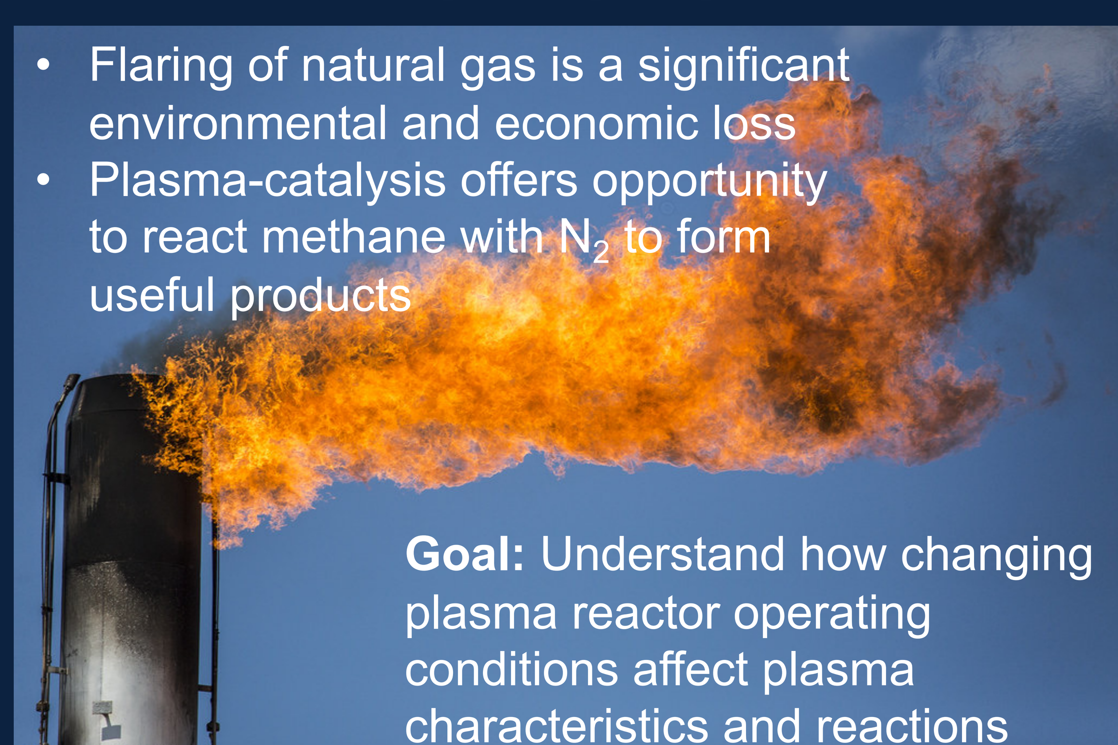
Understanding Temperature Inhibition of Methane Conversion in DBD Plasma Using Electrical Characterization and Optical Emission Spectroscopy

Ibukunoluwa Akintola[1], Gerardo Rivera-Castro[2], Jinyu Yang[1], Jeff Secrist[2], Jason C. Hicks[2], David B. Go[1,2]

[1]Department of Aerospace & Mechanical Engineering, [2]Department of Chemical & Biomolecular Engineering
University of Notre Dame

Motivation



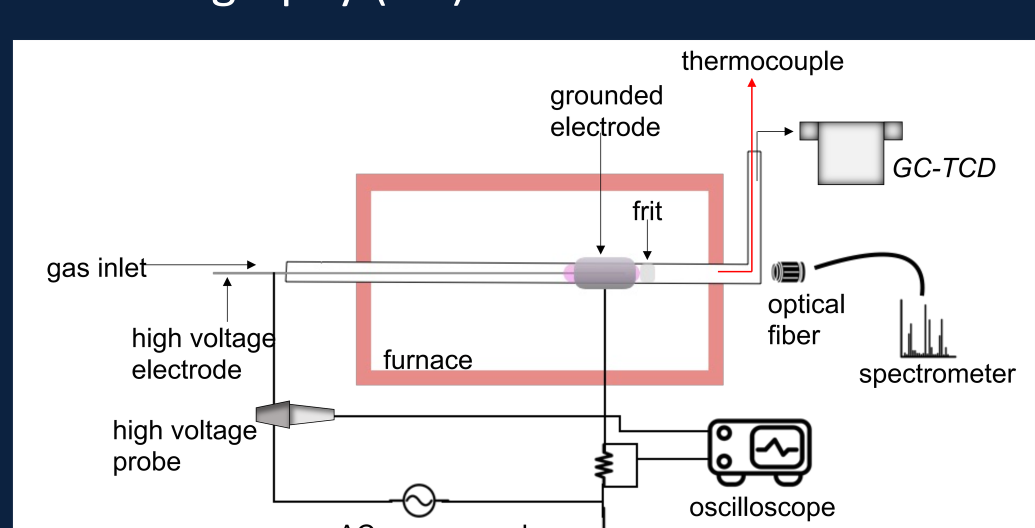
- Flaring of natural gas is a significant environmental and economic loss
- Plasma-catalysis offers opportunity to react methane with N_2 to form useful products

