Solvated Atoms with fs-TALIF

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The talk will first present recent developments and results using advanced optical diagnostics in LTP plasmas at the DOE Princeton Collaborative Research Facility (PCRF). Some of the results include non-equilibrium temperature measurements in nanosecond discharge plasma (with T. Ombrello, AFRL), measurements of electric fields in self-pulsating streamer coronae (with C. Guerra-Garcia, MIT), and demonstrating a high temporal resolution imaging technique using femtosecond Two-Photon Laser Induced Florescence (fs-TALIF) (with Z. Zhang, Univ. of Tennessee).

We will then present direct measurements of atomic species solvated in water at the interface with a plasma jet. Measuring the density of reactive species using direct fluorescent emission in interfacial plasmas is challenging due to the fast quenching of the excited atomic species at and below the gas-liquid interface. Exciting atomic species in a liquid using femtosecond pulses allows for fluorescent emission on time scales comparable or smaller than those allowed by the fast collision rates in a liquid. Using femtosecond two-photon absorption laser induced fluorescence (fs-TALIF) we have succeeded to directly image with high spatial resolution the atomic oxygen created by an atmospheric plasma jet impinging upon water, both above and below the gas/liquid interface. The same resonant UV two-photon excitation of O atoms followed by fluorescent emission in the liquid allowed us to determine that atomic oxygen persists for tens of microseconds in water, penetrating hundreds of micrometers into the liquid (see Fig. 1). The high spatial and temporal resolution of fs-TALIF imaging in liquids opens the door for studying reaction rates of reactive species in interfacial plasmas.

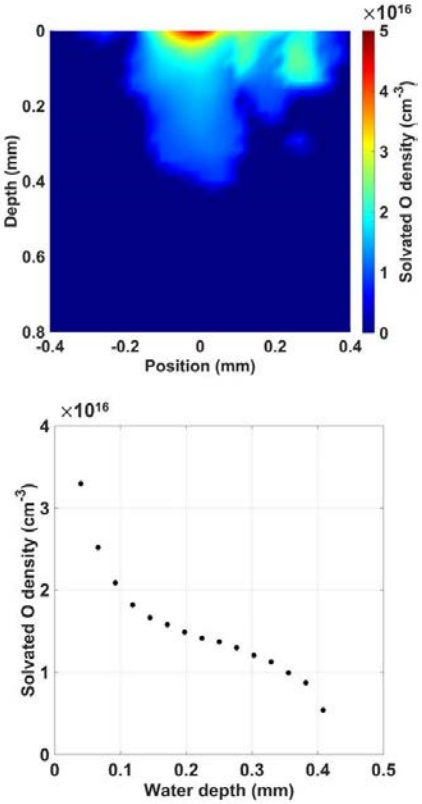

Figure 1 – (top) O TALIF signal below the water surface where the depth denotes the distance to the air/water. (bottom) Atomic oxygen density as a function of depth in the liquid.

Acknowledgements: Arthur Dogariu is supported by the Princeton Collaborative Research Facility (PCRF) under Contract No. DE-AC02-09CH11466 with U.S. Department of Energy (DOE).

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[III-1] Hybrid Ns Pulse / Capacitively Coupled RF Discharges for Plasma Chemistry and Plasma Catalysis

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Control of energy partition in nonequilibrium gas discharge plasmas is critical for applications ranging from energy-efficient plasma chemical conversion to plasma-enhanced catalysis. In many of these applications, high efficiency is achieved when the discharge is operated near the optimum electron impact excitation of vibrational and low-energy electronic states of major molecular species, such as N_2, H_2, O_2, and CO_2. This occurs in DC glow discharges, capacitively coupled RF discharges, and microwave discharges. However, scaling these discharges to high pressures, high input powers, and large volumes is challenging, due to the plasma instability development. The use of a train of high peak voltage, ns duration pulses generated at a high repetition rate extends the range of stable operation to significantly higher pressures and peak powers. On the other hand, this approach greatly reduces the input energy fraction into the excitation of molecular vibrations, due to high peak reduced electric field, E/N. In this work, we employ a non-self-sustained "hybrid" discharge, where the ionization is generated by a ns pulse train and the energy is coupled to the plasma by a sub-breakdown RF waveform, using a single pair of electrodes external to the discharge cell. This approach suppresses the cathode layer instability development and precludes the catalytic reactions on the electrodes. It is shown that the use of the hybrid ns pulse / RF waveform enhances the vibrational excitation of N_2, CO_2, and CO. The hybrid plasma kinetics is quantified using the measurements of the electric field, vibrational temperatures of the molecular species, and the yield of plasma chemistry products.

Electric field in a capacitively coupled, non-self-sustained RF discharge in nitrogen, with external ionization generated by high-voltage ns pulses, is measured by ps Electric Field Induced Second Harmonic generation. The combined ns pulse / sub-breakdown RF waveform generates a diffuse plasma between two plane dielectric-covered electrodes. The measurements are made in the bulk of the plasma and in the sheaths. Reduced RF electric field in the sheaths is significantly higher compared to that in the plasma, exceeding 100 Td, due to the displacement of the electrons by the drift oscillations and the formation of space charge layers. Peak reduced RF electric field in the plasma is significantly lower, from 10 to 40 Td, indicating the efficient vibrational excitation of N_2 by electron impact. The hybrid ns pulse / RF plasmas are used for the selective vibrational excitation of N_2 and CO_2, to study the CO_2 dissociation kinetics in CO_2-N_2 and the plasma-catalytic ammonia generation in N_2-H_2. Vibrational populations of N_2 are measured by CARS. Time-resolved vibrational populations of CO_2 and CO, translational-rotational temperature, and CO number density are measured by mid-IR, tunable Quantum Cascade Laser Absorption Spectroscopy. The NH_3 number density is measured by FTIR absorption spectroscopy. The results identify the effect of the targeted vibrational excitation on the process yield.

The kinetic modeling predictionsindicate that the targeted vibrational excitation of molecular species in a non-self-sustained RF discharge with external ionization is scalable to high coupled power densities, up to ~100 W/cm^3. Scaling the electrode surface area would make feasible sustaining multi-kW "hybrid" ns pulse / capacitively coupled RF plasmas, with independent control of ionization and energy coupling. This approach can be extended to efficient vibrational excitation of other molecular species where vibrational relaxation is relatively slow, such as CO, CO_2, and H_2.

[III-2] Plasma Assisted Combustion and Manufacturing

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In the year of 2022, we have made several progresses in plasma modeling, plasma assisted combustion and detonation, plasma thermal chemical instability with cool flame chemistry, in situ plasma diagnostics, plasma aided chemical looping and materials manufacturing.

In non-equilibrium plasma aided combustion, we have developed a two-dimensional multi-scale adaptive reduced chemistry solver (MARCS-PAC) to include detailed combustion and plasma chemistry and transport properties. The solver was used to study the effects of non-equilibrium excitation and electrode geometry on H_2/air ignition in a nanosecond plasma discharge [1]. In addition, the solver was used to investigates the effects of non-equilibrium nanosecond plasma discharge pulse repetition frequency, pulse number, and flow velocity on the critical ignition volume, the minimum ignition energy, and chemistry in a plasma-assisted H_2/air flow [2]. Furthermore, we experimentally investigated the plasma discharge effect on the transition from deflagration to detonation. We found that detonation transition can be significantly accelerated or delayed by controlling plasma discharge [3].

To understand the mechanism of plasma instability in a reactive flow at low temperature, the cool flame chemistry enhanced plasma thermal-chemical instability was examined in a dimethyl-ether (DME) with a repetitive nanosecond pulsed dielectric barrier discharge. It is found that such nonlinear development of the thermal-chemical instability is controlled by the competition between plasma-enhanced low-temperature heat release and the increasing thermal diffusion at higher temperature [4].

In plasma diagnostics, we developed a sensitive two beam hybrid femtosecond/picosecond pure rotational coherent anti-Stokes Raman scattering (fs/ps CARS) method to simultaneously measure the rotational and vibrational temperatures of diatomic molecules [5]. We developed a femtosecond ultraviolet laser absorption spectroscopy (fs-UV-LAS) and cavity ringdow spectroscopy for simultaneous in-situ measurements of temperature and species [6]. The results show that the temperature and OH concentration could be measured with accuracy enhanced by 29~88% and 58~91%, respectively, compared to the two-narrow-line absorption methods.

In plasma assisted chemicals and materials manufacturing, we studied the effect of non-equilibrium plasma in chemical looping of methane and ethylene with metal oxides [7]. It was shown that plasma can change the reaction pathways and significantly enhance fuel oxidation and chemical looping at low temperature. We also demonstrated by controlling plasma instability with tip enhanced plasma, we are able to stabilize a uniform, volumetric plasma and control the plasma temperature. It is shown that this volumetric plasma can provide a novel platform to manufacturing extreme materials. Moreover, to understand how surface charge affects surface reaction in chemical manufacturing, we conducted the modeling of reaction energetics for N_2 and H_2 dissociative adsorption on catalyst surfaces using DFT calculations. The results showed that the surface may significantly change the relationship between the adsorption energy of H-atom with respect to the adsorption energy of N-atom on a single metal atom catalyst.

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[III-3] Issues of Effectiveness and Energy Efficiency in Plasma-assisted Catalysis for Ammonia Synthesis

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Plasma-assisted catalysis utilizes excited gaseous molecules or new reactive species formed in a non-equilibrium, low temperature gas discharge plasma, along with a catalyst, to enable increases in the activity and selectivity for carrying out desirable chemical reactions at lower temperature and pressure conditions than commonly utilized in commercial thermal heterogeneous catalysis. Plasma chemical reactors also take advantage of the expanding increase of renewable electricity from solar and wind power rather than burning carbon for the synthesis of fuels and chemicals. The synthesis of ammonia (NH_3) from N_2 and H_2 is one of the most important industrial catalytic reactions, and also one of the simplest. Thus, this process is important to study for fundamental insights into the important issues of effectiveness and energy efficiency in plasma-assisted catalysis.

Our recent research has focused on addressing several fundamental questions. What can we learn from simple modeling work about the dominant reaction pathway in plasma-assisted catalysis of ammonia synthesis? Does it give any clue as to why the energy yield is low and how one can try to address it? What are the maximum attainable conversions in plasma-assisted ammonia synthesis? We report on and will discuss several aspects of our work and results from dielectric barrier discharge (DBD) reactors using AC discharges and packed with catalyst support particles or supported metal catalysts in the discharge zone.

Experiments found that there was only minimal difference in the reaction rates over γ-Al_2O_3 and Ru/γ-Al_2O_3.[1] Complementary zero-dimensional plasma kinetic model analysis was also performed using inputs from experimental data. The analysis allowed for gas phase reactions, Eley–Rideal (E–R) reactions, and direct adsorption of radical species on the γ-Al_2O_3 surface. On the metal surface, dissociative adsorption of N_2 and H_2, and Langmuir–Hinshelwood (L–H) reactions were also included. Under the conditions of our experiments, ammonia synthesis proceeds principally by the formation of reactive radicals in the gas phase, which then adsorb and participate in E–R reactions on both the metal and support material surfaces. Measurements using an electron-ionization molecular beam mass spectrometer (MBMS) that was connected directly to a plasma reactor detected NNH radicals in the gas phase, indicating that this reactive species should be considered further in the reaction kinetics.[2] N_2 conversion and plasma parameters were determined for highly porous silica particles and nonporous soda lime glass particles to probe the effects of catalyst support porosity.[3] Modeling indicates that gas-phase radicals can diffuse into the pores of the support to a small extent. Ongoing research is investigating the enhancement of N_2 dissociation due to noble gas species for plasma-assisted catalysis of ammonia production, the application of a helium collisional-radiative model for plasma diagnostics of atmospheric pressure dielectric barrier discharges, and the development and testing of in situ probes for vibrational spectroscopy at catalyst surfaces.

Overall, these results increase our understanding of plasma-catalysis hybrid systems and aid efforts to achieve the strongest synergistic interactions between the plasma and catalyst to increase performance and overall energy efficiency.

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[IV-1] Sandia's Plasma Research Facility (PRF): Status Update and New Solicitation of User Proposals

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Sandia National Laboratories is the largest of the US national labs and has extensive programs in science and energy, in addition to its original and sustained focus on national defense science and technology. Through the course of these research programs, Sandia has developed state-of-the-art experimental and modeling capabilities relevant to low-temperature plasma research. Examples of relevant diagnostics include Laser-Induced Fluorescence (LIF), Photofragmentation LIF (PF-LIF), Laser-Collision-Induced Fluorescence (LCIF), Coherent Anti-Stokes Raman Spectroscopy (CARS), Sum Frequency Generation (SFG), and Molecular Beam Mass Spectrometry (MBMS). Advanced modeling capabilities are also available, including state-of-the-art PIC-DSMC modeling tools and access to Sandia's high-performance computing (HPC) capabilities (many 10K's of cores). This presentation will provide an update on the activities and structure of Sandia's Low-Temperature Plasma Research Facility (PRF), funded by the DOE Office of Science, Fusion Energy Sciences, General Plasma Science program. The PRF is a resource available to anyone in the international Low-Temperature Plasma (LTP) community to access the advanced resources available at Sandia (not restricted to the US collaborators). Capabilities are accessed through an annual proposal process that is open and accepting proposals.

This work used the capabilities of the SNL Plasma Research Facility, supported by the DOE Office of Science, Fusion Energy Sciences. SNL is managed and operated by NTESS under DOE NNSA contract DE-NA0003525.

[IV-2] Nonlinear Optical Diagnostics Laboratory for Low-Temperature Plasma-Assisted Chemistry

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Low temperature plasmas (LTP) provide opportunities for enhancing high-value energy conversion processes in both energy efficiency and chemical selectivity. Nonequilibrium distributions of electronic and molecular rovibronic excitations as well as active speciation in non-thermal equilibrium plasmas offer unique control over the outcome of chemical reactions and process optimization. While the understanding of low temperature plasma assisted chemistry is still emerging, clear benefits have already been demonstrated in such fields as plasma assisted catalysis and plasma assisted combustion. However, improved understanding of the underlying chemical kinetic mechanisms requires new capabilities in in-situ probing of plasma-chemistry interactions.

Ultrafast nonlinear spectroscopy methods provide incisive tools for probing the chemistry and dynamics of low-temperature plasma assisted chemistry. Figure 1 demonstrates recent results from our lab in utilizing both femtosecond (fs)/picosecond CARS and fs E-FISH to study the plasma-assisted deflagration to detonation chemistry. It was further found in these experiments that the LTP induced low-temperature oxidation chemistry as shown through laser induced fluorescence measurements of formaldehyde intermediate. We have implemented a 2-beam E-FISH approach for full suppression of spurious second-harmonic signal, and further assess the effect of the Guoy phase shift on these signals.

