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

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Motivation

- Flaring of natural gas is a significant environmental and economic loss
- Plasma-catalysis offers opportunity to react methane with $\text{N}_2$ to form useful products

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

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

\[ \text{N}_2 + \text{CH}_4 \rightarrow \text{N-C Products} \]

- CH$_4$ conversion used as an indicator for product formation
- $Q_{\text{avg}}$-filament obtained from current/voltage trace
- $\psi_{\text{diel}}$ calculated from slope of Lissajous curve

\[ X_{\text{CH}_4} = \frac{c_{\text{CH}_4,\text{in}} - c_{\text{CH}_4,\text{out}}}{c_{\text{CH}_4,\text{in}}} \]

\[ Q_{\text{avg}} = \sum_{i=1}^{N_f} \left[ \int_{t_i}^{t_{i+1}} \frac{I(t)}{V(t)} \, dt \right] \]

Results

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

Results & Conclusions

- Opposite trends observed for $Q_{\text{avg}}$ as a function of temperature and power
- $Q_{\text{avg}}$ 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

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\begin{align*}
Q &= k(T) \left[ \text{gas} \right] \text{mol} / \text{cm}^3 \\
\text{gas} &= \text{CH}_3 + H (+M) \rightarrow \text{CH}_4 (+M)
\end{align*}
