# Hydrogen Purification and Recycling for an Integrated Oxygen Recovery System Architecture

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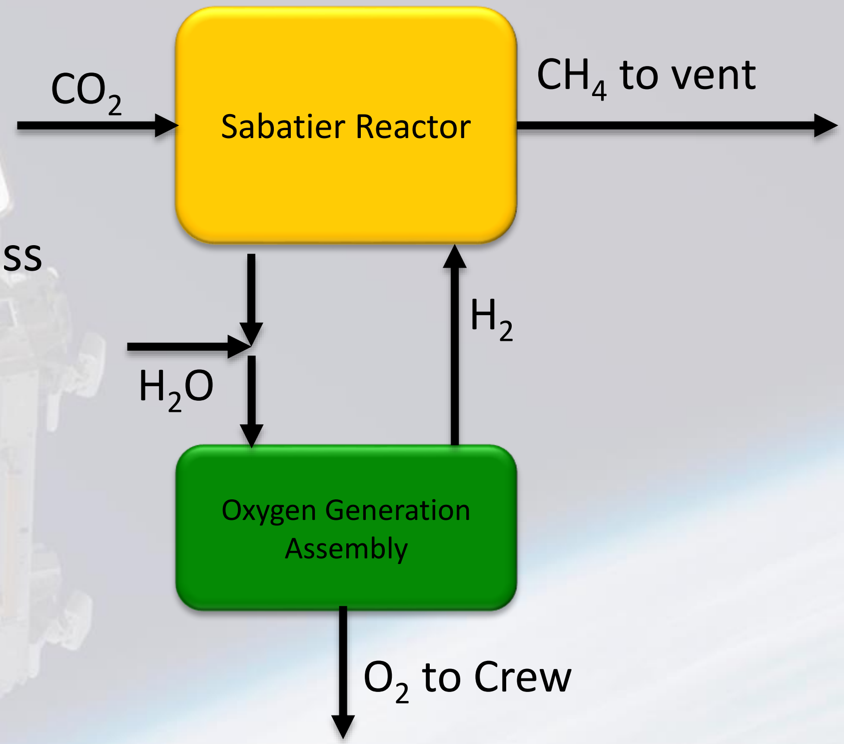
# Overview

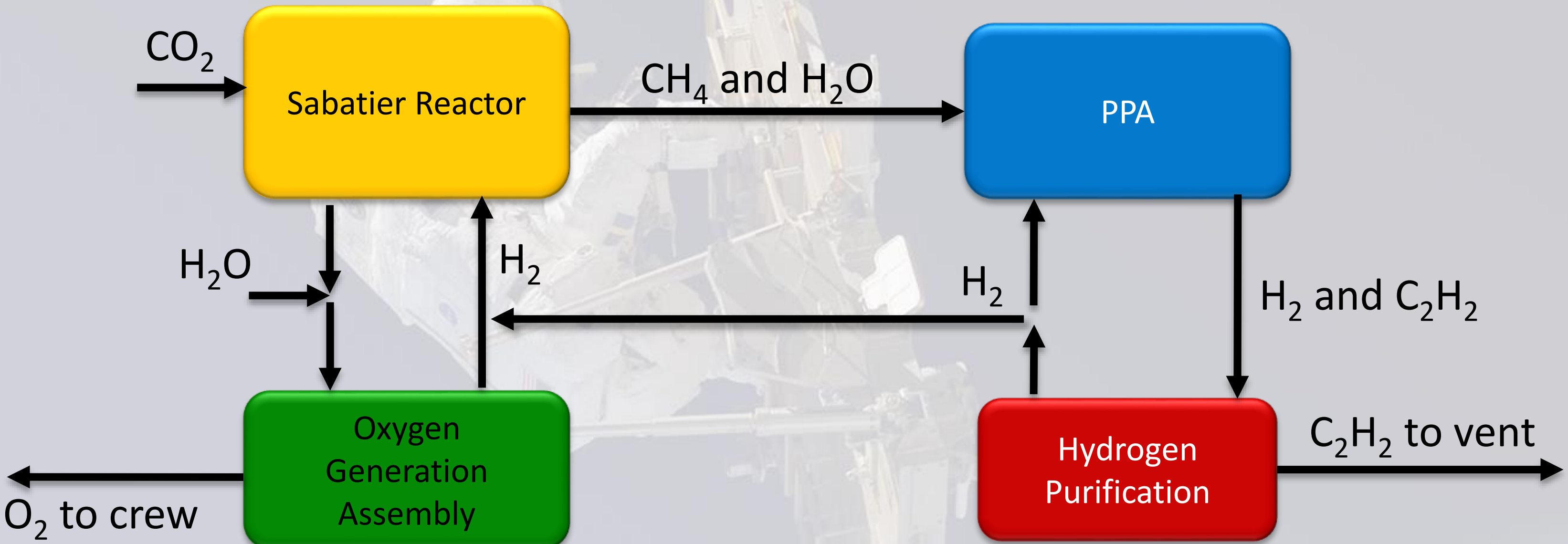
- Background
- Hardware
- Test Setup
- Results
- System Architectural Options
- Conclusion
- Acknowledgements

# State-of-the-Art

- Sabatier Reactor
  - $CO_2 + 4H_2 \rightarrow 2H_2O + CH_4$
  - Water product electrolyzed for oxygen
  - Methane product vented resulting in loss of hydrogen reactant
  - Theoretical recovery of ~54% of $O_2$ recovered from metabolic $CO_2$



# Sabatier Plus Post-Processing




- ~91% $O_2$ recovery from $CO_2$ possible

# PPA Technology Description

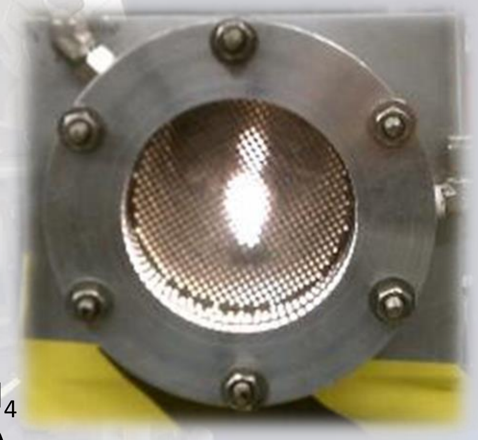
- Developed by UMPQUA Research Co.
- Methane converted to hydrogen and acetylene by partial pyrolysis in microwave generated plasma

- Targeted PPA Reaction:

  $2CH_4 \leftrightarrow 3H_2 + C_2H_2$

- Other reactions:

| | |
|---|---|
| $CH_4$ Conversion to Ethane | $2CH_4 \leftrightarrow H_2 + C_2H_6$ |
| $CH_4$ Conversion to Ethylene | $2CH_4 \leftrightarrow 2H_2 + C_2H_4$ |
| $CH_4$ Conversion to Solid C | $CH_4 \leftrightarrow 2H_2 + C(s)$ |
| CO Production | $C(s) + H_2O \leftrightarrow CO + H_2$ |
| CO Production | $CH_4 + H_2O \leftrightarrow CO + 3H_2$ |



