SLATT UNDERGRADUATE RESEARCH FELLOWSHIP
FINAL REPORT

SCHOLAR NAME:	Jeff Secrist
FACULTY ADVISOR:	David B. Go
PROJECT PERIOD:	5/17/22 — 7/22/22
PROJECT TITLE:	Characterization of Plasma-Catalysis Systems for Upcycling of Light Hydrocarbons
CONNECTION TO ONE OR MORE ENERGY-RELATED RESEARCH AREAS (CHECK ALL THAT APPLY):	**(X) Energy Conversion and Efficiency** **( ) Sustainable and Secure Nuclear** **( ) Smart Storage and Distribution** **( ) Transformation Solar** **( ) Sustainable Bio/Fossil Fuels** **( ) Transformative Wind**

MAJOR GOALS AND ACCOMPLISHMENTS

Summarize your research goals and provide a brief statement of your accomplishments (no more than 1-2 sentences). Indicate whether you were able to accomplish your goals by estimating the percentage completed for each one. Use the next page for your written report.

RESEARCH GOALS	ACTUAL PERFORMANCE AND ACCOMPLISHMENTS	% OF GOAL COMPLETED
Learn how to operate the reactor to collect electrical and optical data	Learned how to generate a dielectric barrier discharge while collecting electrical data and used optical emission spectroscopy to extract optical data.	100
Extract relevant properties of the plasma through electrical and optical characterization	Using electrical characterization, I was able to extract the number of filaments, average current per filament, and areal fraction of the plasma. Using optical characterization, I was able to observe the spectra and under some circumstances, was able to extract vibrational and rotational temperature of molecules, electron density, and electron temperature.	75
Understand how changing operating conditions such as power, temperature, flow rate, and gas composition might affect the plasma	Worked on understanding temperature inhibition of methane conversion by varying power and temperature with three different gas mixtures and electrically characterizing the plasma.	75

RESEARCH OUTPUT

Please provide any output that may have resulted from your research project. You may leave any and all categories blank or check with your faculty advisor if you are unsure how to respond.

CATEGORY	INFORMATION
EXTERNAL PROPOSALS SUBMITTED	(Sponsor, Project Title, PIs, Submission Date, Proposal Amount)
EXTERNAL AWARDS RECEIVED	(Sponsor, Project Title, PIs, Award Date, Award Amount)
JOURNAL ARTICLES IN PROCESS OR PUBLISHED	A journal article is currently being prepared by my graduate student mentor Ibu Akintola.
BOOKS AND CHAPTERS RELATED TO YOUR RESEARCH	(Book Title, Chapter Title, Authors, Submission Date, Publication Date, Volume #, Page #s)
PUBLIC PRESENTATIONS YOU MADE ABOUT YOUR RESEARCH	(Event, Presentation Title, Presentation Date, Location) Summer Undergraduate Research Symposium, Understanding Methane Conversion in DBD Plasma Using Electrical Characterization, July 20, 2022, University of Notre Dame
AWARDS OR RECOGNITIONS YOU RECEIVED FOR YOUR RESEARCH PROJECT	(Purpose, Title, Date Received)
INTERNAL COLLABORATIONS FOSTERED	(Name, Organization, Purpose of Affiliation, and Frequency of Interactions)
EXTERNAL COLLABORATIONS FOSTERED	(Name, Organization, Purpose of Affiliation, and Frequency of Interactions)
WEBSITE(S) FEATURING RESEARCH PROJECT	(URL)

OTHER PRODUCTS AND SERVICES (e.g., media reports, databases, software, models, curricula, instruments, education programs, outreach for ND Energy and other groups)	(Please describe each item in detail) I helped represent the Go Research Group at Science Alive, an event for children held at St. Joe County Public Library where research groups from Notre Dame set up short demonstrations of interesting science phenomenon.

RESEARCH EXPERIENCE

Please let us know what you thought of your research experience: Did this experience meet your expectations? Were lab personnel helpful and responsive to your needs? What else could have been done to improve your experience or achieve additional results?

My research experience through the Slatt Fellowship was a great learning experience for me and helped me grow as a researcher within the engineering field. I worked closely with Ibu Akintola, a graduate student, and Jinyu Yang, a postdoc researcher, and my research was overseen by Professor David Go. I have gained a deeper understanding of the importance of collaboration in research. I attended weekly individual meetings with Professor Go as well as research group meetings and plasma catalysis meetings that other collaborators attended. I learned the benefits of working alongside other experts in the field to generate new ideas and achieve research results. The experience was overall positive because I came to enjoy the research process and will continue with this research moving forward.