Goal: Understand how changing plasma reactor operating conditions affect plasma characteristics and reactions

www.texasstandard.org

Experimental Approach

- A cylindrical flow-through reactor with an integrated dielectric barrier discharge (DBD) was used
- Plasma characterized using optical emission spectroscopy (OES) and electrical measurements
- Reaction products characterized using gas chromatography (GC)

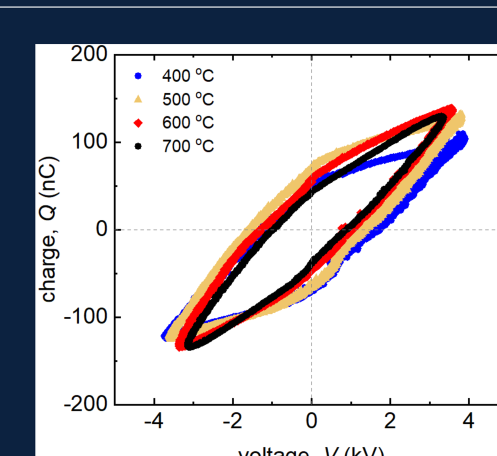


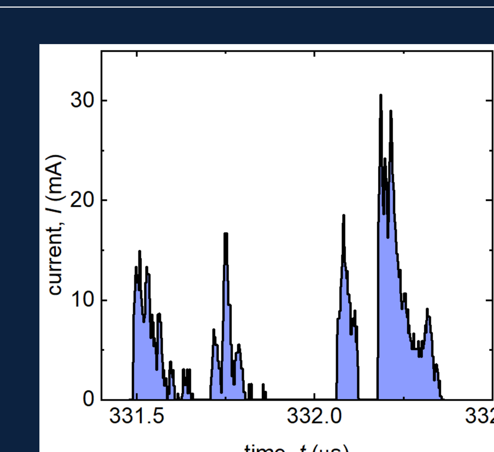
Schematic of the reactor setup for optical and electrical characterization of the plasma and product formation analysis.

Methodology

$N_2 + CH_4 \rightarrow$ N-C Products

- CH_4 conversion used as an indicator for product formation
- Q_{avg}-filament obtained from current/voltage trace
- ζ_{diel} calculated from slope of Lissajous curve

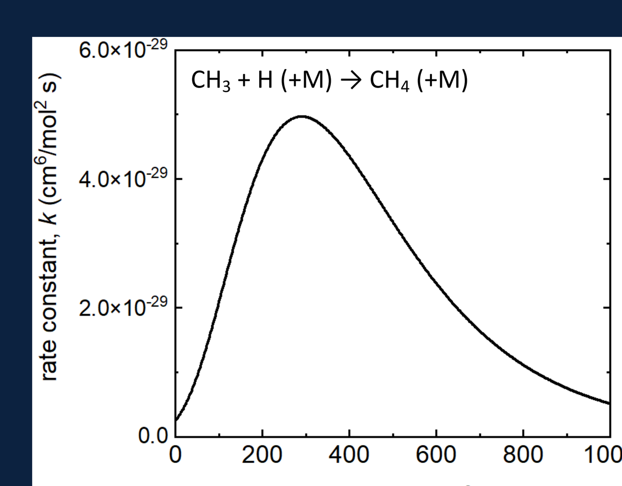




$$X_{CH_4} = \frac{C_{CH_4 in} - C_{CH_4 out}}{C_{CH_4 in}} \qquad Q_{avg} = \frac{\sum_{i=1}^{N_f}\left[\int_{t_{f,i}} I_i dt\right]_i}{N_f}$$