$H_2/CH_4$ Plasma

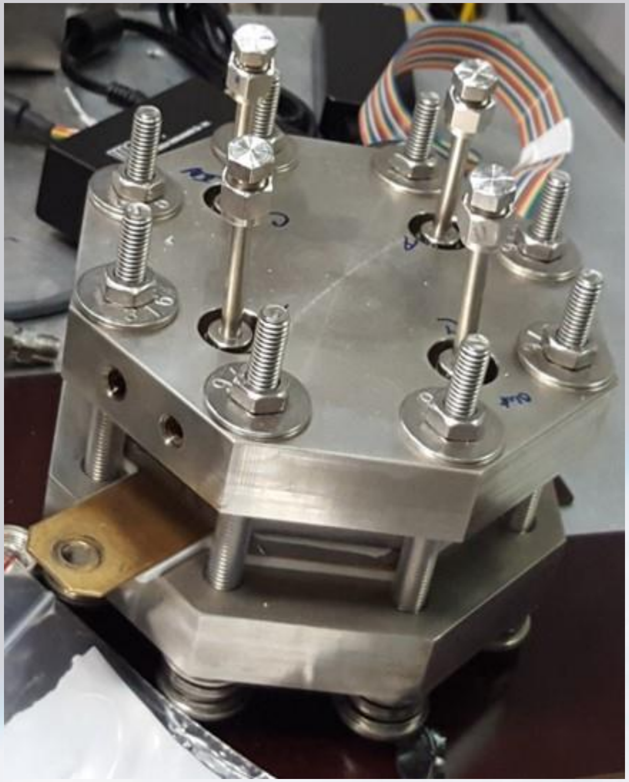

Plasma Pyrolysis Assembly

# Metal Hydride Hardware

- Hydrogen Components, Inc. Metal Hydride Canister
- $LaNi_{4.6}Sn_{0.4}$ metal hydride
- Designed for hydrogen storage

# Electrochemical Hardware

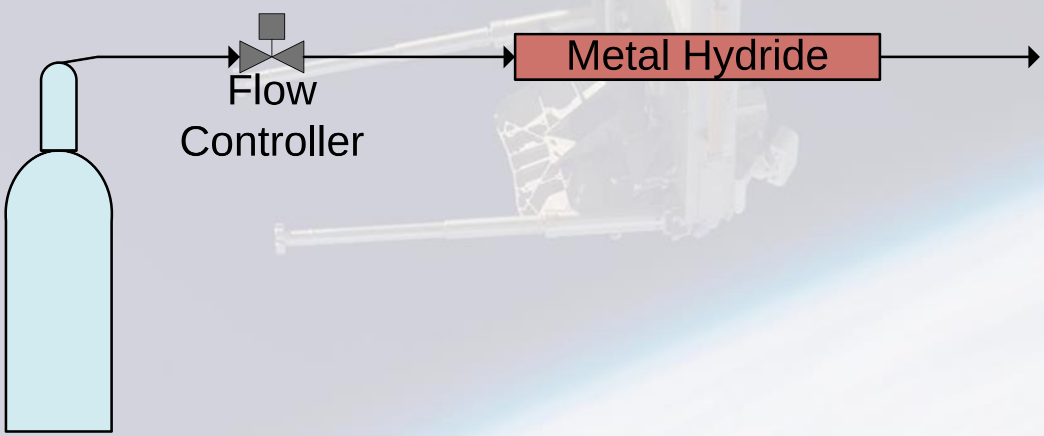
- Electrochemical hydrogen separation
  - $H_2$ electro-oxidized to protons and electrons
  - Protons are electro-reduced, recombined with electrons, in another chamber producing purified $H_2$
- Basic technology was well developed but not compatible with CO
  - CO would preferentially adsorb on catalytic electrodes and interfere with $H_2$ oxidation
- Sustainable Innovations developed electrolyte materials capable of operating above 150°C CO thermal desorption temperature
  - "Basic" and "Advanced" cell stacks delivered to MSFC



Sustainable Innovations Cell Stack

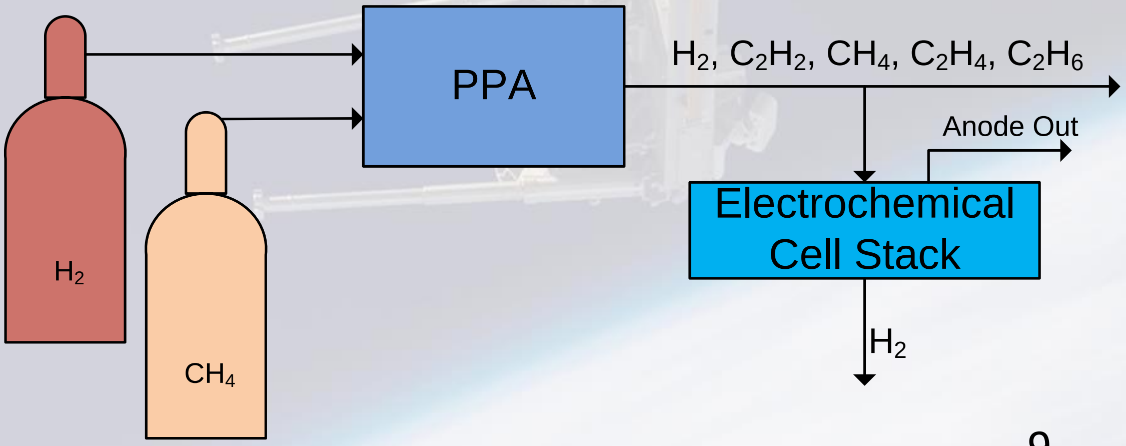
# Test Configurations

- Stand alone
  - Metal hydride to verify safety
    - Literature indicated other metal hydrides had potential to cause violent acetylene decomposition or metal-carbide formation
  - Tested with gas mixture containing 7% $C_2H_2$, 1% $CH_4$, and 92% $H_2$
  - Tested in Marshall Space Flight Center's Component Development Area, usually used for rocket engine component testing



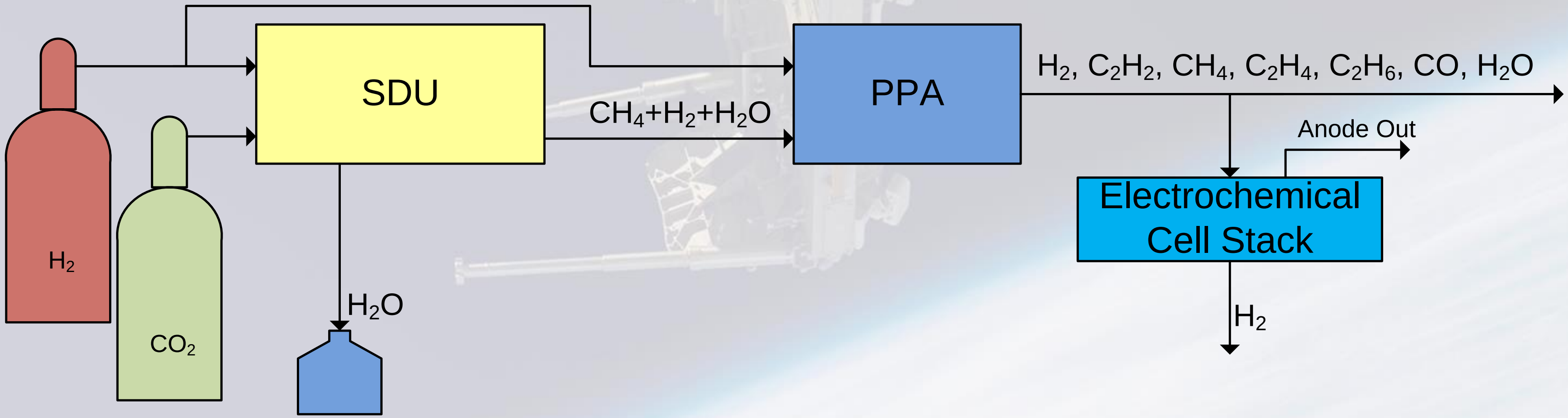
# Test Configurations

- **PPA + $H_2$ Purification**
  - Cell stacks integrated with 2nd Gen. PPA
  - PPA operated with ultra-high purity $H_2$ and $CH_4$ bottles
  - 1 Crew Member processing rate
  - 4:1 ratio of $H_2$:$CH_4$
  - 52 torr
  - 550 W microwave power
- PPA products contained $H_2$, $C_2H_2$, unreacted $CH_4$, $C_2H_4$, and $C_2H_6$
- No CO
- 100 standard milliliters per minute (SmLPM) to cell stack
- Evaluated $H_2$ product and process effluent