Lastly, surface sum-frequency generation has been employed to study the plasma-enhanced ammonia synthesis reaction. Preliminary evidence for plasma-induced reactive surface N-H has been observed, encouraging further development of this approach for LTP applications such as heterodyned signal detection.

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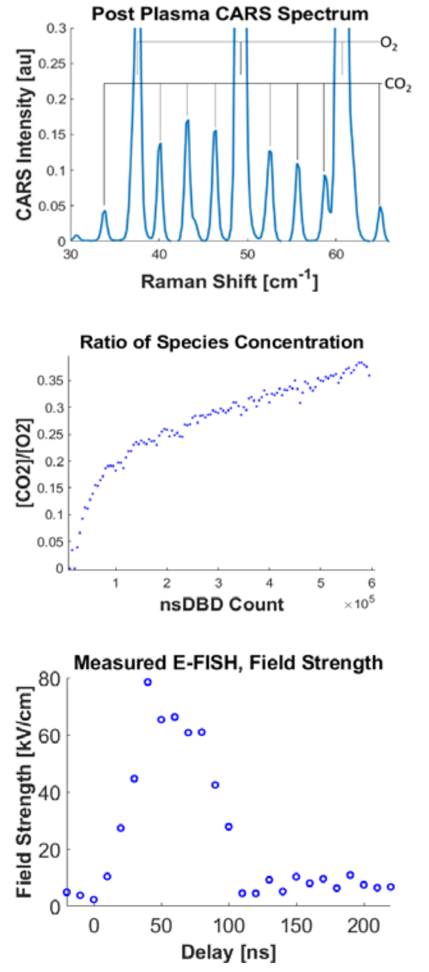

Figure 1 - (Top) Fs/ps CARS spectrum following nsDBD discharge induced conversion of DME/O_2 into CO_2. (Middle) Evaluation of the plasma-induced conversion rate. (Bottom) Electric field during plasma pulse as evaluated with E-FISH.

[IV-3] How Sheath Properties Change with Gas Pressure: Modeling and Simulation

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Here we test a 1D collisional sheath model using particle-in-cell (PIC) simulations, which include charge-neutral collisions using the direct simulation Monte Carlo method. The model was created to predict how several important sheath properties change with gas pressure: the sheath edge values of the ion velocity, relative plasma density, electric field, potential, and sheath width. Currently, there are multiple models for each property, each lacking validation. Instead of testing each model separately, we self consistently extract each property from our fluid model and compare it with the PIC simulations. We find that numerical solutions to our model and simulations agree over a large pressure range (0.01-10,000 mTorr). Both predict the ion velocity (collisional Bohm criterion) and relative density (edge-to-center density ratio) decrease with increasing pressure, while the potential and sheath width increase. The sheath edge electric field is found to remain constant in both. However, the model doesn't account for kinetic effects, like non-Maxwellian features of the electron velocity distribution and temperature gradients. We find that the former leads to small differences between the model and simulations at low pressures (<100 mTorr) and that the difference can become significant at the highest pressures (>100 mTorr). Ultimately, we derive expressions for each quantity that depend only on the pressure and adjust the coefficients to best represent the simulation data with its kinetic effects.

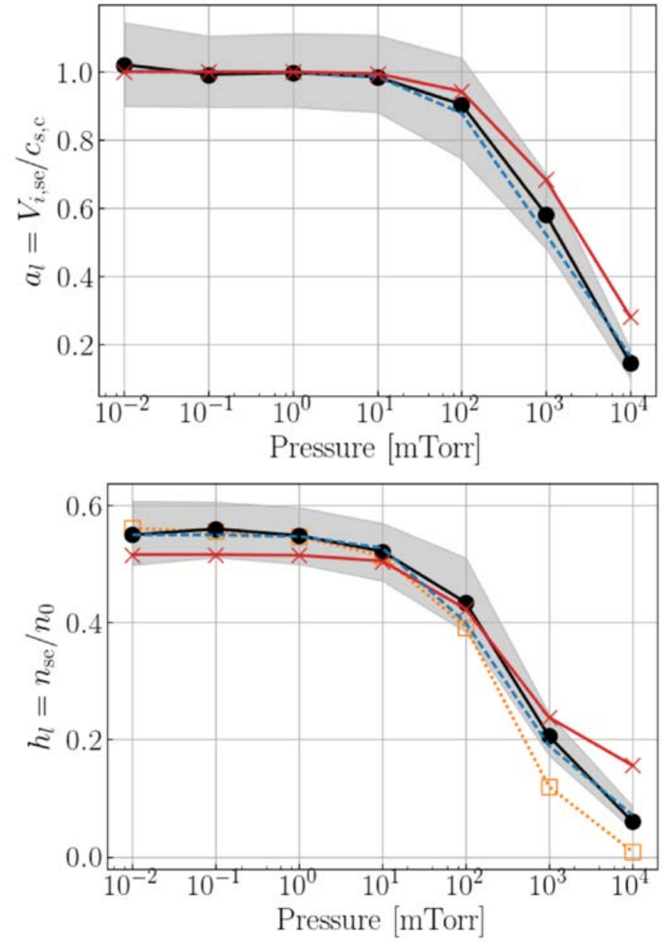

Figure 1 – (Top) Collisional Bohm criterion, a_l, measured from PIC simulations (circles) and from numerical solutions of the fluid model (crosses). (Bottom) Edge-to-center density ratio, h_l, measured from PIC simulations (circles), predicted from numerical solutions of the fluid model (crosses), and predicted using $a_l = 1$ to define the sheath edge location (squares). Analytic expressions derived from approximations to the fluid model are shown in blue and the sensitivity of the PIC measurements to the sheath edge location are illustrated by the grey region.

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[IV-4] Imaging of Methyl Radical and Hydrogen Peroxide in Pulsed Plasmas by Photofragmentation Laser-Induced Fluorescence

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The methyl radical (CH_3) and hydrogen peroxide (H_2O_2) are key reactive species in plasma assisted processes such as combustion, catalysis, surface decontamination, and biomedical treatments. Imaging of CH_3 and H_2O_2 is challenging because they cannot be detected directly by laser-induced fluorescence. We present a technique for 2D imaging of these species in plasmas using photofragmentation laser-induced fluorescence (PF-LIF). The target molecule, CH_3 or H_2O_2, is photodissociated by a UV pump laser to produce CH or OH fragments, respectively. These photofragments are then detected with LIF imaging using an overlapping probe laser beam for LIF excitation. For CH_3 PF-LIF measurements, transitions in the B-X band of CH are excited, and fluorescence from the overlapping A-X(0,0), A-X(1,1), and B-X(0,1) bands is detected with the A-state populated by collisional B-A electronic energy transfer. For H_2O_2 PF-LIF measurements, the probe laser excites transitions in the A-X(1,0) band of OH, and fluorescence is detected from A-X(1,1) and (0,0) bands. These non-resonant detection schemes enable interrogation near surfaces.

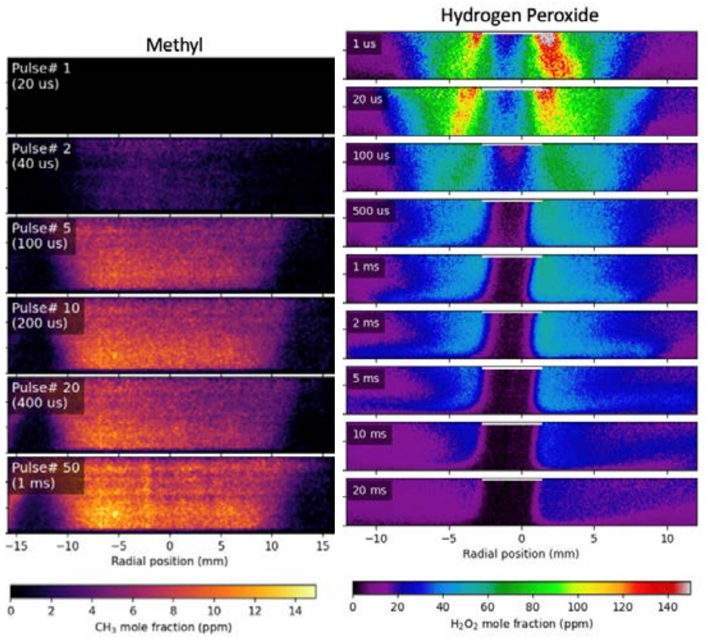

Figure 1 – PF-LIF measurements of (left) CH_3 in a plane-to-plane DBD discharge in CH_4-CO_2-He, (right) H_2O_2 in a DBD jet configuration in a He-H_2O-O_2 discharge.

We demonstrate PF-LIF imaging measurements of CH_3 and H_2O_2 mole fractions in nanosecond pulsed DBD plasmas with mixtures of CH_4-CO_2-He and He-H_2O-O_2, respectively.[1, 2] We have also initiated PF-LIF imaging measurements of the spatiotemporal evolution of methyl in a nanosecond pulsed CH_4-He plasma jet for which companion modeling efforts are underway.

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[V-1] Study of Surface Interactions for Plasma Catalytic Nitrogen Oxidation

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Cold atmospheric pressure plasma is a potential tool for performing nitrogen fixation through nitrogen oxidation. In this study we use a plasma-catalysis reaction chamber that enables quantitative and qualitative measurements of such interactions by infrared spectroscopic methods [1] and study N_2-O_2 plasma interactions with a Pt-Al_2O_3 catalyst. An $N_2/O_2/Ar$ gas stream is flown through an atmospheric pressure plasma jet to the heated catalyst in an enclosed chamber. Species on the catalyst surface are analyzed using Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS), while gas phase species exiting the catalyst bed are analyzed with Fourier Transform Infrared Spectroscopy (FTIR). Plasma power, catalyst temperature and gas flow composition are varied to investigate plasma-catalyst interactions. Bare Al_2O_3 is analyzed as a reference material to isolate the effects of the platinum. Results reveal multiple interactions on the catalyst surface. Interaction seems to be confined to plasma produced NO_x species and catalyst. Minimal evidence of atomic species-catalyst interaction under the tested conditions has been seen. At low temperature, NO and NO_2 adsorb on the catalyst surface. DRIFTS reveals surface compounds to be nitrites and nitrates likely bonded to alumina, as well as NO bonded to Pt. Figure 1 shows downstream species build versus time at different temperature settings of the heating stage.

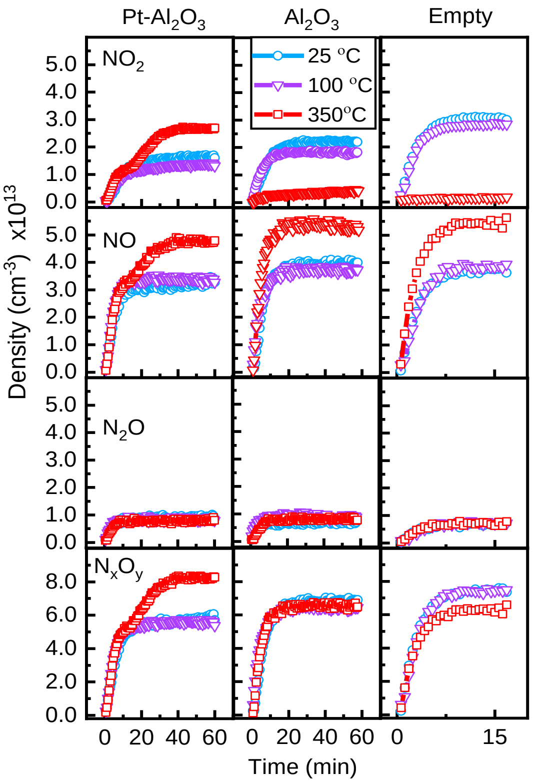

Figure 1 - Downstream gas densities vs. time for Pt catalyst, Al_2O_3 support and empty chamber at 3 different temperatures.

Upon heating to 350 °C NO_x is released from the Pt catalyst surface as NO_2 and NO. The exact temperature of product desorption shifts with operating conditions, but is limited by NO thermal desorption. DRIFTS Pt-NO results are consistent with NO desorption at high temperatures. The Pt catalyst also promotes oxidation of NO to NO_2 by interaction with O_2 compared to the Al_2O_3 case at 350 °C (Fig. 1), a known feature of platinum catalysts. Overall NO_x quantities may increase with a heated catalyst relative to empty chamber or support only, but more investigation is needed. Hysteresis loops created from downstream gas data taken during thermal cycling of the catalyst can be used to separate the plasma and surface contributions to downstream gas species production. We thank B. Bayer, Dr. A. Bhan and Dr. P. J. Bruggeman for helpful discussions. This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Fusion Energy Sciences under award number DE-SC0020232.

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[V-2] Surface Ionization Wave Propagation: Surface Permittivity and Capacitance Effects

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Variation in capacitance and material permittivity will play a significant role in the propagation of surface ionization waves (SIW's) along a dielectric surface separating the SIW from ground. Qualitatively, one would expect that as the electrical thickness of the dielectric layer increases, the reduction in capacitance per unit area will increase the rate of surface charging and result in the SIW propagation velocity increasing, eventually approaching the free ionization wave velocity as the capacitance approaches zero, and resulting in the termination of SIW propagation as the capacitance per unit area increases at very small electrical distances. [1] This has previously been explained as a dependence between SIW velocity and RC time constant for surface charge accumulation. [2]. This has been observed as the dielectric thickness has changed due to material permittivity. [3]. Between these two extremes, a monotonic increase in SIW velocity is predicted.

Experiments were performed where SIW velocity was measured at constant incident ionization wave velocities (initiated by a positive pulse 4kV nominal 500ns cylindrical APPJ) as a function of dielectric thickness over a grounded surface. For sub-millimeter electrical thicknesses, the expected monotonic increase in SIW velocity with increasing thickness is observed. As dielectric permittivity is increased, an expected decrease in the rate of velocity gain per physical thickness is also observed and is consistent with the electrical thickness dependence on dielectric permittivity. As electrical thickness continues to increase, a saturation of SIW velocity below the free ionization wave velocity is observed. This is summarized in Figure 1.