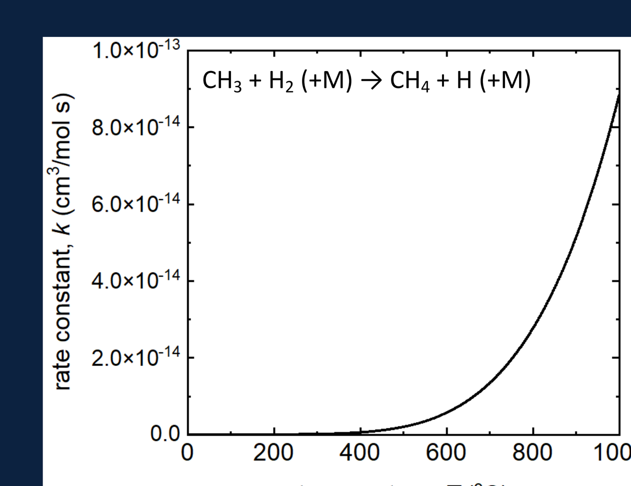
Results

- Conversion of methane decreases with increasing reactor temperature
- Changes to reaction chemistry with temperature which affect conversion
- ζ_{diel} shows increase in dielectric permittivity with temperature



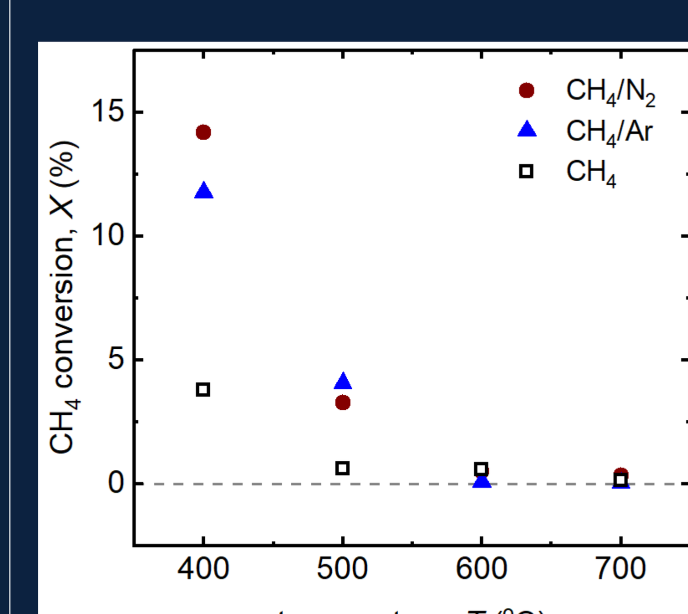
$$k(T) = (9.24\times10^{-23})\left(\frac{T}{298}\right)^{-8.82} e^{-\frac{(41310)}{RT}}$$

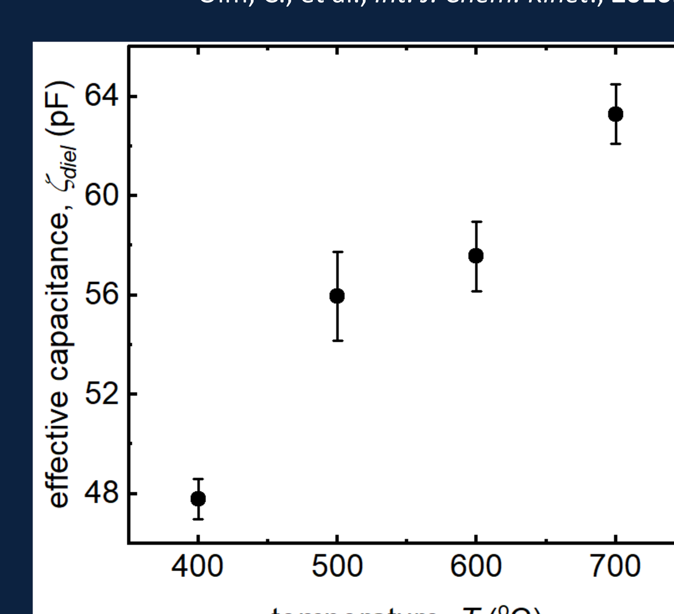
Olm, C., et al., *Int. J. Chem. Kinet.*, **2016**.