# Test Configurations

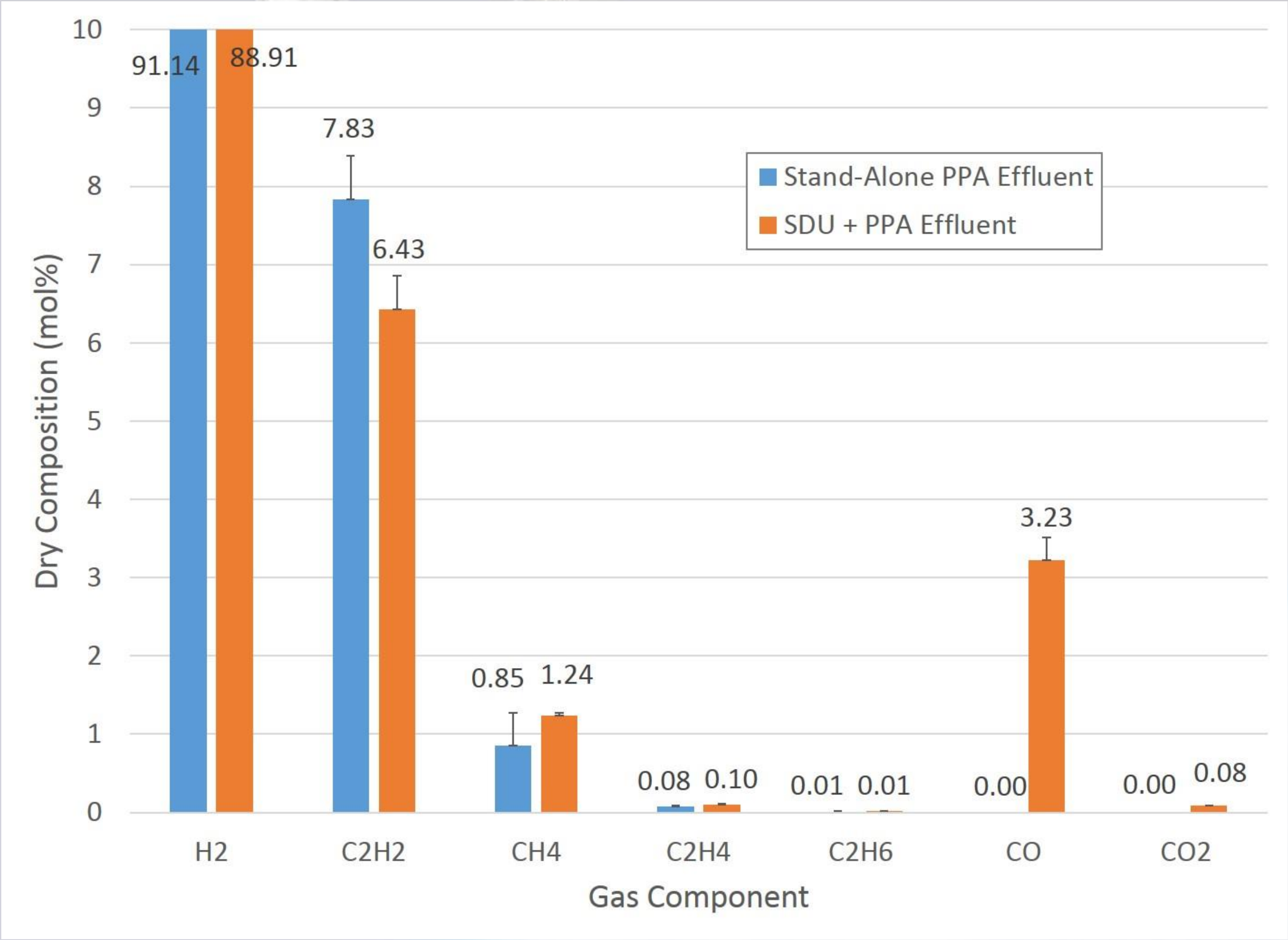
- **Sabatier Development Unit (SDU) + PPA + $H_2$ Purification**
  - Precision Combustion, Inc. SDU integrated upstream of PPA
  - SDU operated to produce 350 SmLPM $CH_4$ with no unreacted $CO_2$
  - Methane product containing 80 mol% hydrogen
  - Water vapor content dew point of 31°C
- PPA operated identically to PPA + $H_2$ testing
- PPA products contained all previously indicated components and CO and $H_2O$



# Metal Hydride Performance

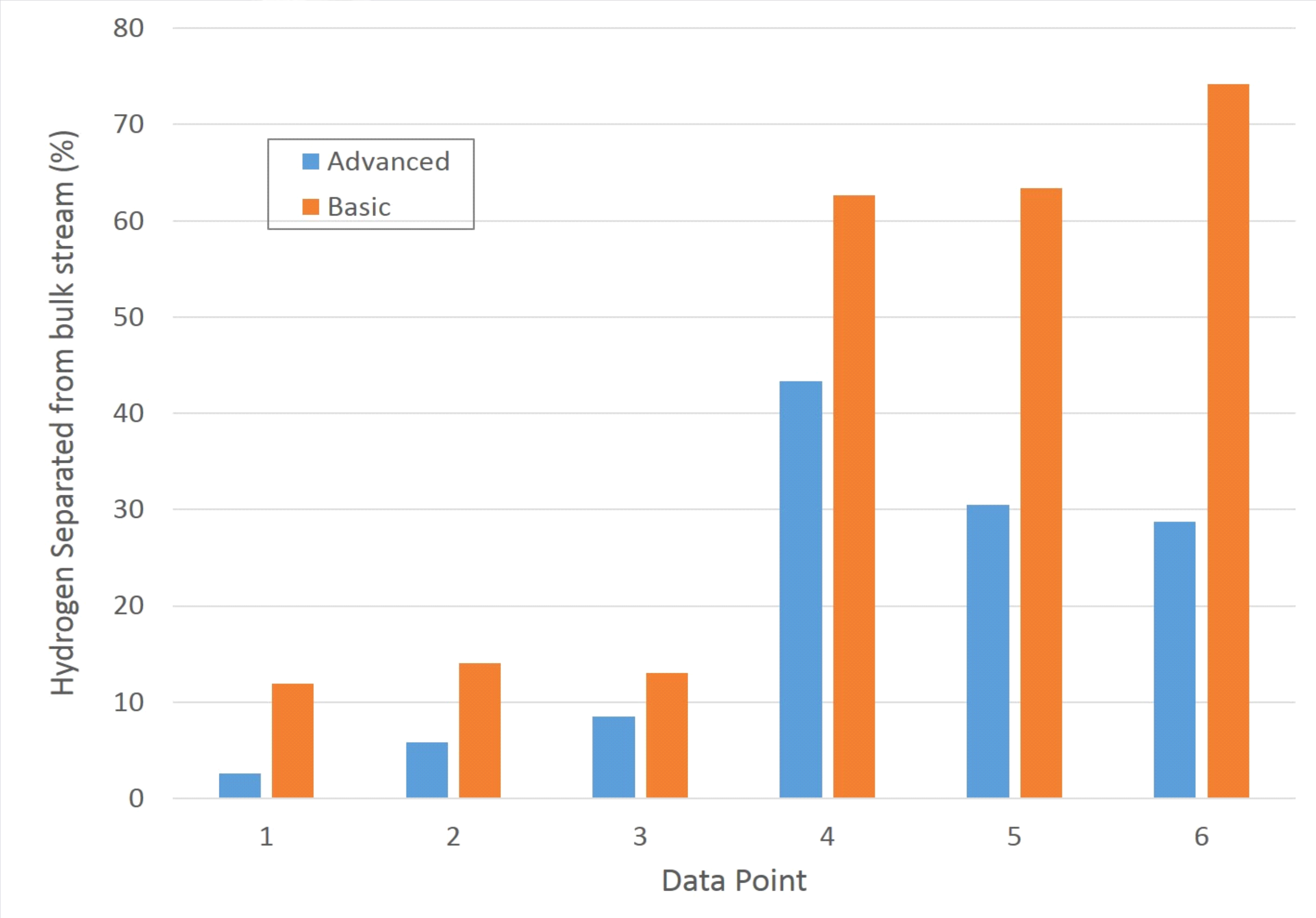
- No measurable pressure or temperature difference between pure $H_2$ runs and acetylene mixed gas runs
- No safety risk under expected operating conditions