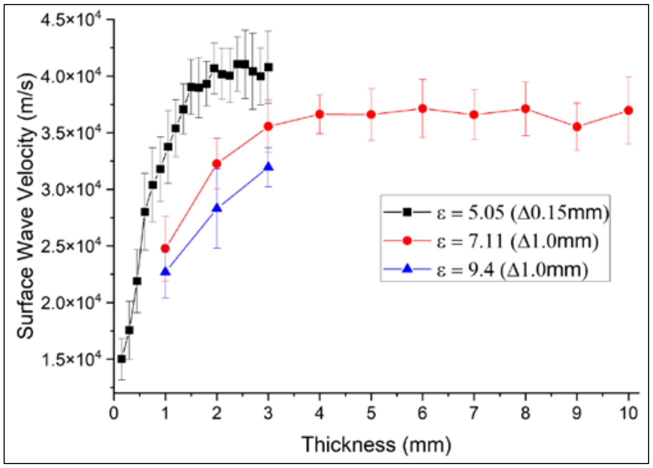

Figure 1 – SIW velocities as a function of thickness for dielectrics of varying physical thicknesses showing monotonic increase in SIW velocity to a critical thickness above which the velocity saturates below the initial free ionization wave velocity.

This velocity saturation below the free ionization wave velocity suggests that the relationship between SIW velocity and dielectric thickness is not a straightforward relationship as described in previous works, and that an explanation to the velocity saturation is needed to better understand plasma interaction with complex interfaces. In this talk, we will present the results of the initial thickness study as well as follow-up experiments that suggest that bulk permittivity and surface permittivity both contribute to SIW velocity, and attempt to quantify the critical electrical thickness that contributes to SIW propagation velocity.

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[V-3] Tomographic Optical Imaging of a Pulsed Atmospheric Pressure Plasma Jet

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Advances in laser diagnostics and models have been leveraged to investigate well-controlled idealized plasmas in 2D fashion, but the spatially complex structure in actual plasmas requires a technique that can provide a more complete 3D picture. To address this limitation, a plasma tomographic optical imaging capability has been developed at Sandia National Labs. The system includes four intensified cameras that measure eight angular projections of the light source. An algebraic reconstruction technique (ART) is used to determine the light intensity at each voxel using the method of projections onto convex sets. Initial efforts have focused on 3D optical emission imaging. The primary development challenges have included reconstruction algorithm development and achieving sufficient 3D spatial and temporal resolution to resolve features of interest. A nanosecond pulsed atmospheric pressure plasma jet is imaged as an initial demonstration of the technique. The diagnostic shows promise for investigating plasma structures and behaviors found on complex surfaces and in streamers, arcs, cathode spots, anode spots, magnetic field interactions, etc.

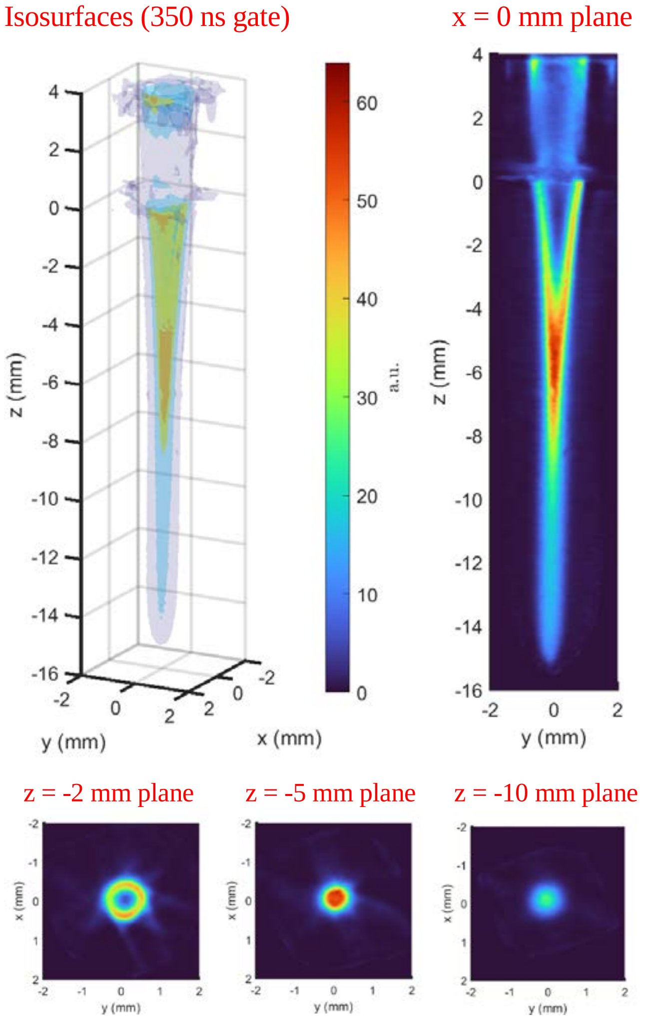

Figure 1 – Tomographic reconstructions of helium pulsed atmospheric pressure plasma jet using four intensified cameras and eight viewing angles. 3D spatial resolution is 20 um.

Acknowledgements
This work was supported by the U.S. Department of Energy Office of Fusion Energy Sciences (DE-SC0020232) and the Sandia Laboratory Directed Research and Development (LDRD) program. Sandia is managed and operated by NTESS under DOE NNSA contract DE-NA0003525.

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[V-4] Characterization of Plasmas in Contact with a Liquid Water Surface

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Characterizing and predicting the electric field distribution in plasmas in direct contact with liquid water, such as in atmospheric pressure plasma jets impinging on a water-covered substrate, is essential for understanding the dynamics of plasma propagation over the surface and the kinetics of plasma chemical reactions. In this work, time-resolved and spatially resolved distributions of the electric field over a microchannel array, alumina ceramic surface placed under an atmospheric pressure N_2-Ar plasma jet powered by a ns pulse discharge is measured by ps Electric Field Induced Second Harmonic (EFISH) generation. The measurements are made both over a dry microchannel surface and over a surface with the channels filled with liquid water. The experimental results exhibit the enhancement of the electric field inside and above the channels. This effect is observed both for the horizontal and vertical electric field components. The distribution of the electric field in the channels is controlled by the propagation of an ionization wave over the surface and is sensitive to the distance between the plasma jet and the measurement location.

Hydroperoxyl radicals (HO_2), hydroxyl radicals (OH), and hydrogen peroxide (H_2O_2) are among the major product species in low-temperature, high-pressure plasmas containing water vapor and oxygen. *In situ* measurements of the absolute, time-resolved concentrations of HO_2 and H_2O_2 in the plasma, at well-defined and reproducible conditions, would help identify the dominant plasma chemical processes of their generation and decay. The generation of H and OH by electron impact dissociation of water vapor, followed by the three-body recombination of H atoms and O_2 is a known process generating HO_2 in the plasma. Reactions between HO_2 radicals also result in the accumulation of H_2O_2, which is known to induce apoptosis (programmed cell death) in a wide range of cell cultures in plasma-treated liquids. Since one of the objectives of plasma activation of water and aqueous solutions is introducing the long-lived reaction products, such as H_2O_2, into the liquid phase, the key species measurements should be made near the liquid surface, using the water vapor as one of the key reactants. In this work, absolute, time-resolved HO_2 radicals are measured in the plasma in direct contact with liquid water, in a controlled environment, using Cavity Ring Down Spectroscopy Diagnostics.

[V-5] Atmospheric Pressure Plasma Interactions with Channels and Embedded Particles

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Atmospheric pressure plasma (APP) catalysis will include some form of plasma-surface interaction, and likely at atmospheric pressure. APPs interacting with those substrates deliver activation energy to the surface that may enhance (or delay) surface reactions. In this paper, we discuss results from experimental and computational investigations of APPs interacting with complex surfaces with the goal of determining if engineering the surface can produce control of plasma-surface interactions. The first experiment consists of fast-camera imaging of plasma jet interactions (argon or helium flowing into ambient air) with flat and profiled dielectric surfaces. The speed of surface ionization waves (SIWs) are measured as a function of dielectric thickness, voltage and surface profile. In the second experiment, electric fields are measured, using the EFISH technique, of a plasma jet (argon flowing into ambient air) producing SIWs over surfaces with square channels, both dry and water filled. The computations were conducted with the 2-dimensional modeling platform *nonPDPSIM*. The experiments were modeled, as were SIWs interacting with ideal surfaces where material properties and shapes were systematically varied. For example, APP interactions with dielectric surfaces with embedded particles having different electrical properties were investigated.

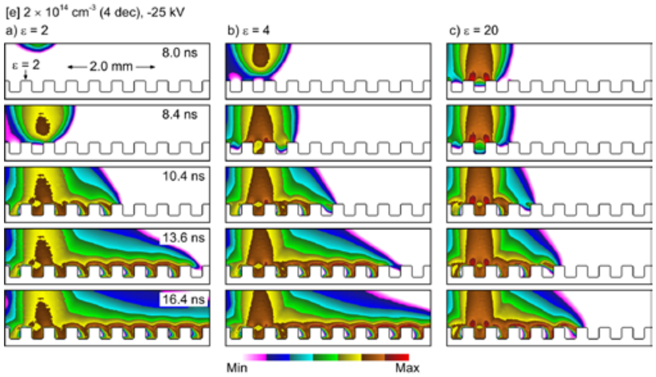

Figure 1 - Predications for electron density during the intersection of an argon plasma jet with a dielectric surface having micro-channels. The permittivity of the surface is a) $\varepsilon_r = 2$, b) $\varepsilon_r = 4$, and c) $\varepsilon_r = 20$. The ionization wave arrives at the surface sooner with large ε_r due to polarization that increases the electric field in the plasma. The SIW propagates faster with small ε_r due to the lower capacitance (F/cm^2).

Control of the speed of SIWs over dielectrics enables control of the dwell time of the plasma over the surface, which in turn controls the fluence of, for example, VUV radiation to the surface. Experiments and modeling showed that the speed of SIWs over flat and profiled surfaces is largely controlled by the capacitance (F/cm^2) of the underlying dielectric, though secondary processes such as photo-emission of electrons can influence that speed. (See Figure 1.) The local intensity of the SIWs is in large part determined by electric enhancement due to the polarization and shape of the dielectric. These processes can lead to hopping of the SIW to sites of electric field enhancement, and reverse ionization waves due to both secondary processes (photo-emission, electric field emission) and electric field enhancement. These hopping and reverse-wave phenomena, are particularly pronounced for water filled channels due, in part, to the high permittivity of the water which, through polarization, leads to large electric field enhancement. Measurements were made of the vertical and horizontal components of the electric field at the surface where a plasma jet make a transition into a SIW. During this transition, measurements and modeling show that the vertical component of the electric field diminishes and the horizontal components grows as the SIW develops, though topography dominates the local magnitude of the electric field.

Abstracts - Poster Session I

[I-1] Plasma-Assisted Chemical-Looping Combustion: Low-Temperature Methane and Ethylene Oxidation with NiO

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In this work, the low-temperature plasma-assisted oxidation of CH_4 and C_2H_4 by NiO in a heated, coaxial, dielectric barrier discharge (DBD) reactor was investigated through time-dependent species measurements by an electron-ionization molecular beam mass spectrometer (EI-MBMS) and reactive molecular dynamics simulations (ReaxFF). In the experiments, 10% methane or 10% ethylene mixed with noble gasses was flown at 50 sccm through 1g NiO particles dispersed in quartz wool under both plasma and non-plasma conditions in a fixed-bed flow reactor, as seen in Fig 1. Time-dependent gas-phase species profiles were collected through the EI-MBMS approximately every 13s of the entire mass spectra up to a molecular weight of 220u.

We observed enhancement of methane oxidation from the plasma discharge between 400 and 500 °C, with no CO_2 was recorded at such temperatures without the presence of plasma. At higher temperatures, the differences between plasma and non-plasma conditions were limited, as complete oxidation occurred without the need for plasma and with the plasma not producing significantly more or different intermediate oxygenated species. At lower temperatures, there was less oxidation and significantly more plasma reforming measured, with C_2H_4 being the chief plasma reforming species observed in the CH_4 plasmas. NiO accumulated significant carbon buildup under both non-plasma and plasma conditions with CH_4.

To gain kinetic insights into the reaction pathways that occur on the surface of NiO particles, Reactive Molecular Dynamics (ReaxFF) simulations were performed for the oxidation of CH_4 and C_2H_4 in a non-plasma environment on the ns timescale on NiO nanoparticles. The simulations were able to correctly predict the build-up of carbon on NiO. Reaction pathways for CH_4 and C_2H_4 reacting with NiO based the observed species from the MBMS spectrum in plasma and non-plasma experiments, along with the ReaxFF simulations, were created. Such pathways provide a framework from which future, more complex plasma-assisted CLC mechanisms can be crafted from.

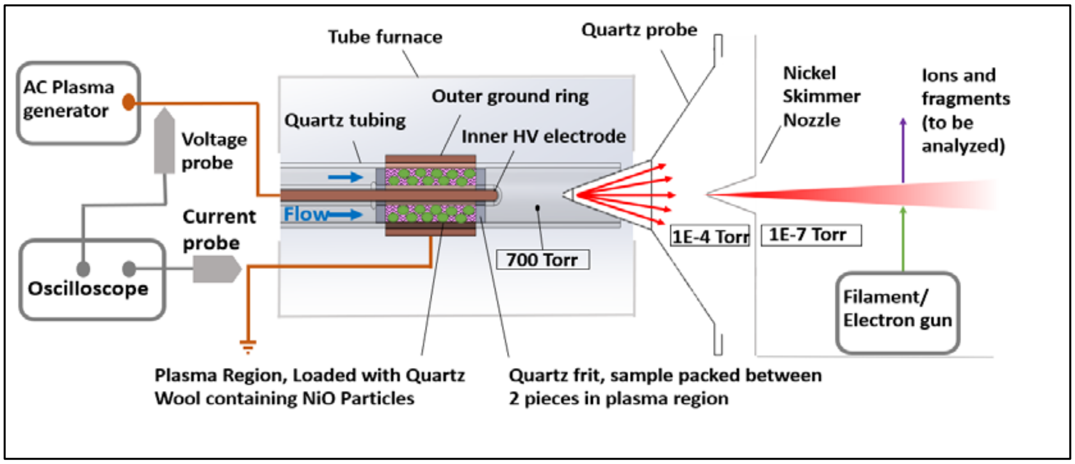

Figure 1 - Schematic of fixed bed flow reactor loaded with NiO particles.

[I-2] Sensitive and Single-shot OH and Temperature Measurements by Femtosecond Cavity Enhanced Absorption Spectroscopy

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In many low temperature plasmas (LTPs), the OH radical and temperature represent key properties of plasma reactivity. However, OH and temperature measurements in weakly ionized LTPs are challenging, due to the low concentration and short lifetime of OH and the abrupt temperature rise caused by fast gas heating. To address such issue, this work combined cavity enhanced absorption spectroscopy (CEAS) with femtosecond (fs) pulses to enable sensitive single-shot broadband measurements of OH and temperature with a time resolution of ~180 nanoseconds (ns) in LTPs [1]. Such combination leveraged several diagnostic benefits. With the appropriately designed cavity, an absorption gain of ~66 was achieved, enhancing the actual OH detection limit by ~55× to the 10^{11} cm^{-3} level (sub-ppm in this work) compared to single-pass absorption. Single-shot measurements were enabled while maintaining a time resolution of ~180 ns sufficiently short for detecting OH with a lifetime of ~100 µs. With the broadband fs laser, ~34000 cavity modes were matched with ~95 modes matched on each CCD pixel bandwidth, such that the fs-CEAS signal became immune to the laser-cavity coupling noise and highly robust across the entire spectral range. Also, the broadband fs laser allowed simultaneous sensing of many absorption features, which were exploited to enable simultaneous multi-parameter measurements with enhanced accuracies.