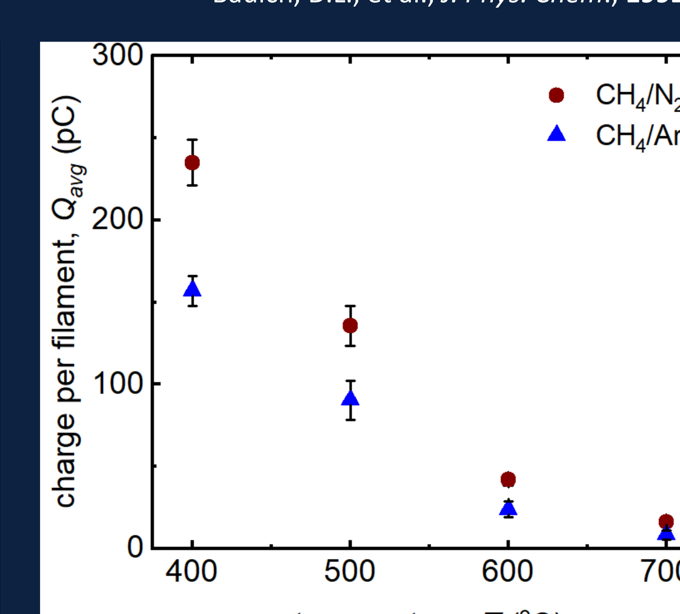


$$k(T) = (6.86\times10^{-14})\left(\frac{T}{298}\right)^{2.74} e^{-\frac{(39410)}{RT}}$$

Baulch, D.L., et al., *J. Phys. Chem.*, **1992**.