# PPA effluent composition as a function of configuration



# $H_2$ separation performance comparison between Basic and Advanced cell stacks



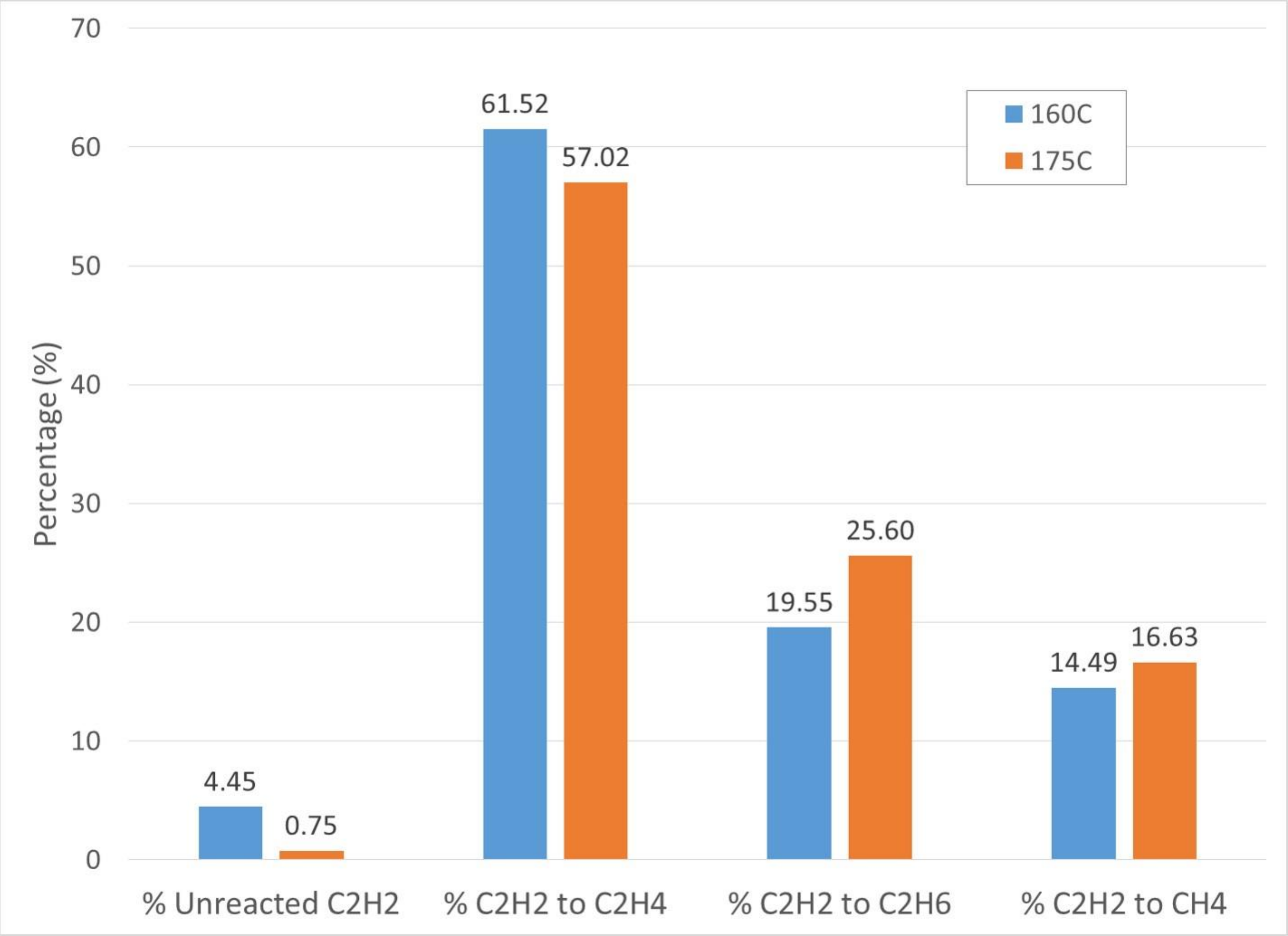



- Varied gas feed from PPA, stack temperature, inlet composition, and applied voltage
  - Conditions for each data point were identical
- **All recovered $H_2$ pure within measurable limits of µGC**