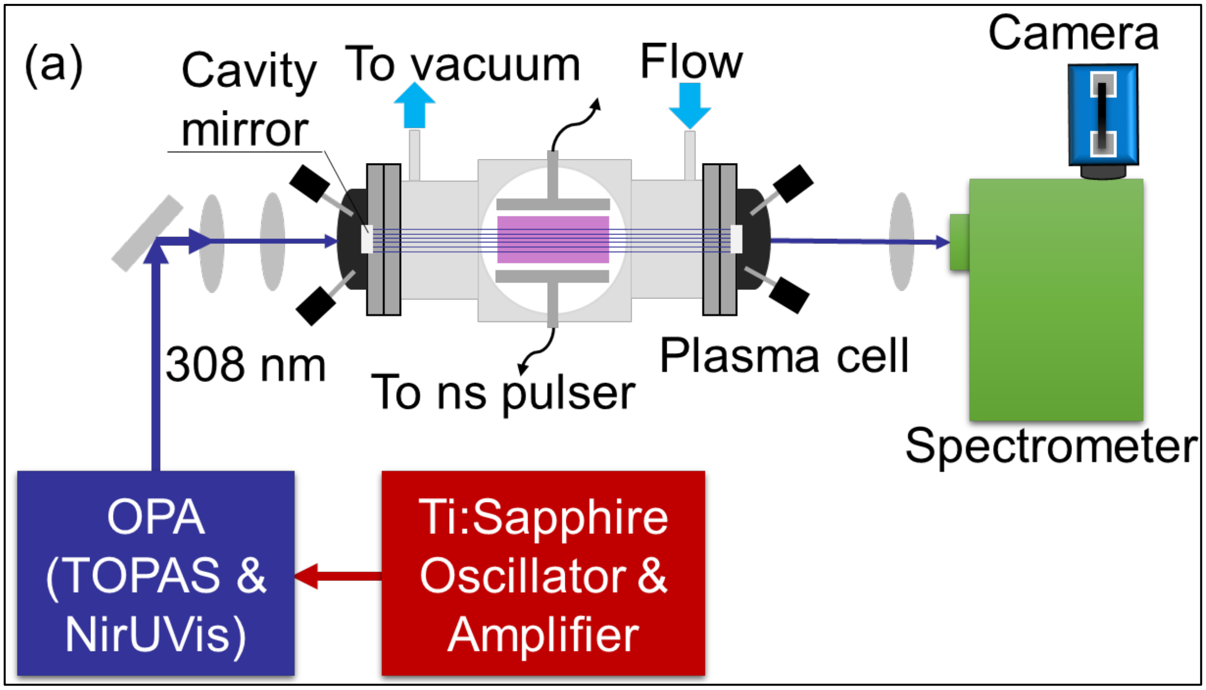

Figure 1 Fs-CEAS setup for *in-situ* single-shot OH and temperature measurements.

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[I-3] Laser Induced Fluorescence Measurements of Vibrationally Excited Oxygen Produced by Recombination of O Atoms

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Vibrationally excited oxygen and nitrogen have been long recognized to be of critical importance in nonequilibrium high-enthalpy flows, primarily encountered behind hypersonic shock waves [1]. To date, no direct experimental verification of vibrational state resolved dissociation rates predicted by different kinetic models has been undertaken, in large part due to the difficulty of measuring these rates directly. We present a framework where state specific recombination rates can be inferred from the time-resolved measurements of O_2 vibrational populations, such that the state specific dissociation rates can be obtained from the detailed balance.

In the present approach, recombination reactions of atomic oxygen are monitored in the afterglow of a diffuse ns pulse discharge burst in an O_2-Ar mixture. The time evolution of $O_2(v)$ populations in the recombining mixture are measured by ps Laser Induced Fluorescence (LIF) in the Schumann-Runge bands, with absolute calibration by NO LIF. By varying the output wavelength of the ps laser / Optical Parametric Oscillator (OPO) system used for the excitation in the 250-450 nm range, levels from v"=8 to 21 have been detected. Two-photon Absorption LIF (TALIF) at 226 nm has been used to measure the atomic oxygen concentration, with absolute calibration by Xe [2].

Within ~0.2 ms after the discharge burst, a rapid decay of $O_2(v)$ is observed, indicating vibration-vibration (V-V) and vibration-translation (V-T) relaxation of vibrational states populated by electron impact and by quenching of the excited electronic states of Ar. After the rapid initial decay, the vibrational populations level off and slowly decay, on the timescale of about 3 ms, suggesting the presence of a persistent source of vibrational excitation due to chemical reactions. In addition to O atom recombination to form vibrationally excited O_2 molecules, at low temperatures atomic oxygen may also recombine with O_2 and form ozone. The latter may well generate vibrationally excited O_2 in $O + O_3 \rightarrow O_2 + O_2$ reaction, which would therefore affect the present results. Ozone formation is controlled by heating the excited mixture up to 800 K, as well as increasing the pressure and reducing the O_2 mole fraction, when the production of ozone is reduced significantly. However, ozone production can not be reduced completely, therefore modeling calculations are necessary to understand the mechanisms by which $O_2(v)$ is produced. To successfully implement the model, the gas temperature in the afterglow must be known. This is acheived by Rayleigh scattering thermometry.

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[I-4] CO_2 Dissociation in Ns Pulse and “Hybrid” Ns-RF Discharge Plasmas

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Time-resolved CO_2 and N_2 vibrational populations and translational-rotational temperature are measured in a CO_2-N_2 plasma sustained by a ns pulse discharge burst in plane-to-plane geometry. Time-resolved, absolute number density of CO generated in the plasma is also inferred from the experimental data. CO_2 and CO vibrational populations are measured by mid-IR, tunable Quantum Cascade Laser Absorption Spectroscopy, and N_2 vibrational populations are measured by the ns broadband vibrational CARS. Transient excitation of N_2 and CO_2 asymmetric stretch vibrational energy modes is detected during the discharge burst. The time-resolved rate of CO generation does not correlate with N_2 or $CO_2(\nu_3)$ vibrational temperatures, indicating that CO_2 dissociation via the vibrational excitation is insignificant at the present conditions. The rate of CO generation decreases gradually during the discharge burst. Comparison of the experimental data with the kinetic modeling analysis indicates that CO_2 dissociation in collisions with electronically excited N_2 molecules is the dominant channel of CO generation at the present conditions, although the inferred CO yield in these processes is significantly lower than 1. The effect of vibrational energy transfer between N_2 and CO_2 on the plasma chemical processes is insignificant. The kinetic model underpredicts a rapid reduction of the N_2 and $CO_2(\nu_3)$ vibrational temperatures during the later half of the discharge burst and in the afterglow. V-T relaxation of CO_2 by O atoms has a significant effect on the relaxation rate.

CO_2, N_2, and CO vibrational temperatures, CO number density, and gas temperature are also measured in the CO_2-N_2 plasma generated by a ns pulse discharge combined with a sub-breakdown RF waveform. The results demonstrate significant additional vibrational excitation of all three molecular species by the RF field, including the CO_2 reactant. This results in a higher specific CO production rate, as well as a significant increase of the gas temperature. Additional measurements are planned to isolate the effect of vibrationally enhanced CO_2 dissociation from the thermal chemical reactions.

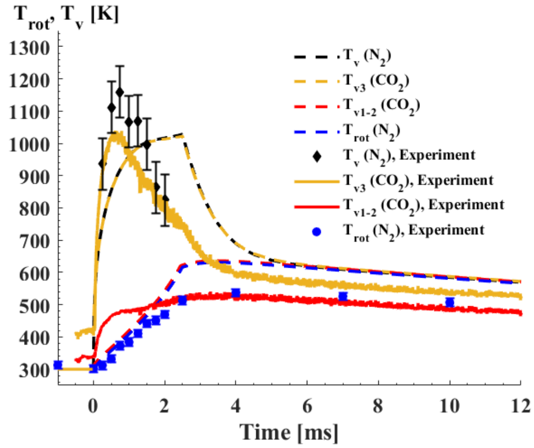

Figure 1 - Vibrational and rotational temperatures of N_2, CO_2, and CO during and after ns pulse discharge burst in 5% CO_2-N_2 mixture at P=100 Torr, compared with kinetic modeling predictions.

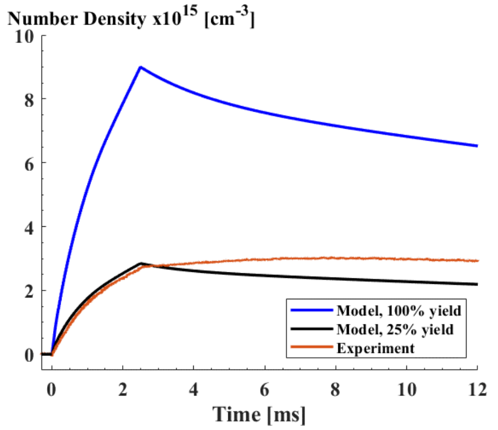

Figure 2 - Experimental and predicted CO number density during and after the discharge burst. The modeling predictions are plotted for 100% and 25% CO yield in N_2^* quenching reactions by CO_2.

[I-5] LTP-PIC, an Open-source, Three-dimensional Kinetic Code for Modelling Low-temperature Plasmas on Modern Supercomputing Architectures

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The Princeton Plasma Physics Laboratory has been developing a new kinetic particle-in-cell code designed for use by the low-temperature plasma community. LTP-PIC models complex geometry on a uniform Cartesian mesh in two and three dimensions, incorporating a geometric multigrid linear algebra solver for the Poisson equation. LTP-PIC can handle an arbitrary number of charged species, which can interact with a fixed neutral background via elastic and inelastic collisional processes, including ionization and charge exchange. Surface interactions including secondary electron emission and charge accumulation on dielectric surfaces can also be modelled.

These capabilities have and are being demonstrated through simulations of various plasma devices such as Hall thrusters, Penning discharges, and CCP-RF discharges [1]. The code is designed from the ground up for performance and scalability, demonstrated on several high-performance clusters, including heterogenous CPU+GPU systems. LTP-PIC is also portable and can be run on systems ranging from personal computers to supercomputers. With an upcoming open-source release on GitHub we welcome the low-temperature plasma community, and others in need of a robust electrostatic PIC code, to engage with us on using and improving LTP-PIC.

Acknowledgments

Code development was funded by the PPPL LDRD program and is supported by the Princeton Collaborative Research Facility (PCRF) supported by the US Department of Energy through contract DE-AC02-09CH11466

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[I-6] Enhancing the Perfluorooctanoic acid (PFOA) Degradation through Plasma Reactor Parametric Optimization

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Drinking water of more than 200 million Americans could be contaminated by poly- and perfluoroalkyl substances (PFAS). Known as the *Forever Chemicals*, PFAS are a large family of over 12,000 highly persistent and toxic chemicals that don't occur in nature. The widespread use of PFAS in military, aerospace, automotive, construction and electronic industries, combined with their complicated environmental release, mobility, fate and transport has resulted in multiple exposure routes for humans. The high stability of the carbon-fluorine bonds comprising PFAS renders them virtually indestructible and thus resistant to conventional water treatment methods [1]. Electrical discharge plasma however destroys PFAS at timescales and energy efficiencies that rival those of other technologies by up to several orders of magnitude [2].

This study investigates the influence of various experimental parameters on the removal of perfluorooctanoic acid (PFOA), a representative PFAS, in a gas-liquid pin-to-ring electrical discharge plasma reactor with gas bubbling. By applying a high voltage pulse to the pin electrode, the plasma was initiated in the gas phase and allowed to interact with the liquid (bubble) surface. The influence of experimental parameters such as PFOA initial concentration (10 - 300 ppm), applied voltage (24 - 37 kV), inter-electrode gap (0 - 15 mm), and gas type (Ar, He, N_2 and air) and flowrate (2 and 5 LPM, Figure 1) on the PFOA removal/defluorination and plasma properties was studied. Results indicate that the highest PFOA defluorination rates are obtained for the lowest compound concentration, highest applied voltage and lowest inter electrode gap. In addition, the highest defluorination rates were measured for argon and lowest for helium. These results show the strong influences of the electric field, discharge nature, and bulk liquid mass transport in PFOA degradation.

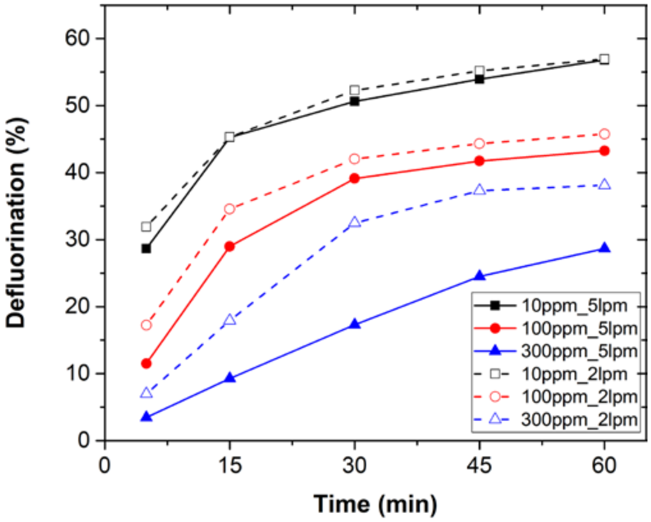

Figure 1 – PFOA deflurination profiles for various initial compund concentrations and argon gas flowrates. The applied voltage is -30 kV and discharge frequency 60 Hz.

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[I-7] Study of Plasma-catalyst Surface Interactions for Nitrogen Oxidation

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Cold atmospheric pressure plasma in conjunction with a catalyst is potentially useful for nitrogen oxidation. In this study quantitative and qualitative measurements of plasma-catalyst interactions are measured by infrared spectroscopic methods [1] for N_2-O_2 plasma with a Pt-Al_2O_3 catalyst. N_2/O_2/Ar gas is flown through an atmospheric pressure plasma jet towards heated catalyst powder in a sealed reaction chamber. Catalyst surface species are analyzed using Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS), while Fourier Transform Infrared Spectroscopy (FTIR) is used to quantify gas phase species exiting the catalyst bed. Catalyst temperature and plasma conditions are varied to investigate plasma-catalyst interactions. Results show multiple interactions on the catalyst surface. Interaction seems to be primarily between plasma produced NO_x species and the catalyst. DRIFTS reveals surface compounds formed under plasma exposure to be nitrites and nitrates likely bonded to alumina, as well as NO bonded to Pt. Figure 1 shows surface species build up on the catalyst surface under steady state conditions.