FINAL WRITTEN REPORT

(Please use the space below to describe your research project and objectives, any findings and results you can share, and graphs, charts, and other visuals to help us understand what you achieved as a result of this research experience.)

Introduction:

This research project is funded by the Department of National Energy (DOE) National Energy Technology Laboratory (NETL) (DE-FE0031862). The goal of the project is to use a plasma-catalytic system to create value-added liquid chemicals from light hydrocarbons. In the oil and gas industry, the process of flaring leads to a global, annual loss of more than 140 billion m^3 of natural gas and emits more than 400 metric tons of CO_2 to the environment.[1] Plasma-catalytic systems present one potential way to harness the energy from natural gas and reduce greenhouse gas emissions.

Non-thermal plasmas (NTPs) produce highly reactive chemical environments made up of electrons, ions, radicals, and vibrationally excited molecules.[2] These reactive species, when combined with catalysts, can help drive thermodynamically unfavorable chemical reactions at low temperatures and atmospheric pressure. In order to effectively create these plasma catalytic systems, it is imperative that there is a fundamental understanding of the plasma-phase chemistry alone. While there have been many studies on plasmas, there is limited understanding on how changing operating conditions (e.g., feed ratio, plasma power, operating temperature) affect the plasma properties and ensuing plasma chemistry. In this work, we generate nitrogen (N_2) and methane (CH_4), argon (Ar) and methane, and helium (He) and methane plasmas to understand the effects of varying plasma parameters and gas compositions on product formation. Relevant electrical measurements such as the number of filaments and average current per filament are studied to better understand the processes behind methane conversion. Our findings show an inhibition of CH_4 conversion with an increase in temperature, which seems independent of gas composition due to similar trends in the plasma properties in all three gas mixtures.

Experimental Method:

In this research, we were working with a dielectric barrier discharge (DBD). This DBD was generated by applying a high voltage on the order of kilovolts between two electrodes with one electrode being covered by a cylindrical quartz tube (a dielectric material). It utilizes an alternating current and the dielectric prevents a termination of charges. The alternating flow allows the discharge to consist of many filaments spread across the electrode area. The plasma current and voltage traces from a typical DBD are shown in Fig. 1 where the plasma current peaks correspond to the individual microdischarge filaments. These filaments are characterized by electron densities up to 10^{15} cm^{-3} and strong electric fields up to 10^5 V/cm which is why these filaments influence the plasma chemistry.[3] The filamentary nature of plasma has been used by Ozkan et al. to better understand CO_2 conversion.[4] My interest this summer was concerned with observing the filamentary nature of plasma and using it to better understand CH_4 conversion.

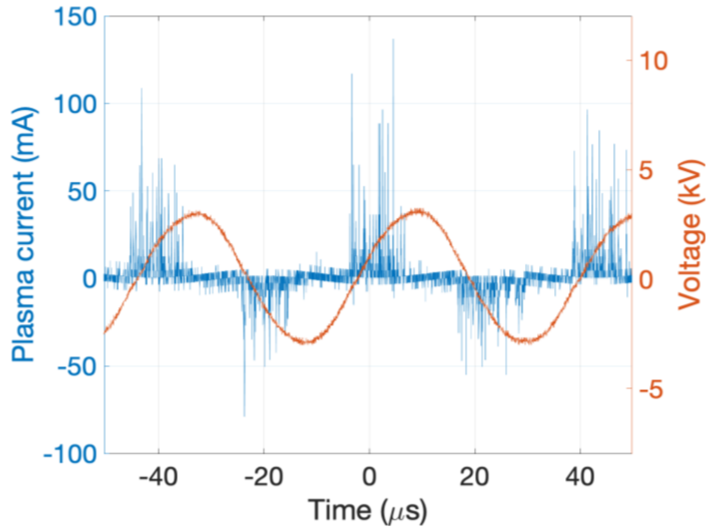

Figure 1: *Plasma current and applied voltage versus time.*

Results:

Understanding CH_4 conversion is critical to the overall goal of this research project. CH_4 conversion is defined as:

$$\frac{\text{moles of } CH_4 \text{ converted}}{\text{moles of } CH_4 \text{ in the feed}} \times 100\,\%.$$

In CH_4/N_2 plasma, CH_4 conversion increases with an increase in power but decreases with an increase in temperature as depicted in Fig. 2. CH_4/Ar plasma also follows this same trend with an increase in temperature.