# Hydrogenation

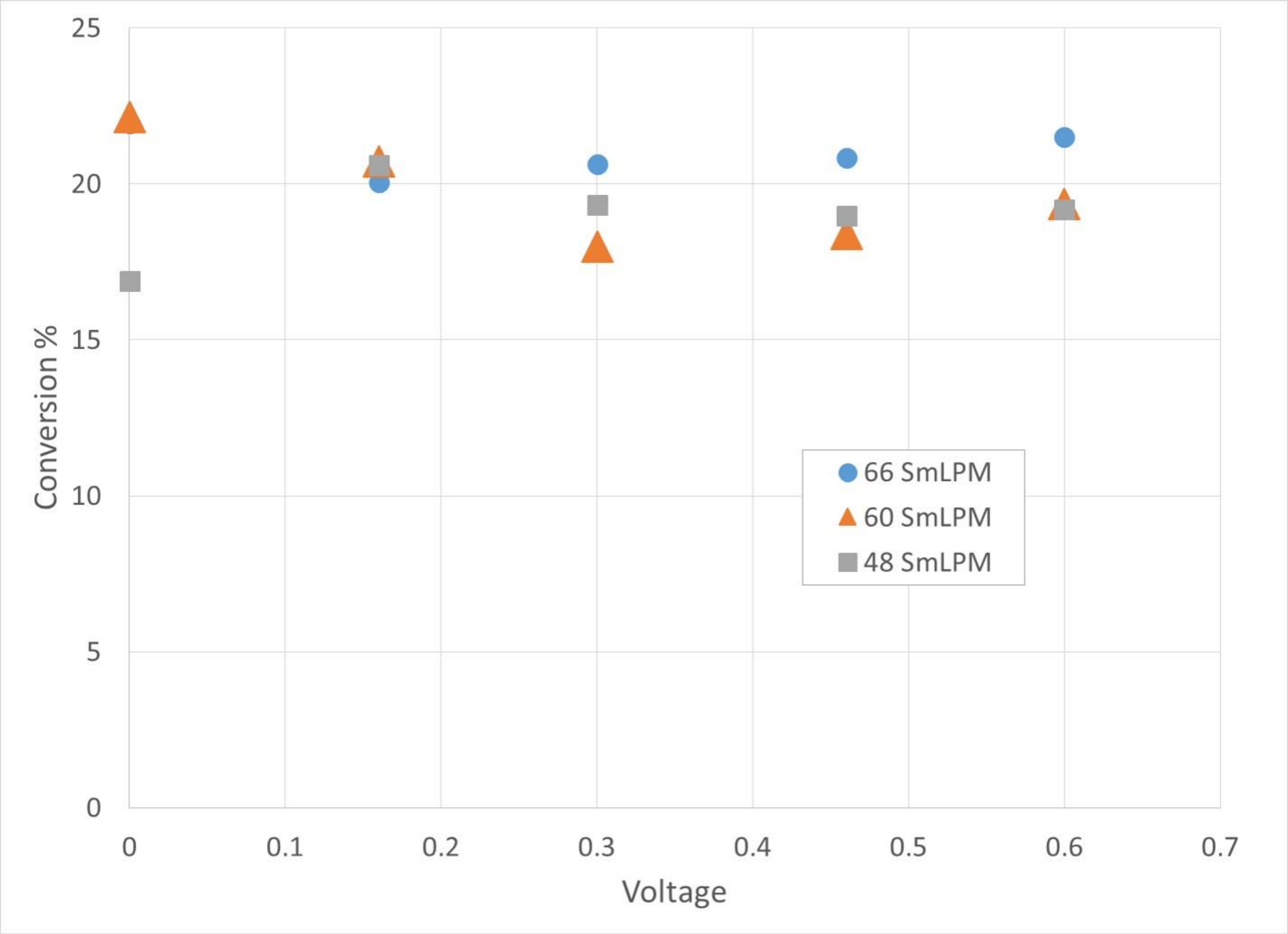
- Expected similar gas mix (minus $H_2$) leaving anode as entering
- High levels of $C_2H_4$ and $C_2H_6$ were observed with minimal or no $C_2H_2$
- Overall chemical equations:
  - $CH_4$ Conversion to Ethane $2CH_4 \leftrightarrow H_2 + C_2H_6$
  - $CH_4$ Conversion to Ethylene $2CH_4 \leftrightarrow 2H_2 + C_2H_4$
- Ethane Formation from $CH_4$ with free radical intermediates:
  $CH_4 + CH_4 \leftrightarrow CH_3^* + CH_3^* + H^* + H^* \leftrightarrow C_2H_6 + H_2$
  - $CH_4$ forms $CH_3^*$ free radicals which then recombine to form $C_2H_6$
  - $C_2H_6$ is converted to $C_2H_4$ and $C_2H_4$ is converted to $C_2H_2$
  - Reverse reactions also occur providing a mechanism for $C_2H_2$ hydrogenation to the other hydrocarbons