Heating the catalyst to 350 °C after plasma exposure causes NO_x to be released from the Pt catalyst surface as NO_2 and NO. DRIFTS Pt-NO absorbance-peak changes are consistent with NO desorption at high temperatures. The Pt catalyst also promotes oxidation of NO to NO_2 by interaction with O_2. Overall NO_x quantities may increase with a heated catalyst relative to the bare support material, but more investigation is needed. Thermal cycling of the catalyst creates hysteresis loops in downstream gas data that can be used to separate the plasma and surface contributions to downstream gas species production. We thank B. Bayer, Dr. A. Bhan and Dr. P. J. Bruggeman for helpful discussions. This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Fusion Energy Sciences under award number DE-SC0020232.

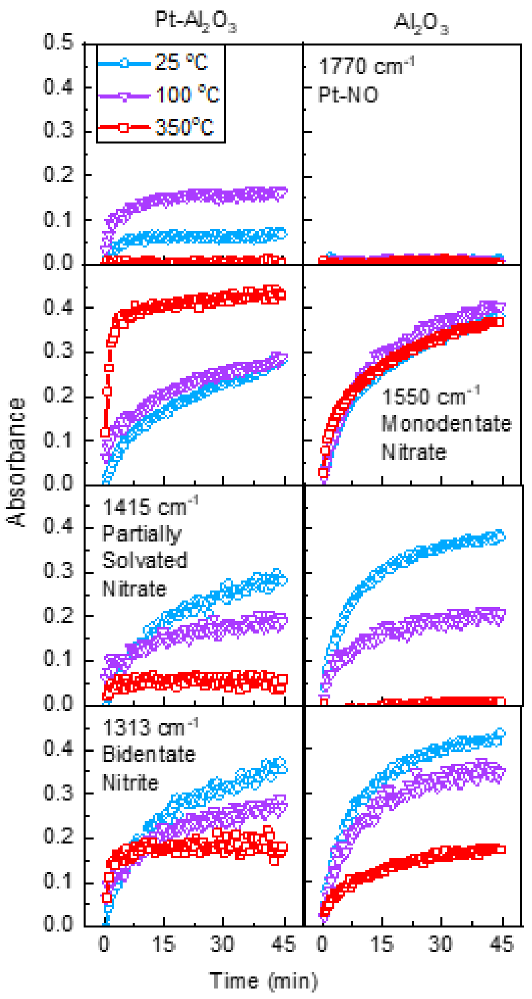

Figure 1 – DRIFTS Peak Absorbances over Time. Platinum appears to hinder nitrate formation at low temperatures and accelerate it at high temperatures.

Reference

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[I-8] Cold Plasma Treatment of Biomass Can Enhance Nitrate Content and Reduce Ammonia Evaporation

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Nitrogen-based synthetic fertilizer production and distribution generates a significant amount of greenhouse gasses. It has recently been postulated that non-thermal air plasmas can acidify biowaste by generating nitric acid and, thus, can hold promise for producing "green fertilizer" using renewable electricity [1]. To this end, our research aims to verify two hypotheses: 1. plasma treatment of biowaste will enhance plant-available nitrogen content by increasing the nitric acid content (due to the nitrate anion); and 2. plasma treatment will prevent ammonia evaporation from biowaste by ionizing ammonia to ammonium.

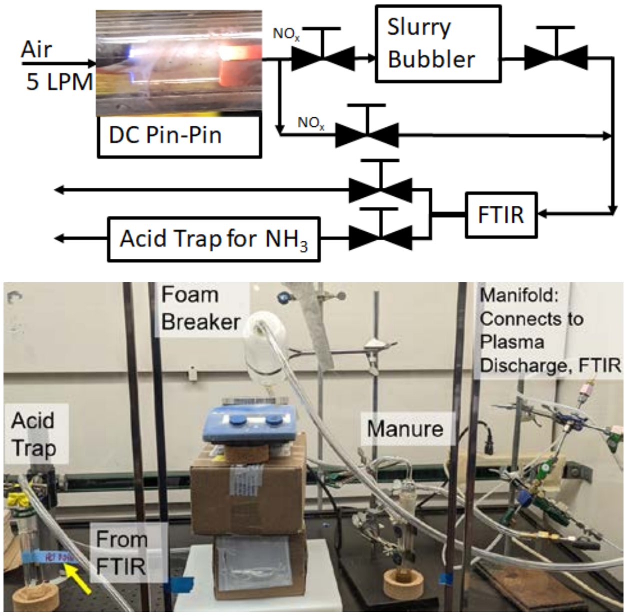

Figure 1 – Top: Experimental setup. The manifold enables routing of gas between the plasma discharge, FTIR, and fumehood for measurement. Bottom: Physical realization of experimental setup.

The above hypotheses are being tested by flowing plasma effluent generated by a DC pin-pin discharge with 5 LPM air as feed operating at 6 kV and 40 mA with an 11 mm pin distance to generate 5000 ppm NO$_x$, of which 0.5 LPM is diverted into the bioslurry. The first hypothesis is tested by examining the change in total nitrogen in the slurry (Total Kjeldahl Nitrogen) vs NO$_x$-nitrogen absorbed (as measured by FTIR). NO$_x$ absorption will be controlled by varying the time for which NO$_x$ vapor is bubbled through the manure slurry. We will also compare the change in nitrate/nitrite (generated from plasma) with the nitrogen absorbed via FTIR. This analysis can reveal the presence of process-limiting side reactions.

The second hypothesis is examined by stripping ammonia out of the slurry by flowing air through the treated/untreated biowaste slurry and capturing ammonia in the acid trap. The captured ammonia concentration is then measured via ion-selective electrodes. We will present how the ammonia captured in the acid trap varies as slurry exposure to plasma-generated NO$_x$-nitrogen varies. We will also present how the slurry pH varies in the same interval. If we observe a high conversion of absorbed N to nitrate/nitrite and a reduction in ammonia loss, then the next steps are to compare plasma-slurry contacting modes. Significant losses will drive us to consider gas-phase N-fixation to generate plasma activated water-based fertilizer to blend with manure.

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[I-9] Propagation of Surface Ionization Waves Over Dry and Wet Microchannels Exposed to an Atmospheric Pressure Plasma Jet

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Electric field over dry and wet microchannels machined in a macor ceramic cylindrical tube, placed under an Atmospheric Pressure Plasma Jet (see Fig. 1(a)), with a 10% N_2-Ar main flow and N_2 co-flow and powered by high-voltage ns pulses, has been measured by ps Electric Field Induced Second Harmonic (EFISH) generation, developed in our previous work [1].

A cylindrical tube is used instead of a plane microchannel plate to enable the laser beam positioning close to the surface. The measurements are made approximately 70 µm above the grooves and the ridges of the rectangular cross section microchannels, 250 µm × 250 µm, underneath the jet, as shown schematically in Fig. 1. The measurements are repeated with the channels filled with deionized water, due to the capillary flow. Plasma emission imaging shows that a surface ionization wave propagates along on the surface of the ceramic tube, away from the location of the plasma jet impingement. Simulations of the SIW are performed using nonPDPSIM, a two-dimensional plasma hydrodynamics model [2]. The simulation includes the solution of Navier-Stokes equations for the neutral gas flow, coupled with the transport equations for charged particles and Poisson's equation for the electric potential. Photoionization is represented using Green's function approach.

The experimental results and the model predictions show the periodic enhancement of the electric field inside and above the dry channels, as well as above the channels willed with liquid water. This effect is observed both for the horizontal and vertical electric field components. The distribution of the plasma and electric field in the channels is sensitive to the axial distance between the plasma jet and the measurement location.

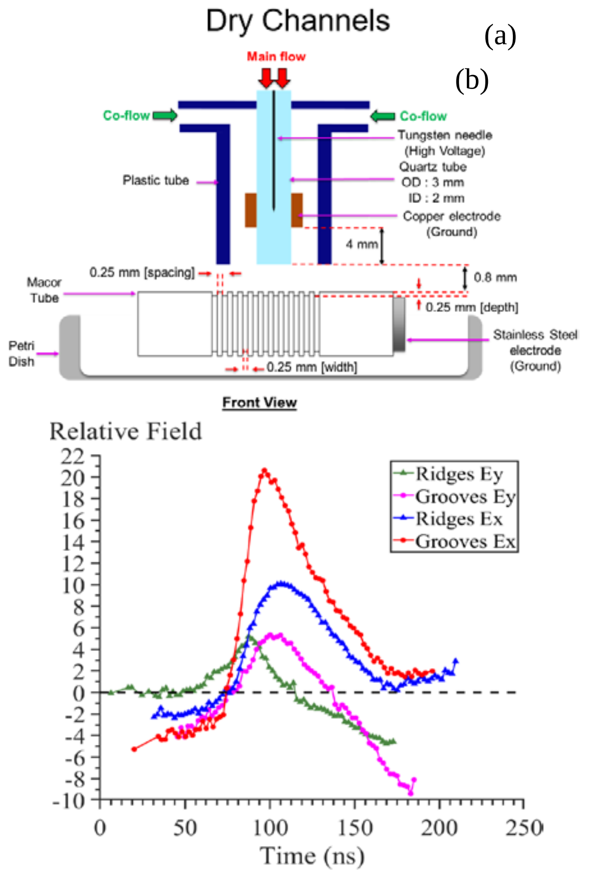

Figure 1. (a) Schematic of the plasma jet impinging over dry microchannel array. (b) Vertical and horizontal electric field components inside a groove and over a ridge, 2.5 mm and 2.75 mm away from the center of the jet, respectively.

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[I-10] Falling Liquid Film Plasma Reactor – A New Approach to Study Temporally Resolved Plasma-Liquid Interactions

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Plasma-liquid interactions enable many applications including nanoparticle synthesis, wound healing, bacterial inactivation and wastewater treatment [1]. However, due to the limited penetration depth of short-lived plasma-generated species in the liquid phase, a experimental study of these near interfacial species is exceedingly difficult [1]. Controlled plasma-liquid interaction studies for which transport in the liquid phase is dominantly due to diffusion have been reported for formate decomposition in a plasma-droplet reactor [2]. This study showed that formate conversion is diffusion limited for nearly all investigated plasma conditions. The impact of convective transport has been studied mostly through numerical modeling with studies performed by Kaneko et al [3] and Ghosh et al [4] using a liquid jet. Studies regarding simultaneous control of both convective and diffusive transport in the liquid phase are yet to be reported in spite their critical impact on plasma-liquid interactions.

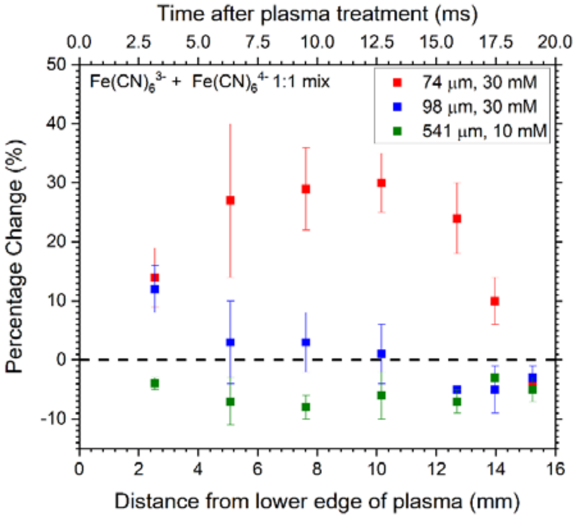

Figure 1 – Time-resolved conversion of 1:1 ferricyanide/ferrocyanide mixture for different film thicknesses. Both the position of the measurement and the corresponding time axis assuming liquid plug flow are shown. A positive and negative percentage change indicates oxidation of ferrocyanide and reduction of

A falling liquid film plasma reactor has been developed, operating at flow velocities between 0.5 – 2 m/s and film thicknesses ranging between 50 to 800 μm, allowing us to control convection and diffusion timescales perpendicular to the flow direction. The reactor enabled the in situ measurement of the temporally resolved conversion of various compounds using absorption spectroscopy. Figure 1 shows the conversion of the redox couple ferricyanide and ferrocyanide on a millisecond time scale. The results allow us to identify three dominant reactions at the plasma-liquid interface: (a) electron driven reduction for film thicknesses > 100 μm, (b) OH driven fast oxidation on time scales of 2-10 ms and (c) a previously unreported reduction process on a time scale of 20 ms which we ascribed to reductions reactions of HO_2^- for a film thickness < 100 μm. These results show the capability of this system to investigate for the first time near interfacial reactions in the liquid in thin liquid films. This capability uniquely allows linking liquid phase processing to gas phase plasma processes and providing new insights in plasma-liquid interactions.

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[I-11] OH and Water Vapor Concentration Measurements Surrounding Water Droplets in an Atmospheric Pressure Plasma

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The interaction of liquids with low temperature atmospheric pressure plasmas has been investigated intensively due to its large potential in many applications including pollution control, decontamination and fertilizer production. These applications are enabled by plasma-produced reactive oxygen and nitrogen species. An effective approach to enhance the production of reactive species in liquids is using micrometer size droplets dispersed in plasma for which the large surface-to-volume ratio significantly enhances the transport of gas phase reactive species to the liquid. However, the strong coupling between the plasma and the droplet can lead to huge changes in the local plasma properties surrounding the droplet which can significantly impact the transport of reactive species into the droplet. These local variations in the near droplet region has not been studied in detail partly because the associated diagnostic challenges and is the focus of this work.

We report on the measurement of the spatial distribution of water vapor and OH radicals by planar laser induced fluorescence (PLIF) along the trajectory of a droplet passing through an atmospheric pressure RF plasma in He + 17% Ar mixture. To enable this measurement, we generate a train of droplets with a diameter of ~40 μm with a droplet residence time in the plasma 10 times smaller than the dispensing period [1].

Figure 1 shows the water vapor concentration and OH density near the droplet. The evaporation of the droplet, in spite the moderate size reduction of the droplet, produces a local enhancement of water vapor concentration up to 1500 ppm within 1 mm near the position of the droplet. Interestingly, the OH density variation remains within a factor of three while previously a square root dependence of the OH density on the water concentration was reported [2]. This result suggests a significant impact of the droplet on the local plasma kinetics.