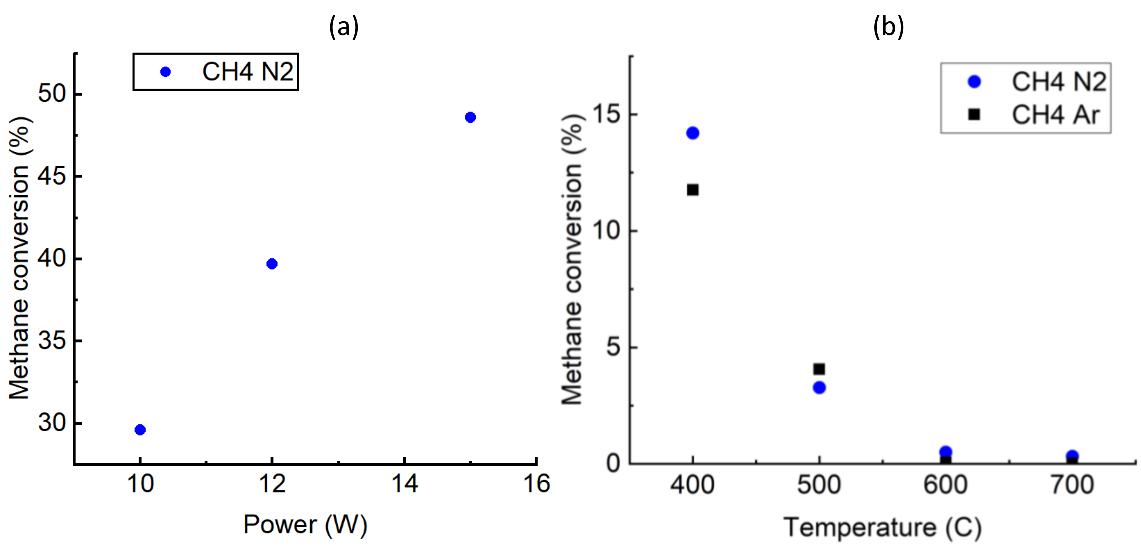

Figure 2: *(a) Methane conversion of 50 sccm (1-1 CH_4/N_2) discharges at 500 °C as a function of power. (b) Methane conversion of 50 sccm (1-1 CH_4/N_2, Ar) discharges at 10W as a function of temperature.*

The filamentary nature of discharges was then studied using different gas mixtures to observe how the electrical properties of the plasma changed as a function of power and temperature. For CH_4/N_2 and CH_4/Ar plasmas, the number of filaments increased as power was increased but the average current per filament stayed relatively constant as shown in Fig. 3. The increase in voltage led to an increase in current through an increase in the number of filaments, *not* through an increase in the current per filament.

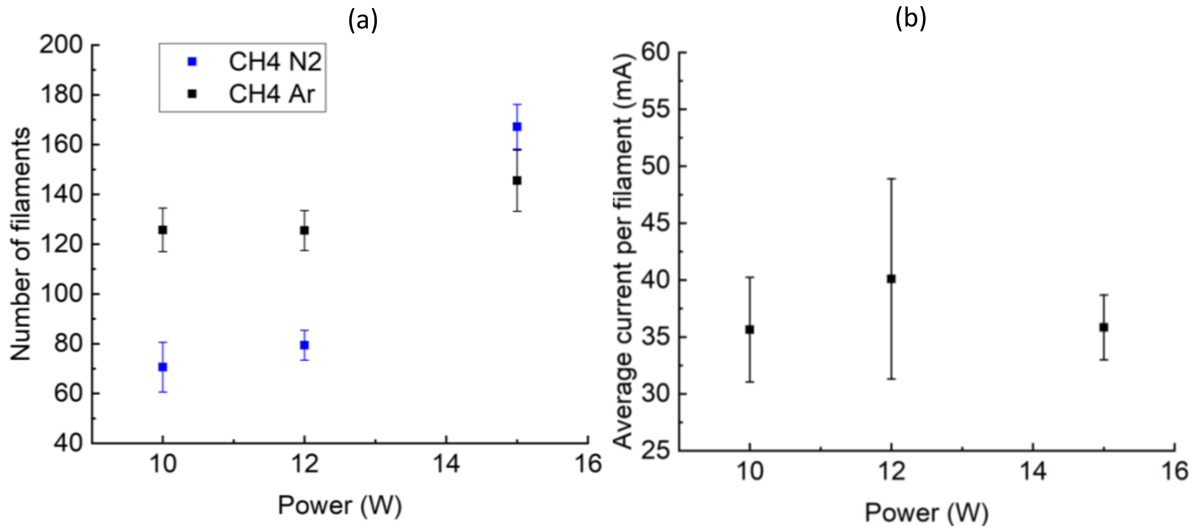

Figure 3: *(a) Number of filaments of 50 sccm (1-1 CH_4 /N_2, Ar) discharges at 500 ºC as a function of power. (b) Average current per filament of 50 sccm (1-1 CH_4 /Ar) discharges at 500 ºC as a function of power.*