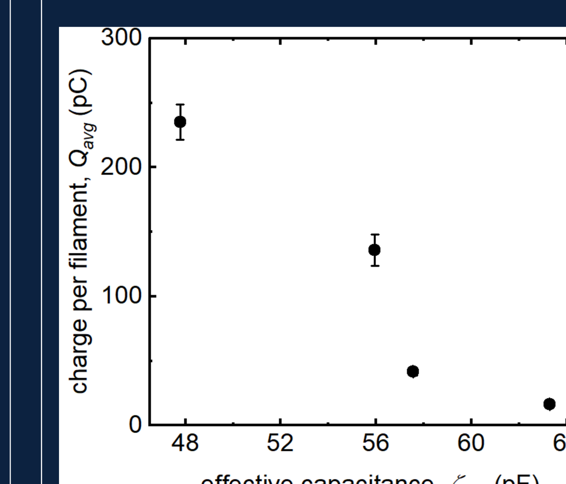


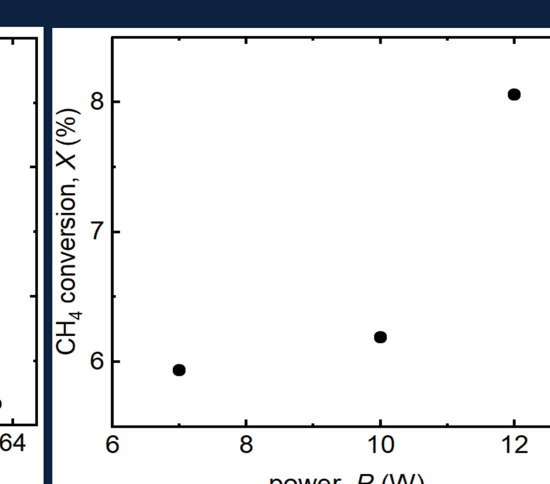


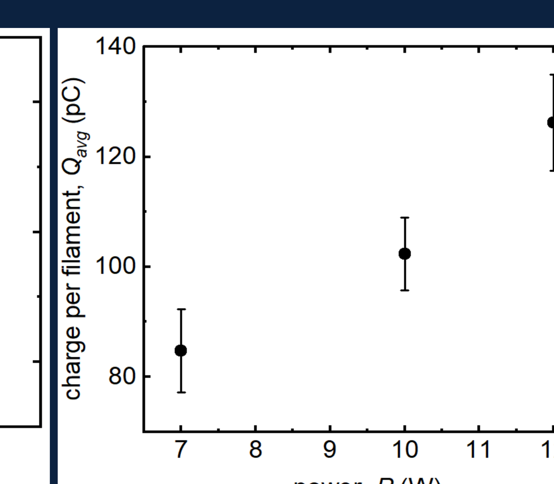


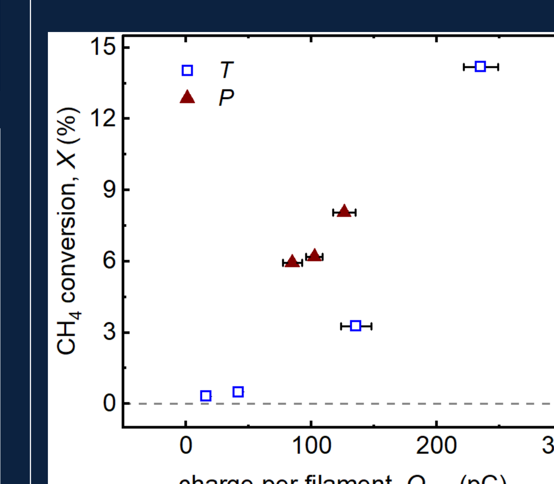
Experimental conditions: 50 sccm flow rate (1-1 CH_4/gas), 10 W power, Pressure: 1 atm

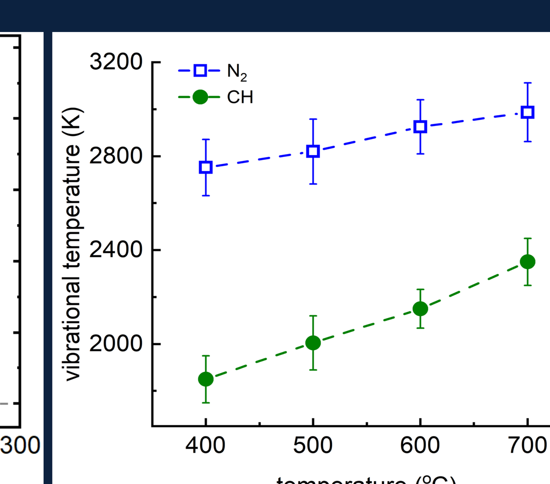
Results & Conclusions

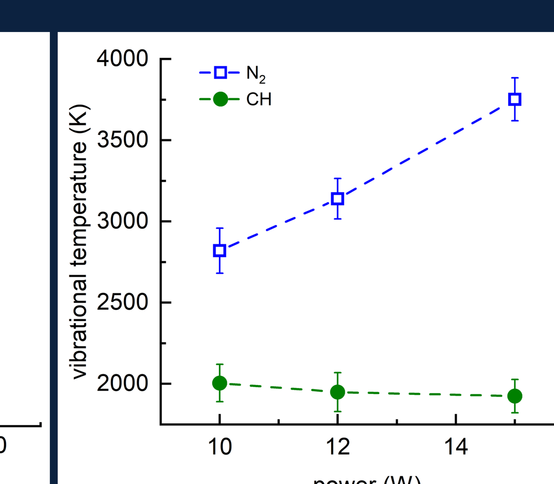












- Opposite trends observed for Q_{avg} as a function of temperature and power
- Q_{avg} follows same trend as conversation regardless of experimental conditions
- Unexpected inverse relationship between C-H vibrational temperature and conversion of methane
- Plasma properties also have good correlation with observed trends in conversion

Future Work: Understand the individual effects of reaction chemistry and plasma properties on conversion, determine how it changes with catalyst present, and if conversion inhibition is subject to CH_4 only

Acknowledgements:
This work was supported by the U.S. Department of Energy by National Energy Technology Laboratory under Award Number DE-FE0031862.