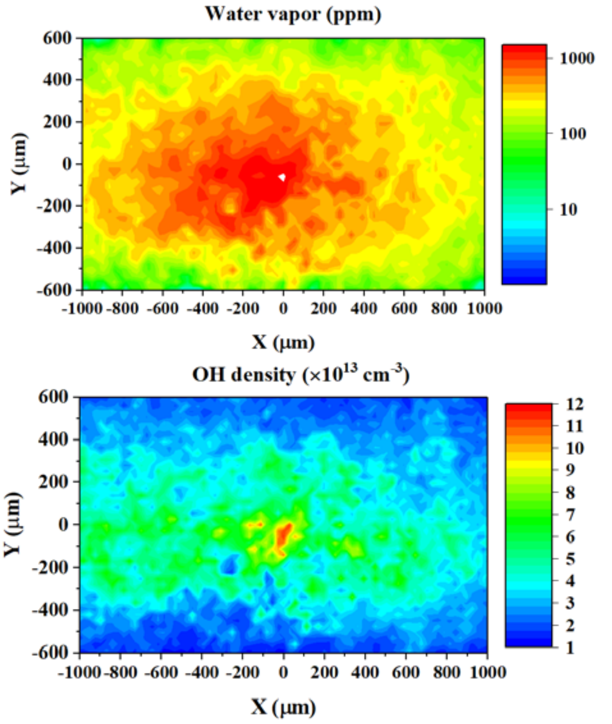

Figure 1 – Distributions of water vapor concentration (upper figure) and OH densities (lower figure) densities in the surrounding of a 40 μm droplet in RF plasma in He+17% Ar (5.8 W). The droplet is positioned at (0,0) and the shown cross section is perpendicular to the electrodes with the electrodes positioned at |Y| ~ 2000 μm.

We implemented PLIF in a plasma-droplet reactor with the capability to study near interfacial processes in plasma-droplet interactions allowing the investigation of local kinetics and transport processes which we will discuss in detail during the presentation.

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[I-12] Photoemission Induced Plasma Breakdown in Argon

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Laser-induced photoemission of electrons offers opportunities to trigger and control plasmas and discharges. However, the underlying mechanisms are not sufficiently characterized to be fully utilized. Photoemission is highly nonlinear, achieved through multiphoton absorption, above threshold ionization, photo-assisted tunneling, etc., where the dominant process depends on the work function of the material, photon energy and associated fields, surface heating, background fields, etc. To characterize these effects, Townsend breakdown experiments were performed and interpreted using a circuit model and quantum model of photoemission. At low currents, it is found that laser induced photoemission can be sufficiently de-coupled from space charge effects to be observable. Photoemission induced plasma breakdown is investigated with high-speed imaging for different reduced electric fields and laser intensities and photon energies (<6.3 eV). Experiments were performed using a tunable picosecond laser that allowed the use of a two-temperature model for electrode heating.

Acknowledgements
This work was supported by Sandia National Laboratories' Plasma Research Facility, funded by the U.S. Department of Energy Office of Fusion Energy Sciences. Sandia is managed and operated by NTESS under DOE NNSA contract DE-NA0003525. The MSU team was supported by Office of Naval Research (ONR) YIP Grant No. N00014-20-1-2681 and U.S. Department of Energy Office of Science, Grant No. DE-SC0022078.

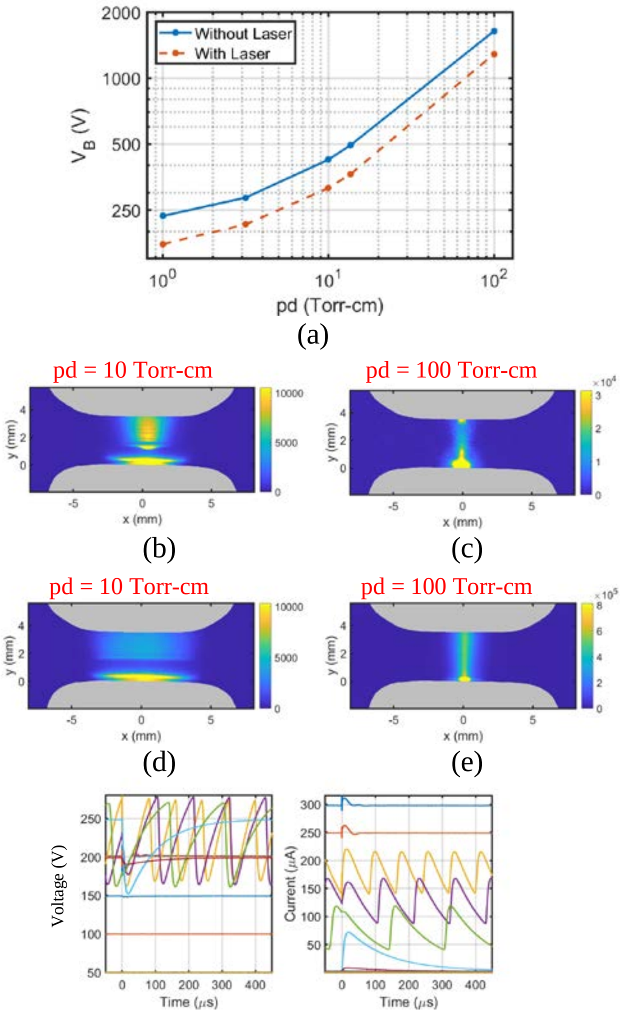

Figure 1 – (a) Effect of laser-induced photoemission on breakdown voltage (V_B) in argon. (b) and (c) images of breakdown aquired through a 750 nm bandpass filter without the laser and (d) and (e) with the laser. Colorbars have arbitrary units. (f) Gap voltage and current with decreasing applied voltage at pd = 1 Torr-cm. The ps laser pulse arrives at t = 0 μs. At lower voltages, breakdown can be achieved with laser-induced photoemission.

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Abstracts - Poster Session II

[II-1] Modelling the Effect of Surface Charging on Surface Reactions During Plasma Synthesis of NH_3 Using DFT

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Ammonia is one of the most important industrial gases. The conventional ammonia synthesis method - the Haber–Bosch process – converts atmospheric N_2 into ammonia using H_2 and metal catalysts. This process employs high pressures (100 - 200 atm) and temperatures (700−800 K). These thermodynamic conditions are required to maintain the equilibrium in the favor of producing ammonia as well as to maintain the efficiency of the catalysts [1]. Plasma assisted synthesis of ammonia can overcome these thermodynamic limitations. Non-equilibrium plasmas have shown to synthesize ammonia using N_2 and H_2 at relatively lower temperatures and pressures (~473 K and 1 atm) in the presence of various catalysts [2]. The rate limiting step for thermal catalysis of ammonia is the dissociation of N_2 on the catalytic surface. N_2 is a strongly bonded molecule and its dissociative barrier on commonly used catalysts like Fe and Ru is quite high (60–115 kJ mol-1) [2]. This high barrier leads to the requirement of high temperature for thermal-catalytic production of ammonia in an industrial setting. It has been hypothesized that non-equilibrium plasma environment arguably circumvents high dissociation barrier for N_2 by vibrationally exciting the molecule [3]. However, the short vibrational excitation timescale means other plasma induced effects could play an important role is plasm-assisted catalysis of NH_3 [4].

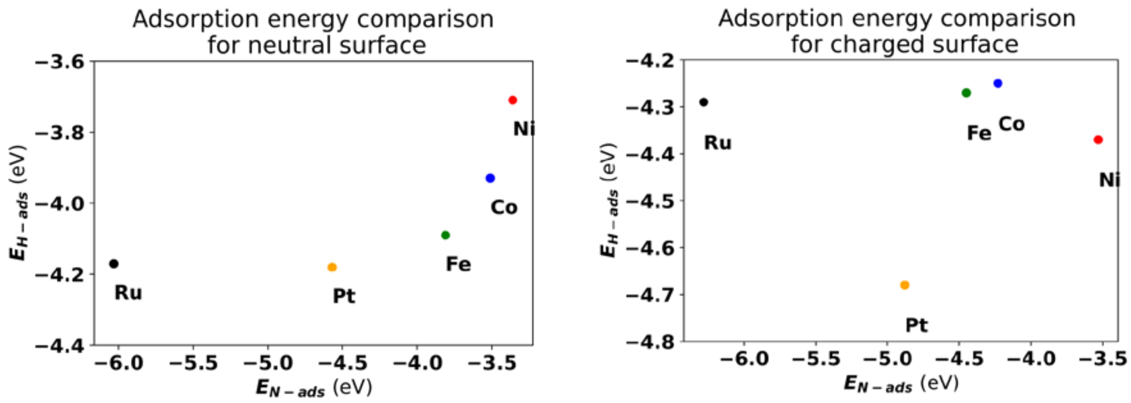

Figure 1 – Adsoprtion energy of H-atom with respect to the adsoprtion energy of N-atom on a single metal atom catalyst (left) on a neutral surface and (right) on a charged surface.

One of the less explored aspects of plasma induced effects is the negative charging of the catalytic surface [2]. We used density functional theory (DFT) calculations [5] to show that in case of single metal atom catalysts, the adsorption energy trends change upon considering negatively charged surface (see Fig. 1). However, metal clusters or nanoparticles on a dielectric support such as γ-Al2O3 are a better representation of the plasma catalysis systems. In this poster, we present the changes to the reaction energetics of ammonia synthesis surface reactions due to the surface charge on metal clusters. Reaction energetics for important surface reactions such as N_2, H_2 dissociative adsorption as well as the intermediate reactions are explored using DFT calculations.

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[II-2] Effects of Inter-pulse Coupling on Nanosecond Pulsed High Frequency Discharge Ignition in a Flowing Mixture

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This work numerically investigates the effects of non-equilibrium nanosecond plasma discharge pulse repetition frequency, pulse number, and flow velocity on the critical ignition volume, minimum ignition energy, and chemistry in a plasma-assisted H_2/air flow at 300 K and 1 atm using a multi-scale adaptive reduced chemistry solver for plasma assisted combustion (MARCS-PAC) [1, 2]. The interactions between discharges/ignition kernels spanning decoupled, partially-coupled and fully-coupled regimes in a pulse train are studied. For a single pulse discharge, increased flow velocity increases the minimum ignition energy required due to the increase of convective heat loss and flame stretch. The results show that the minimum ignition kernel growth at the critical ignition kernel volume increases with the flow velocity. The minimum critical ignition volume decreases with the increase of plasma discharge energy. For sequential two-pulse discharges, ignition fails at both decoupled and partially-coupled regimes even when the total discharge energy is above the minimum ignition energy, but succeeds only in the fully-coupled regime at a shorter inter-pulse time. Overlap of the OH radical pool between the sequential two-pulse discharges and the increase of the chemistry effect due to the increase of reduced electric field in the fully-coupled regime contribute to the ignition enhancement. In addition, for two-pulse discharges in the fully-coupled discharge regime, the mixture can be ignited at a total energy below the minimum ignition energy of a single pulse with the same flow conditions. Moreover, for a given total discharge energy with multiple pulsed discharges, the enhancement of the ignition kernel volume has a non-monotonic dependence on discharge frequency and pulse number. The effective ignition enhancement can be achieved with an optimal pulse repetition frequency and pulse number. This work provides a new understanding of the mechanism for repetitive plasma ignition and insights for the optimization of plasma ignition in a reactive flow.

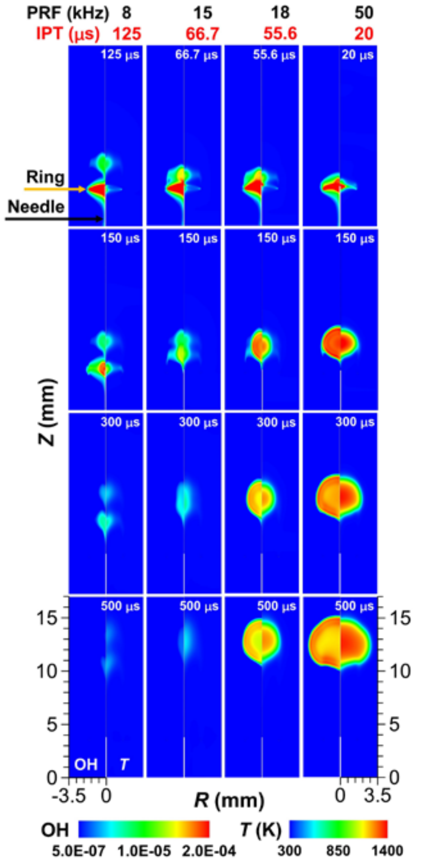

Figure 1 Time evolutions of mole fractions of OH (left) and temperature (right) in 20 m/s flowing H_2/air mixtures at various pulse repetition frequency (PRF)/inter-pulse time (IPT) conditions with 2 pulses,

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[II-3] Electric Field Distribution in a “Hybrid” RF Discharge with Ionization Generated by Ns Discharge Pulses

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Electric field in a capacitively coupled, non-self-sustained RF discharge nitrogen plasma with external ionization generated by high-voltage ns pulses at P=150-250 Torr has been measured by ps Electric Field Induced Second Harmonic (EFISH) generation, developed in our previous work [1].

The measurements are made both in the bulk of the plasma and in the sheaths, as shown schematically in Figure 1, using absolute calibration by the Laplacian field between two plane electrodes. The results are compared with kinetic modeling calculations, showing good agreement. RF electric field in the sheaths is significantly higher compared to that in plasma, due to the displacement of the electrons by the drift oscillations and the resultant plasma self-shielding. However, kinetic modeling predictions indicate that the electron impact ionization in the sheaths is largely ineffective, due to the low electron density there.

At the present conditions, the reduction of the RF electric field in the plasma by the self-shielding in the sheath is moderate, such that the energy is coupled to the plasma by the below breakdown field. Peak RF field in the plasma is in the range of E/N = 15-30 Td, indicating the efficient vibrational excitation of N_2 by electron impact. The modeling predictions suggest that the targeted vibrational excitation of molecular species in a non-self-sustained RF discharge with external ionization is scalable to high pressures and discharge power densities. This approach can be extended to the efficient vibrational excitation of other molecular species where vibrational relaxation is relatively slow, such as CO, CO_2 and H_2.