However, when temperature increases, there is a reduction in both the number of filaments and the average current per filament for CH_4/N_2, CH_4/Ar, and CH_4/He gas mixtures shown in Fig. 4. At 600 and 700 ºC, the number of filaments for CH_4/Ar and CH_4/He plasma dramatically reduces, possibly indicating a mode change towards a homogenous glow discharge. According to Fig. 2b, these plasma conditions are shown to be ineffective for CH_4 conversion.

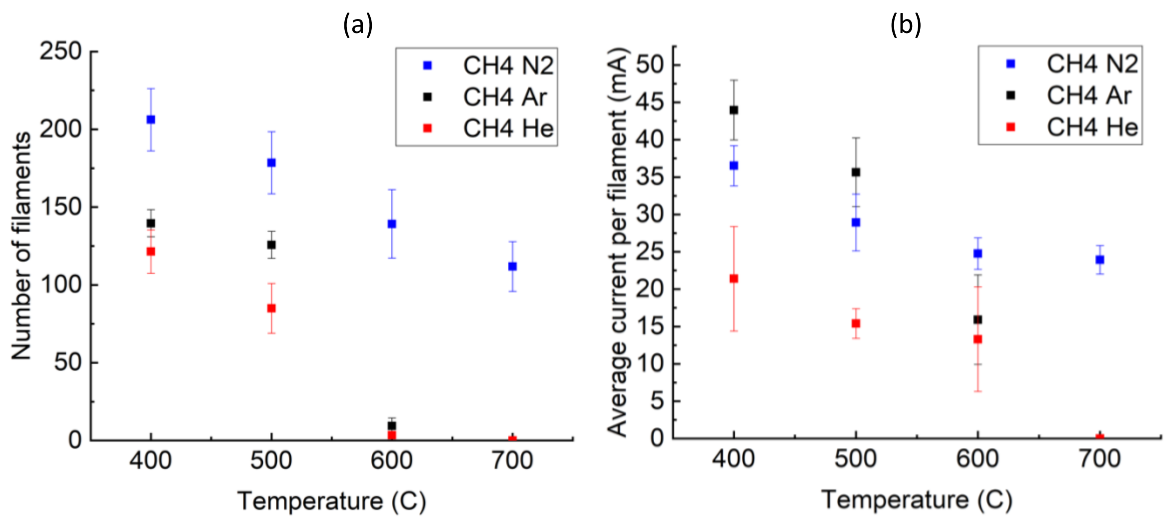

Figure 4: *(a) Number of filaments and (b) average current per filament of 50 sccm (1-1 CH_4 /N_2, Ar, He) discharges at 10W as a function of temperature.*

Conclusions:

Power and temperature both play a key role in the behavior of plasma and the conversion of CH_4. It appears that the gas mixture does not affect trends in filamentary behavior. From our work, a relationship can be drawn between CH_4 conversion and the filamentary nature of the discharge. When the filamentary behavior of the plasma is lost at 600 and 700 ºC, the plasma is very ineffective at converting CH_4. This relationship between the number of filaments and CH_4 conversion is not proven to be causative so future work may look at filament lifetime and the charge carried per filament. This may lead to a better understanding of the temperature inhibition of methane conversion.

References:

[1] National Energy Technology Laboratory. *Process intensification by a one-step, plasma-assisted synthesis of liquid chemicals from light hydrocarbons*. netl.doe.gov. Retrieved July 6, 2022, from https://netl.doe.gov/node/9643

[2] Go, D. B. (2018). *Ionization and ion transport : a primer for the study of non-equilibrium, low-temperature gas discharges and plasmas*. Morgan & Claypool Publishers.

[3] Peeters, F. J. J. (2015). *The electrical dynamics of dielectric barrier discharges*. Technische Universiteit Eindhoven.

[4] Ozkan, A *et al*. 2016 *Plasma Sources Sci. Technol*. **25** 025013