# Effect of temperature on $C_2H_2$ hydrogenation, Advanced Cell Stack

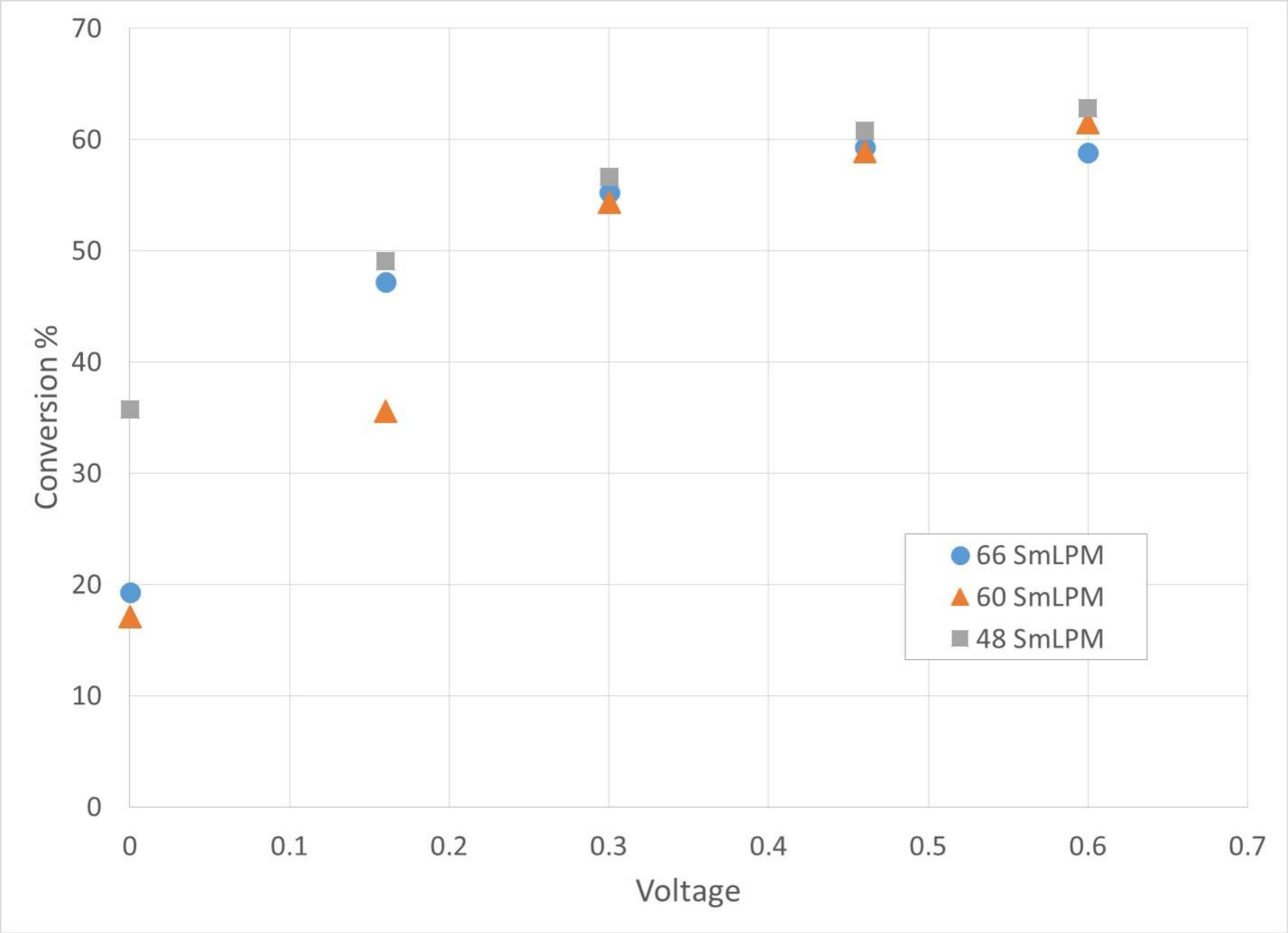


# Acetylene conversion to methane in Advanced cell stack as a function of voltage and anode feed rate.



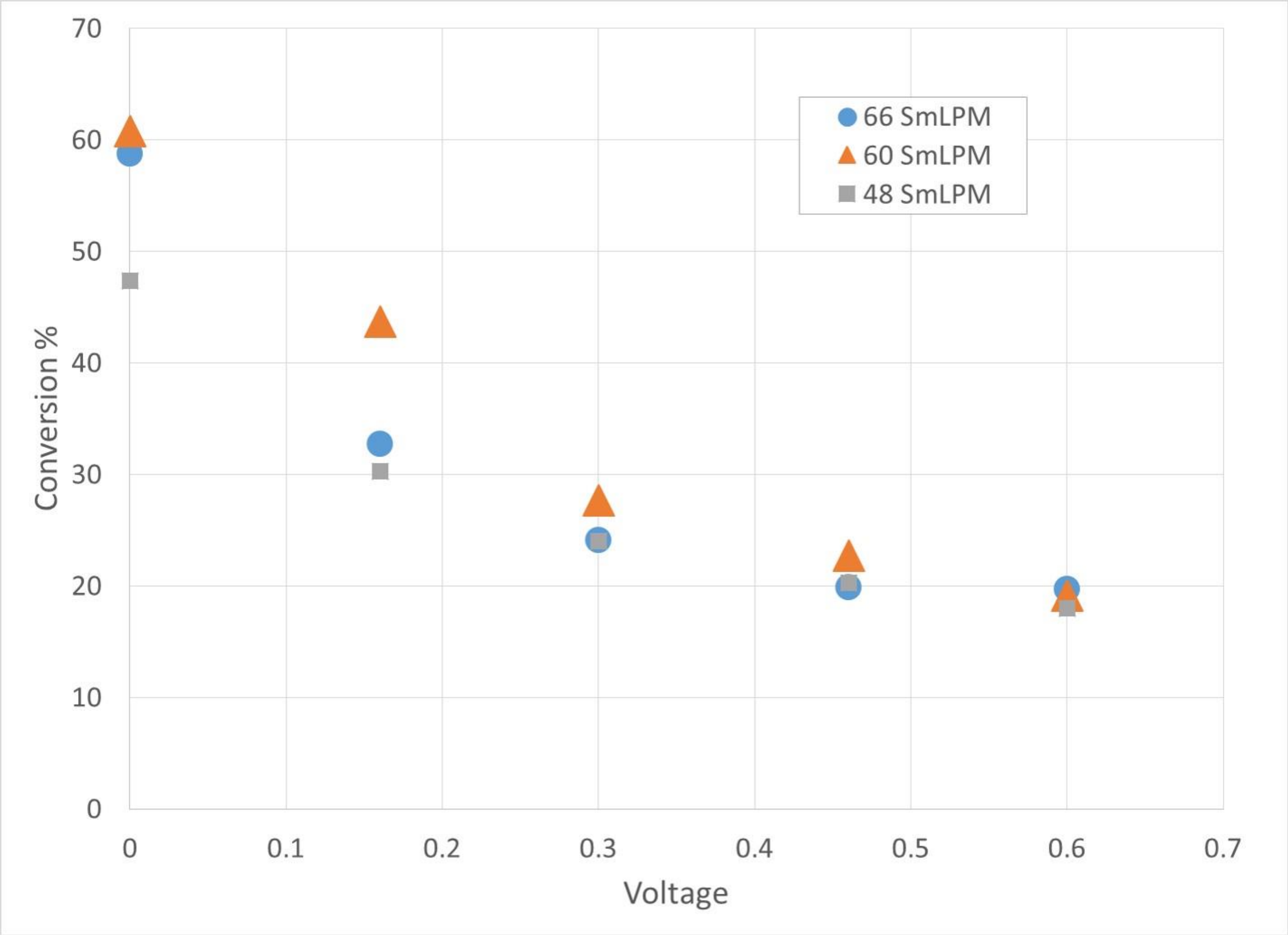


# Acetylene conversion to ethylene in Advanced cell stack as a function of voltage and anode feed rate.



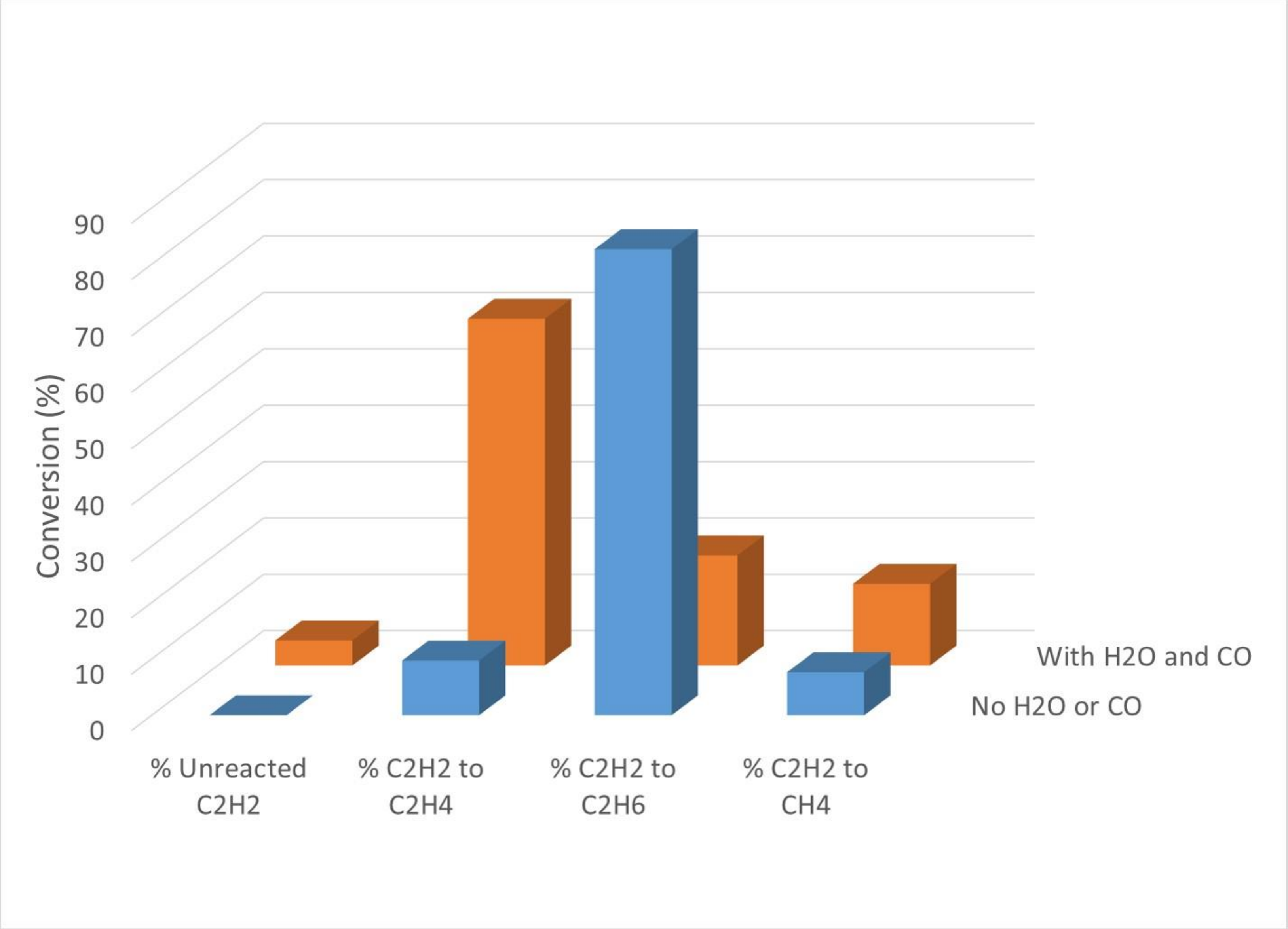
# Acetylene conversion to ethane in Advanced cell stack as a function of voltage and anode feed rate.