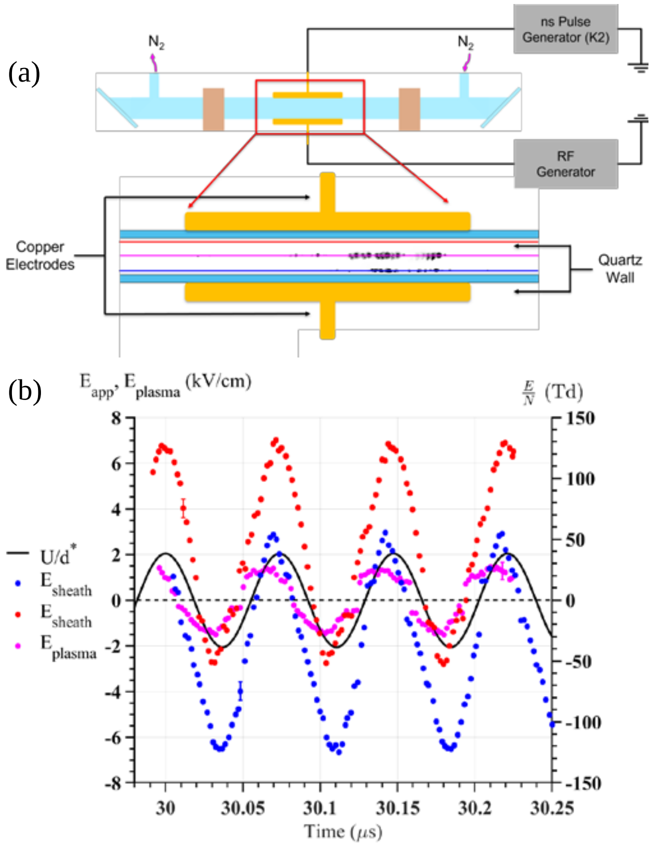

Figure 1. (a) Schematic of the discharge cell. The lines illustrate laser beam locations, determined by imaging of laser induced breakdown in ambient air, used for electric field measurements in the sheaths and in the plasma. (b) Electric field in the sheaths and in the plasma at t = 30 μs after beginning of the RF burst in N_2 at P=150 torr. Reduced electric field is indicated on the right axis.

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[II-4] Plasma-assisted Deflagration to Detonation Transition in a Microchannel

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This study investigated the affect of nanosecond dielectric barrier discharge (ns-DBD) plasma on the kinetics of a fuel-lean, $\Phi = 0.7$, dimethyl ether (DME), oxygen (O_2), and argon (Ar) premixture during deflagration to detonation transition (DDT) experiments in a microchannel. Fast-Frame Imaging, Hybrid fs/ps Coherent Anti-Stokes Raman Scattering (CARS), Electric Field Induced Second Harmonic Generation (EFISH), and Formaldehyde (CH_2O) Laser Induced Fluorescence (LIF) experiments are conducted to examine the influence of the plasma discharge on the premixture and on the resultant deflagration to detonation transition (DDT) onset time and distance. Nanosecond dielectric barrier discharges (ns-DBDs) are applied across the length of the microchannel before ignition, producing active species and radicals as well as fast and slow heating of a mixture to promote ignition due to energetic electrons, ions, and electronic and vibrational excitations. A high-speed camera is used to trace the time histories of flame front position and velocity and to identify the dynamics and onset of DDT. The results show that plasma discharge can nonlinearly affect the onset time and distance of DDT. It is shown that a small number of plasma discharge pulses prior to ignition result in reduced DDT onset time and distance by 60% and 40%, respectively, when compared to the results without pre-excitation by ns discharges. When instead, an increase of number of the plasma discharge pulses is applied the resultant DDT onset time and distance are extended by 224% and 94%, respectively. Analysis of the ignition delay time in alongside the fast-frame observed time history of the deflagration wave speed of DME and the analysis of ignition timescale suggest low temperature ignition may play a role for DME near the isobaric choking condition of the burned gas and the DDT. Plasma-induced low temperature oxidation of the reactive mixture is assessed via the CO_2 to O_2 ratio as measured through fs/ps CARS during the gas excitation in discharges. CARS measurements also confirm negligible vibrational and rotational heating of the gas by discharge. EFISH experiments in the channel quantify the field strength of the ns-DBD plasma in the cell. Laser Induced Fluorescence experiments show an increase in CH_2O concentration ahead of the flamefront suggesting that the applied plasma is enhancing the low temperature chemistry of the fuel. The present experiments demonstrate the ability of nonequilibrium plasma to alter the chemistry of DME/O_2/Ar premixtures for the control DDT in applications for advanced propulsion engines.

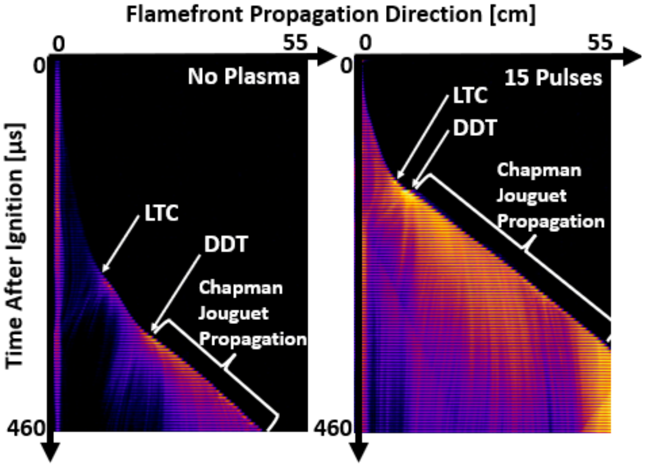

Figure 1 – Stacked fast frame images of flame front progression without (left) and with (right) plasma pre-pulses. Color is added to highlight chemiluminescence intensity.

[II-5] Interconnections among Pulse and Plasma Properties and Chemical Reactions in Gas-liquid Plasma

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Plasma electron density and temperature were characterized in a continuous flowing gas-liquid film reactor with argon carrier gas by time-resolved optical emission spectroscopy. The plasma parameters were studied as a function of time for varying pulse widths and frequencies. Pulse frequency was varied between 1 and 10 kHz at 16 kV (input voltage) and 40 ns (pulse width) using an Eagle Harbor Technologies, Inc. (EHT) power supply and 5–100 kHz using an Airity Technologies, LLC (AT) power supply. The pulse width was varied between 40 and 200 ns at 16 kV, 2 kHz with the EHT power supply. Optimal frequencies of 5 and 20 kHz were observed for peak electron density with EHT and AT power supplies, respectively. The peak electron density increased with increasing pulse width between 40 and 200 ns using the EHT power supply. Hydrogen peroxide exiting the reactor in the liquid phase increased with discharge power irrespective of the power supply or pulse parameters. Mineralization of 12.5, 50, and 200 ppm perfluorooctanoic acid (PFOA) dissolved in DI water to fluoride (F^-) correlated to the peak electron density.

Acknowledgement

This work was performed at the Princeton Collaborative Research Facility (PCRF) supported by the US Department of Energy through contract DE-AC02-09CH11466.

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[II-6] Degradation of Poly- and Perfluoroalkyl Substances (PFAS) in a Plasma Spinning Disk Reactor

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A plasma spinning disc reactor has been used for the treatment of short- and long-chain poly- and perfluoroalkyl substances (PFAS). Due to the strength of the carbon fluorine bonds comprising these compounds, PFAS are resistant to oxidation and can only be chemically reduced. In this work, we compare the efficacy of the plasma spinning disc reactor (Figure 1) in degrading these compounds to that of the enhanced contact plasma bubbling reactor, which has one of the best PFAS treatment efficiencies. Results show that higher long- and short-chain PFAS defluorination rates can be achieved in the plasma spinning disc reactor compared to the bubbling reactor, indicating that thin films may be as effective in ensuring the compound's proximity to the plasma-liquid interface as bubbling. To develop a mechanistic understanding of the PFAS degradation in the plasma spinning disc reactor, the roles of liquid thickness, convective transport, and energy deposition were studied through parametric reactor optimization. Results indicate that at low PFAS concentrations (10 ppm) discharge energy played a minimal role in the degradation. In contrast, liquid surface renewal rates played a dominant role in the PFAS degradation and defluorination indicating the importance of bulk liquid processes in interfacial kinetics.

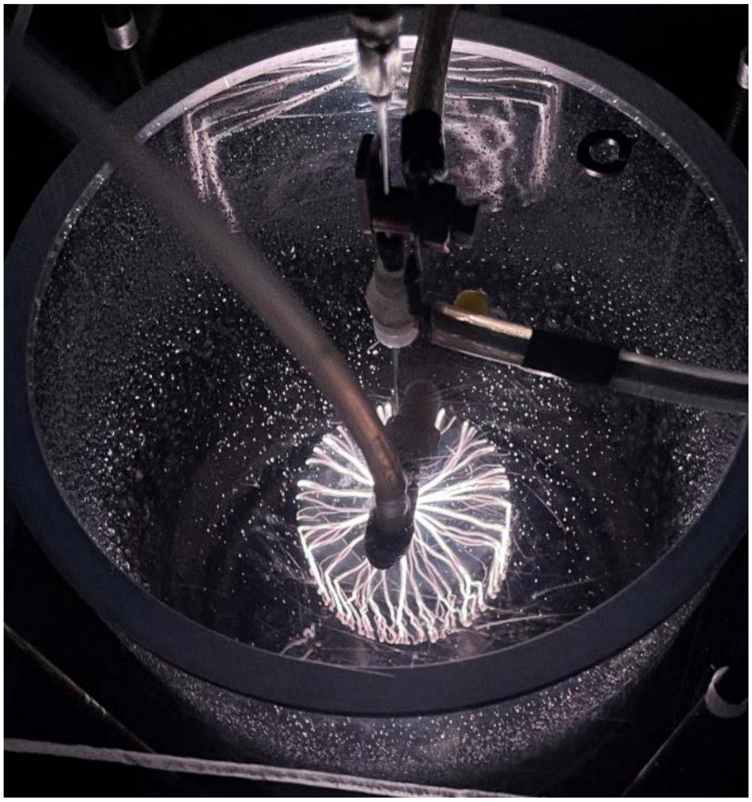

Figure 1 - Image showing the spread of plasma over a rotating disc in the plasma spinning disc reactor.

[II-7] Low-Temperature Plasma Interaction with Rectangular Microchannels

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Most topical areas of existing materials are usually a combination of non-planar and/or non-uniform interfaces. Low-temperature plasmas have been widely used to treat different materials: from biological tissue to plastics. A detailed understanding of fundamental physics and chemistry of interaction of plasmas generated with various sources and applied to different materials is needed. Rectangular microchannels that are discussed in this work can represent natural irregularities on the surfaces. One example of such surfaces is wrinkled skin. It is widely known that low-temperature plasmas have potential benefits if applied to biological tissue.

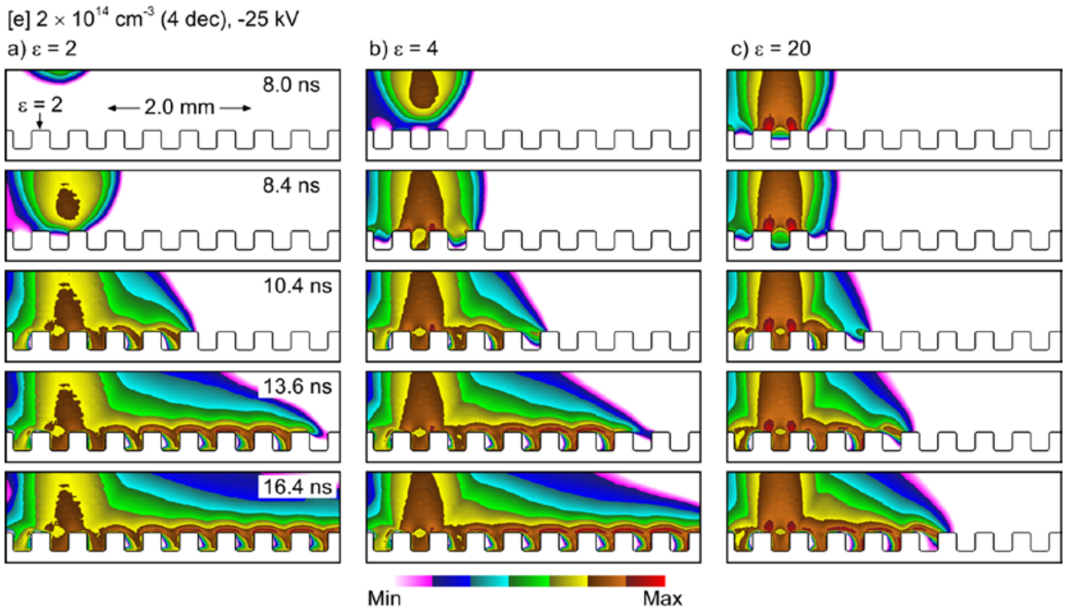

Figure 1 – Electron density time evolution during plasma interaction with dielectric substrate of different dielectric permittivities.

This work contains a discussion of the computational investigation of the interaction of atmospheric pressure argon plasma jet with rectangular microchannels using a platform nonPDPSIM [1]. The results are supported by experimental data obtained using a method to measure electric field EFISH [2]. Various parameters are investigated, including voltage magnitude, feeding gas, conductivity and permittivity of the substrate (Fig. 1).

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[II-8] Reaction Mechanisms for Atmospheric Pressure Plasma Treatment of Organic Molecules in Solution*

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Small, portable atmospheric pressure plasma jets (APPJs) (e.g. KINpen, COST-jet) are gaining traction in the biomedical community as sources of reactive oxygen and nitrogen species (RONS) for bactericidal applications. Plasma-produced RONS solvate into solution and are thought to interact with amino acids located on cell walls to irreversibly change the functionality of cells in solution. To further understand this phenomena, fundamental investigations utilizing APPJs have focused on plasma-liquid interactions of RONS with simple biologically-relevant amino acids (e.g., cysteine) in solution. Experimental studies have shown RONS are depleted in reactions with simple amino acids in solution, producing oxidation products that are similar to those formed by treatment of organic molecules with gas-phase plasmas [1]. Reaction mechanisms for plasma-liquid interactions are generally available for solutions that do not contain organic molecules and materials. Development of these mechanisms could provide insight for biological applications.

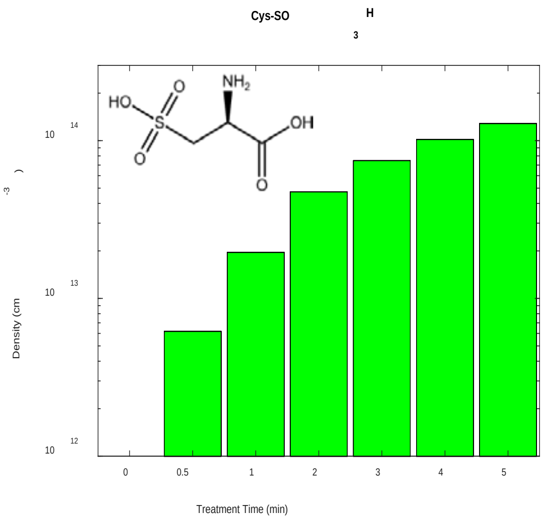

Figure 1 – Density of sulfonic acid (cysteine oxidation product) after 5 minutes treatment by $He/O_2/N_2$, 5 W, COST-jet produced APPJ.

In this work we develop a reaction mechanism for the computational investigation of APPJ treatment of cysteine in water. A global plasma chemistry model adapted for plasma-liquid interactions was used to investigate the effects of plasma activated oxidizing (e.g., O, OH) species on the final oxidation states of cysteine in solution. The interactions of these RONS with the cysteine molecule will be discussed and compared to experiments.

* Work supported by the US NSF, DOE Fusion Energy Sciences and the Army Research Office MURI Program.

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[II-9] Density Functional Theory Coupled with Microkinetic Model to Study Plasma-Catalyst Interactions in Ammonia Synthesis

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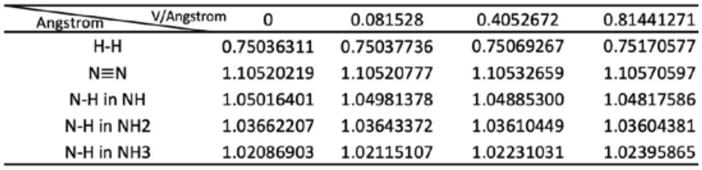
Plasma catalysis has shown promise for the electrification of chemical synthesis, including NO_x synthesis, methane reforming and NH3 synthesis, via complex and not-well-understood synergies between plasma and catalysts. However, there have been limited studies on how the surface electric field will impact surface reaction rates in terms of changing their corresponding activation energy or entropic barriers [1]. Current microkinetic models for ammonia synthesis assume constant sticking probabilities for surface reactions, while density functional theory (DFT) studies seldom consider the impact of electric field on the surface reactions [2,3].

The main objective of our work is to study the impact of electric field on plasma-catalytic ammonia synthesis by leveraging microkinetic modeling and DFT. To this end, we have coupled DFT calculations with a 0D microkinetic model in ZDPlasKin [4] to describe the production rate and energy efficiency of plasma-assisted NH_3 synthesis on Fe catalyst. The kinetic rate coefficient expressions in the microkinetic model are modified to use activation energy and entropic barriers from DFT. We consider the two main types of surface reactions, i.e., Langmuir-Hinshelwood (LH) and Eley-Rideal (ER) mechanisms. We adopt an activation-energy-based rate coefficient expression for LH reactions, while the sticking probabilities for ER reactions are informed by entropic barriers. Furthermore, no transition state is considered for the ER reactions, whereas an entropic barrier is used for gas phase molecules that react with the surface adsorbed molecules [2,3]. According to DFT calculations, large and nonlinear variations of activation energies for LH reactions are observed under different electric field strengths. Our DFT calculations also reveal a fairly large sensitivity of molecule bond lengths to electric field (see Table 1), implying the impact of entropic contributions due to vibration and rotation that in turn influence the ER reaction coefficients. This is because the bond length corresponds to both the vibrational frequency and moment of inertia, which determine vibrational and rotational entropies. These results indicate electric field can impact surface reactions in a complex and nonlinear way.



V/Angstrom Angstrom	0	0.081528	0.4052672	0.81441271
H-H	0.75036311	0.75037736	0.75069267	0.75170577
N≡N	1.10520219	1.10520777	1.10532659	1.10570597
N-H in NH	1.05016401	1.04981378	1.04885300	1.04817586
N-H in NH2	1.03662207	1.03643372	1.03610449	1.03604381
N-H in NH3	1.02086903	1.02115107	1.02231031	1.02395865

Table 1 – Impact of electric field on bond lengths of molecules. The changes in bond lengths correspond to the changes of vibrational and rotational entropy contributions.

We are currently investigating the entropy changes for ER reactions using Quantum Espresso [5]. This way our microkinetic model can account for the impact of electric field on surface reactions, allowing us to study the plasma-catalytic ammonia synthesis process more comprehensively. In future, using the DFT coupled microkinetic model, we will leverage state-of-the-art multi-fidelity and multi-objective data-driven optimization methods to systematically explore the plasma and catalyst parameters that maximize NH3 production while minimizing energy cost.

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[II-10] Kinetics of HO_2 Radical Formation and Decay in Ns Pulse O_2-He Plasmas over a Liquid Water Surface

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Time-resolved, absolute number density of HO_2 radical in O_2-He mixtures excited by a repetitive ns pulse discharge over a pool of distilled water is measured in situ by Cavity Ring Down Spectroscopy (CRDS). The discharge cell with external electrodes to generate the plasma and a water reservoir are integrated into the CRDS cavity. The schematic of the CRDS plasma flow reactor is shown in Fig. 1(a). The diagnostic is described in detail in our previous work. The experimental results are obtained at near room temperature, both during the discharge pulse burst and in the afterglow. The HO_2 number density is inferred from the CRDS data using a spectral model exhibiting good agreement with previous measurements of absolute HO_2 absorption cross sections. HO_2 is generated during the discharge burst and decays in the afterglow between the bursts, on a ms time scale. The HO_2 number density is also measured vs. the O_2 fraction in the mixture. Comparison with the kinetic modeling predictions demonstrates good agreement with the data (see Fig. 1(b)) and identifies the dominant HO_2 generation and decay processes. HO_2 in the plasma is formed predominantly by the recombination of H atoms, generated by the electron impact of water vapor, with O_2 molecules. Reactions with O atoms and OH radicals are among the main HO_2 decay processes in the afterglow. The predicted HO_2 number density is sensitive to the O_2 fraction in the mixture, as expected. The sensitivity to the water vapor fraction is significantly lower, resulting in about a 25% change when the water vapor fraction is varied by a factor of 4. HO_2 is also detected when O_2 is not present in the mixture. In this case, it is generated primarily by the recombination of OH radicals, via the formation of H_2O_2. The results indicate that CRDS can also be used for HO_2 and other plasma chemical reaction product measurements in atmospheric pressure plasma jets impinging on a liquid water surface in ambient air. These measurements are currently underway.

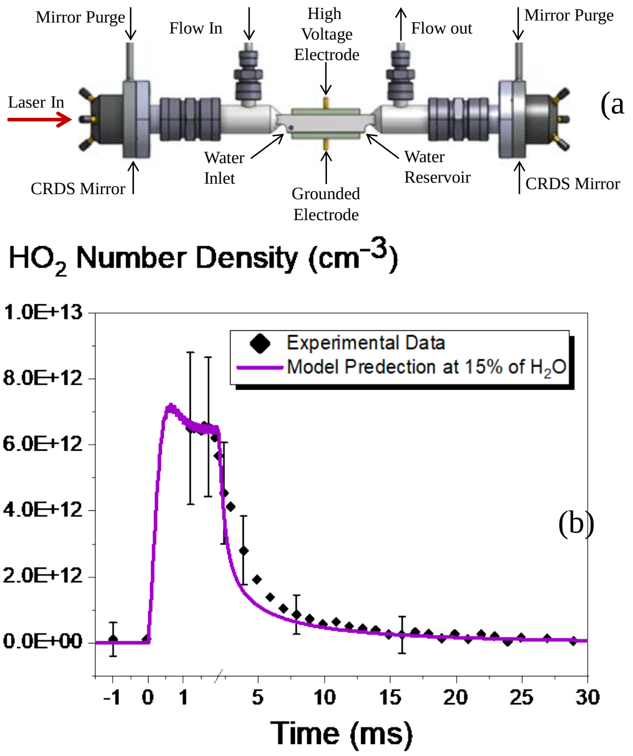

Figure 1 - (a) Schematic of CRDS plasma flow reactor, (b) Comparison of experimental HO_2 number density with kinetic modeling predictions.

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[II-11] Reactor Setup and Experimental Methods to Deduce Reaction Pathways and Timescales in Atmospheric Pressure Plasma Catalysis

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Investigation of gas-phase and surface-mediated reaction pathways and timescales for atmospheric pressure plasma catalysis requires a reactor setup that produces controllable, quantifiable densities of desired reactive species, allows transport of reactive species from the plasma to the catalyst, minimizes gradients in species densities, and exhibits well-defined hydrodynamics. Here, we discuss the reactor setup we used to investigate pathways and timescales for NH_3 synthesis by plasma catalysis and how it accomplishes these goals.

We use a radiofrequency (RF)-driven atmospheric plasma jet to produce measurable densities of radical (N, H) and vibrationally excited species ($N_2(v)$) and place nonporous, unsupported metal wools (Fe, Ni, and Ag) downstream of the discharge to catalyze reactions among the plasma-derived species. We quantify densities of N, H, $N_2(v)$, and NH_3 by molecular beam mass spectrometry (MBMS) using previously developed background subtraction and threshold ionization techniques [1]. Reactive species are transported from the plasma plume to the catalyst bed with millisecond (ms) or shorter convection times to prevent excessive quenching in the gas phase from the tip of the plume to the catalyst bed.

To analyze pathways for NH_3 production, we measured species densities at the inlet (Figure 1a) and outlet (Figure 1b) of the reactor and performed a mass balance to compare NH_3 production with consumption of reactive species. We show that NH_3 formation correlates linearly with N consumption across a range of process conditions that have sufficient H and H_2 densities over Fe, Ni, and Ag. To isolate the effects that H and H_2 have on NH_3 production, we introduce a setup (Figure 1c) that feeds Ar and N_2 through the plasma jet to produce N and feeds H_2 after the plasma jet to prevent H formation. In this reactor setup, NH_3 yield from N is at least 50% lower than the setup where H is present, indicating that H is necessary to selectively produce NH_3 from N.

To analyze timescales for NH_3 production, we vary the residence time in the catalyst bed by varying the bed length and flow rate at conditions where consumption of reactive species is below complete conversion. We note that the timescale for NH_3 formation is the same as N consumption, further demonstrating that reactions involving N are responsible for NH_3 formation at our reaction conditions.

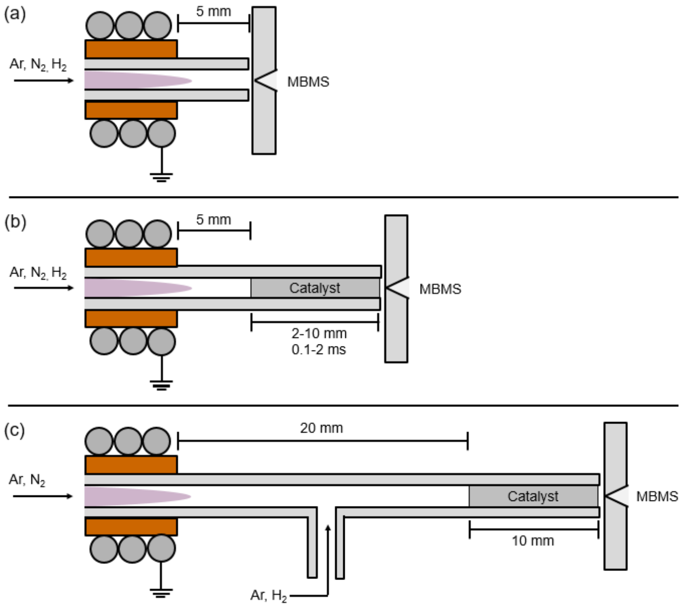

Figure 1 – Setups used to measure species densities at the (a) inlet and (b) outlet of a catalyst bed with different residence times. (c) Setup used to investigate reactions between N and H_2

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[II-12] Propagation of Surface Ionization Waves on Dielectric Substrates with Simple Patterns

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The interaction of atmospheric pressure plasma jets (APPJs) with dielectric surfaces is important to applications ranging from plasma catalysis to biotechnology.[1,2] The propagation of ionization waves produced by an APPJ was investigated as they impact dielectric surfaces of different thicknesses and geometric barriers and convert to surface ionization waves (SIWs). The APPJ was powered by a positive ns-DC pulse 500ns width at 1kHz repetition with helium as the working gas flowing into room air. Imaging was performed using an ICCD camera.

SIW propagation along different dielectric materials covering a ground plane as a function of thickness was studied. Thickness and dielectric constants, 0.15 -10mm, and ε=5.05-9.4, respectively were studied. These studies show SIW velocity is strongly dependent on target thickness and dielectric constant, increasing 3x for thicknesses of 0.15mm to 1.65mm. For thicknesses greater than 1.65mm the velocity plateaus. SIW velocity ranges from $1.5\text{-}4.5x10^4$ m/s. Axial velocities of incident ionization waves ranged from $1.55\text{-}2.0x10^5$ m/s. The velocities of SIW is also shown to decrease with increasing dielectric constant, which is consistent with predictions in literature.[3]

SIW propagation has also been studied for simple geometric barriers such as steps and single channels. For these experiments, the ICCD camera is placed along the axis of the plasma source and can image the SIW as it expands radially along the dielectric target surface. For simple steps it is seen that when the SIW meets a step-up boundary, it requires a SIW with higher pulse energy to overcome than a step-down boundary. The portion of the SIW that does make it down the step has higher light emission than SIWs that did not meet a barrier.

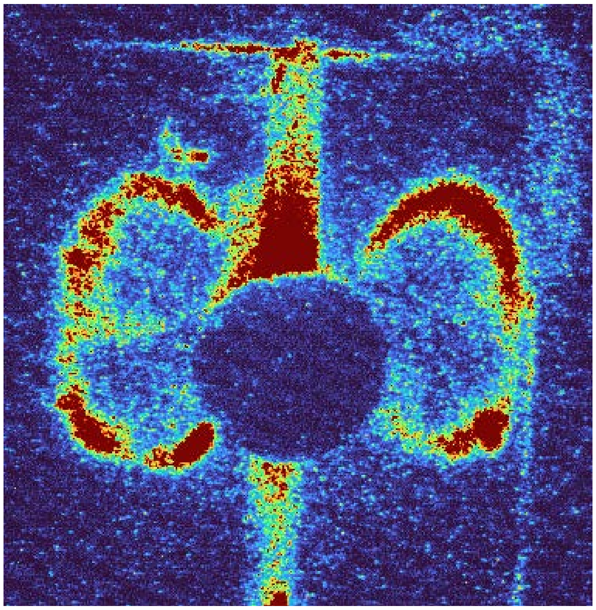

Figure 1 –The dark circle in the center of the image is a blocking plate to prevent ICCD saturation.

In the case of single channels, the SIW is initiated inside of the channel. In this case, only a portion of the wave escapes the channel and propagates outward. **Figure 1** shows the result of the escaping ionization wave. Some energy remains within the channel and propagates along it. The angle θ that defines the escape of the SIW from the channel shows a dependence on SIW energy. Supported by the US-DOE Center for Plasma Interactions with Complex Interfaces DE-SC0020232

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