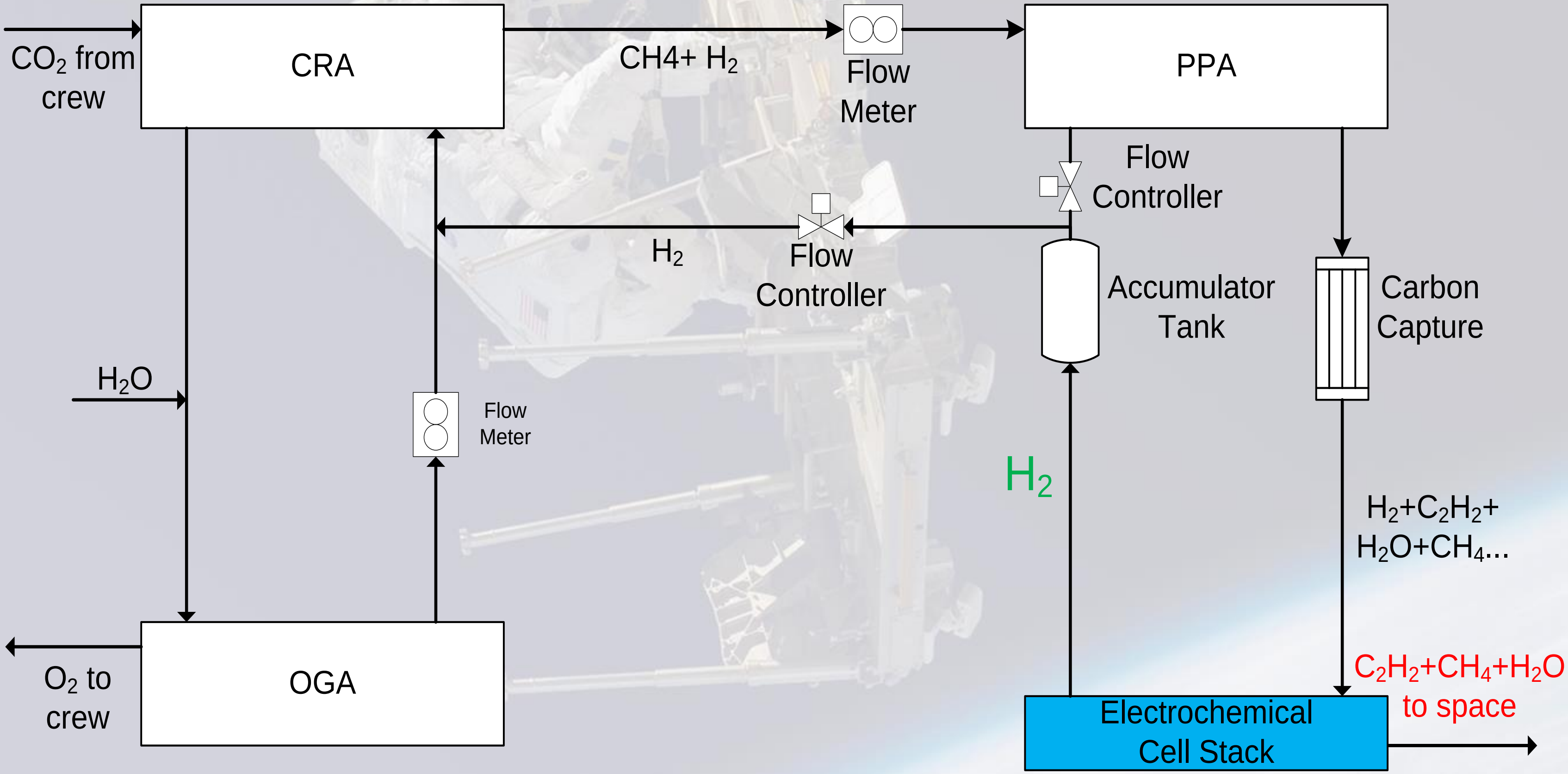


# Effect of water vapor and CO on hydrogenation of $C_2H_2$.

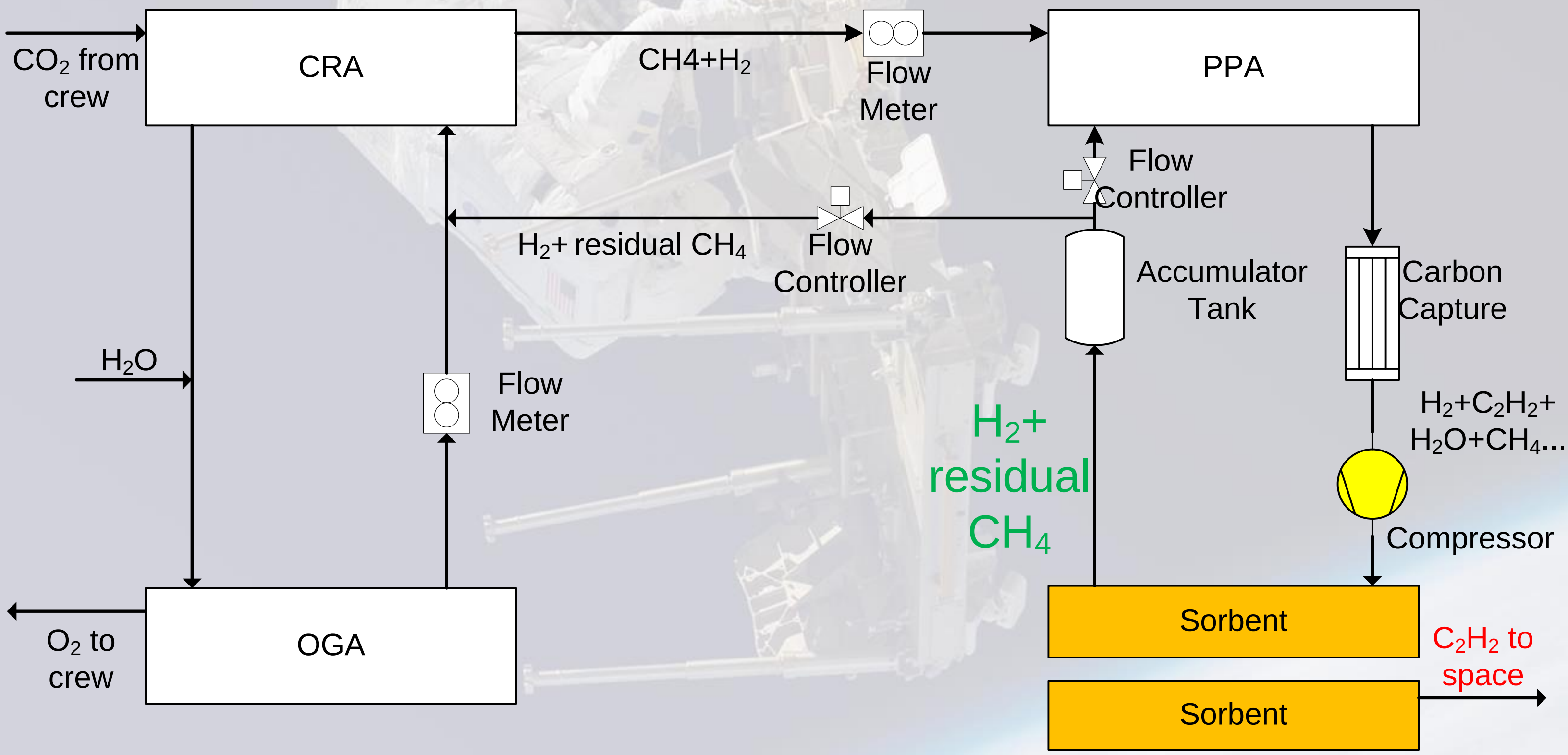




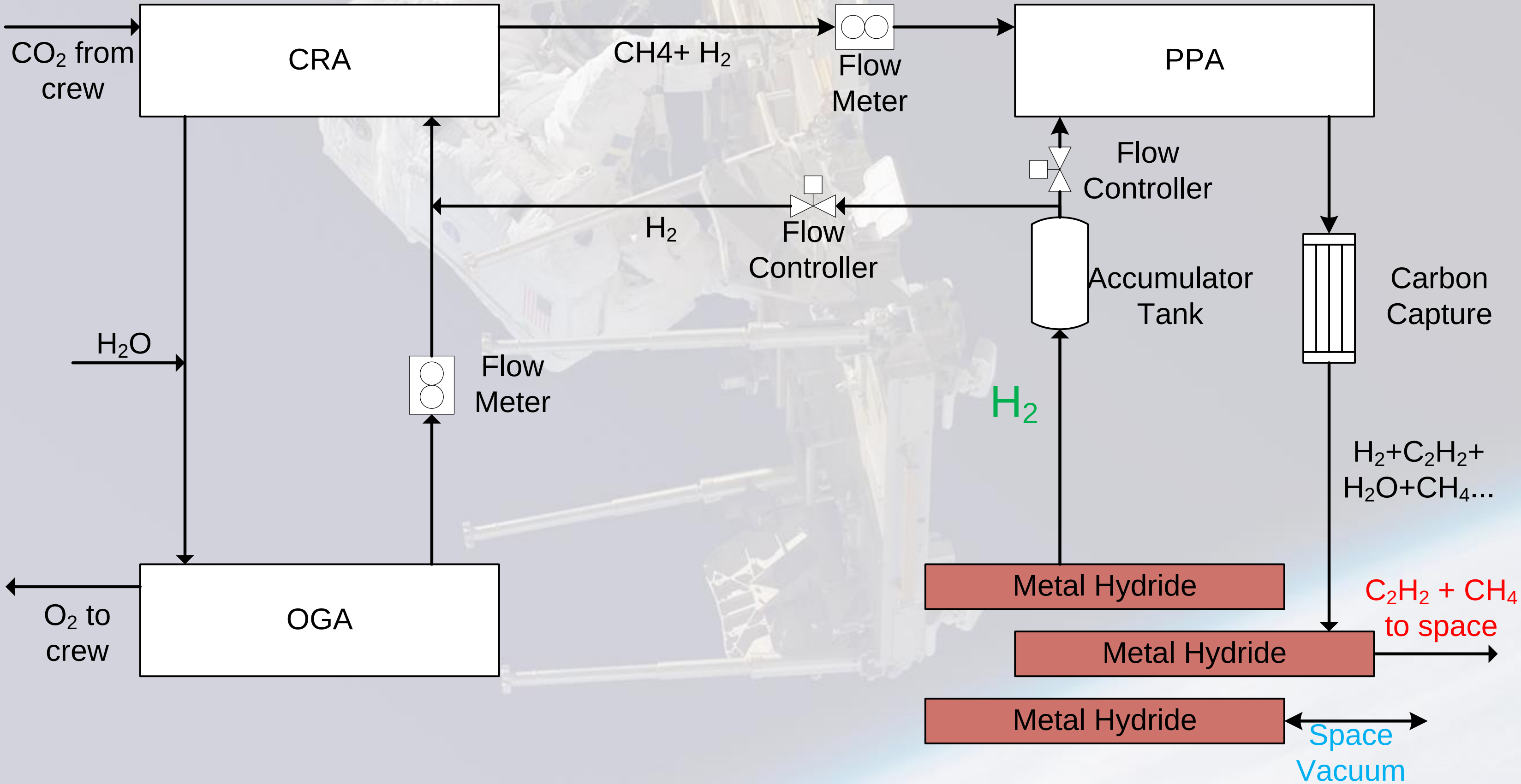
# SI Cell Stack Architecture



# Sorbent Architecture



# Metal Hydride Architecture

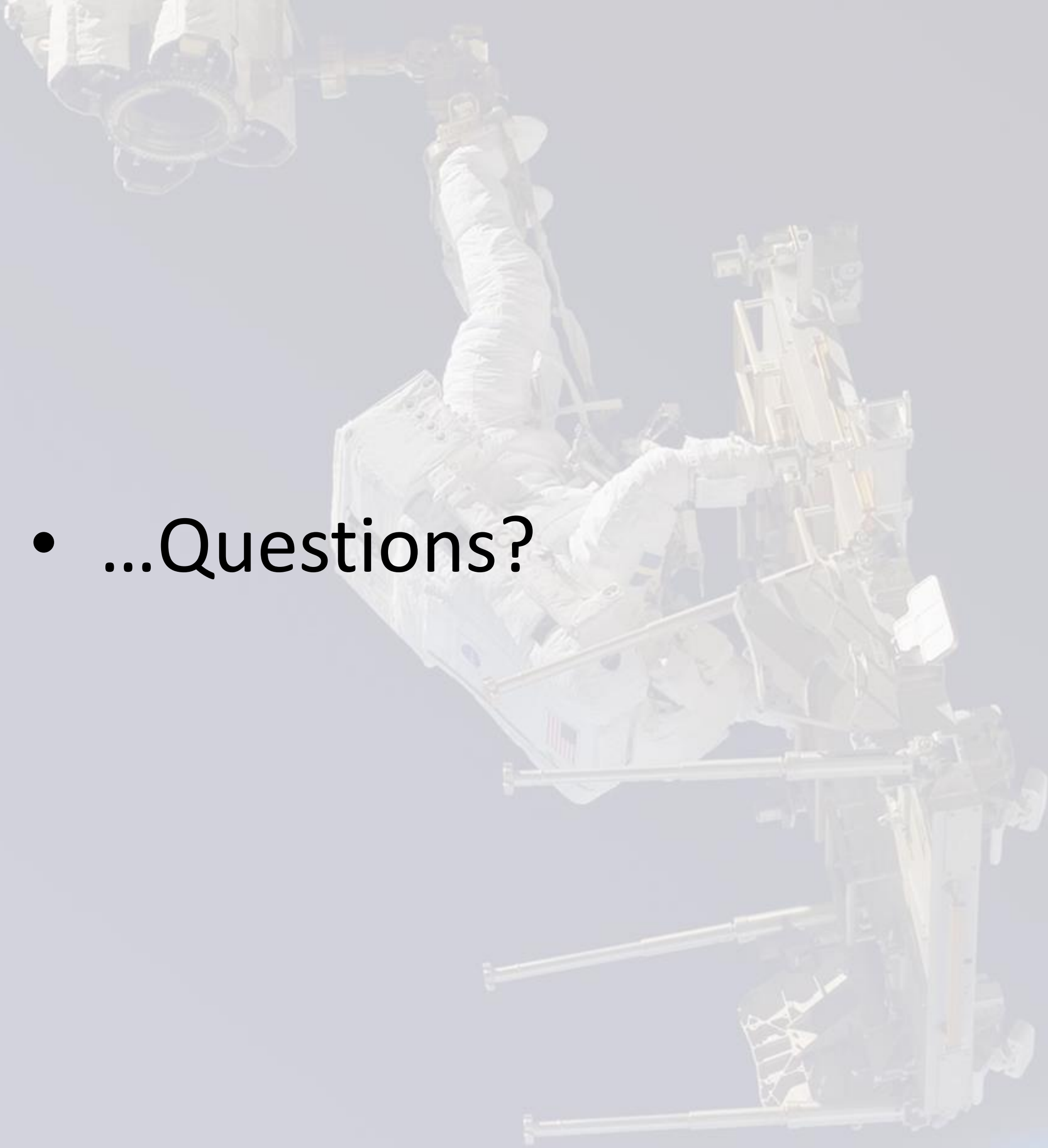


# Conclusion

- Effective acetylene separation technology is essential for Sabatier + PPA architecture
- Future work:
  - Reduce acetylene hydrogenation in cell stacks
  - Test UMPQUA sorbent based hydrogen separation system
  - Test metal hydride

# Acknowledgements

- Kenny Bodkin, Tom Williams, and Jeff Richardson for technical and software support
- Human Exploration and Operations Mission Directorate's Advanced Exploration Systems Program's Life Support Systems Project



- …Questions?