

Hydrogen Purification and Recycling for an Integrated Oxygen Recovery System Architecture

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46rd International Conference on Environmental Systems

Vienna, Austria

July 10-14, 2016

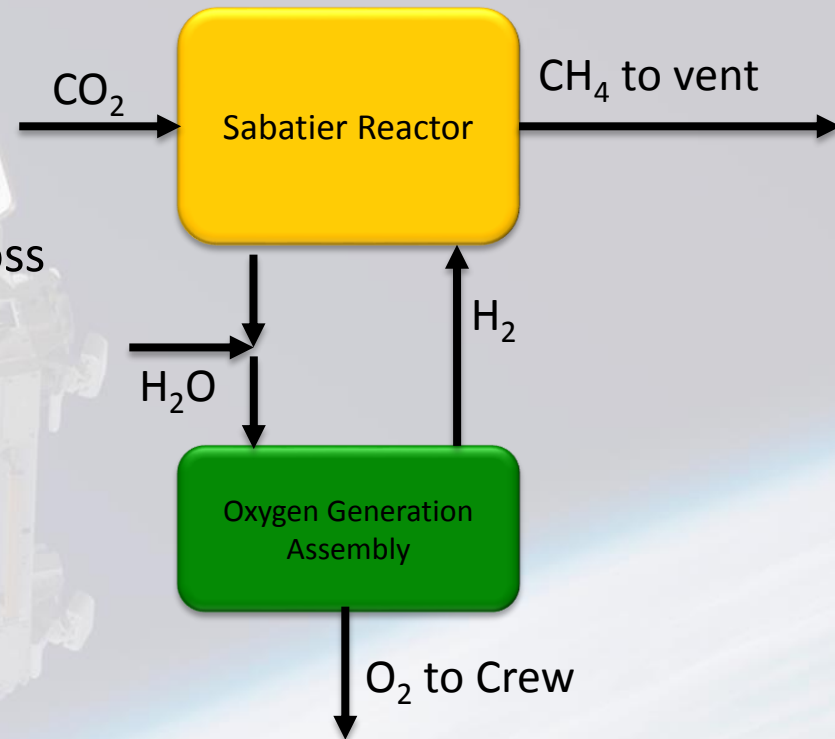
Overview

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

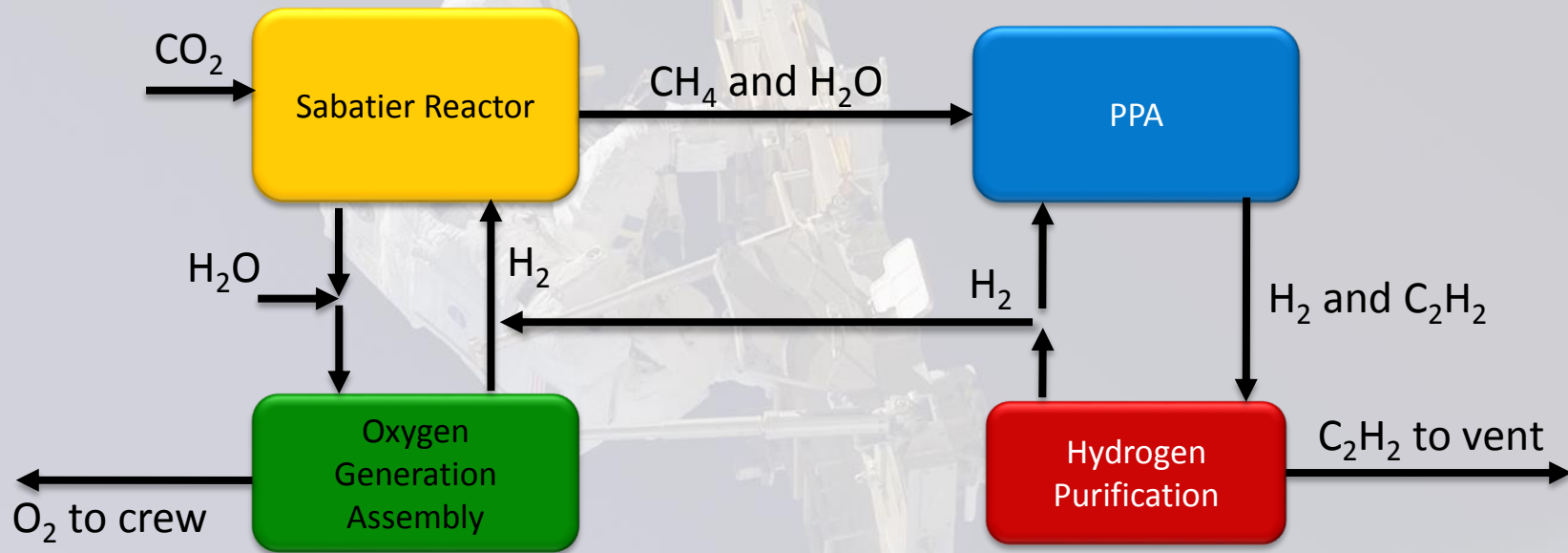
State-of-the-Art

- Sabatier Reactor

- $\text{CO}_2 + 4\text{H}_2 \rightarrow 2\text{H}_2\text{O} + \text{CH}_4$
- 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% O₂ recovery from CO₂ possible

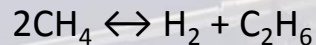
PPA Technology Description

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

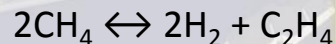
- Targeted PPA Reaction:
$$2\text{CH}_4 \leftrightarrow 3\text{H}_2 + \text{C}_2\text{H}_2$$

- Other reactions:

CH₄ Conversion to Ethane



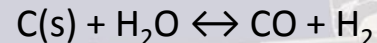
CH₄ Conversion to Ethylene



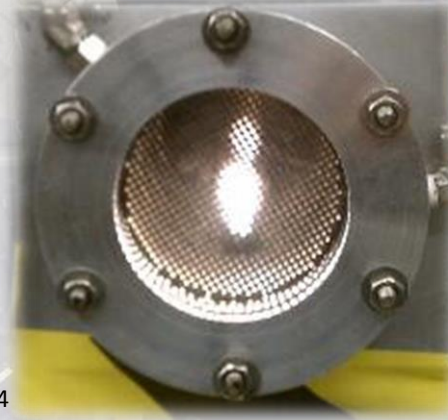
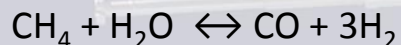
CH₄ Conversion to Solid C



CO Production



CO Production



H₂/CH₄ Plasma



Plasma Pyrolysis
Assembly

Metal Hydride Hardware

- Hydrogen Components, Inc. Metal Hydride Canister
- $\text{LaNi}_{4.6}\text{Sn}_{0.4}$ metal hydride
- Designed for hydrogen storage



Electrochemical Hardware

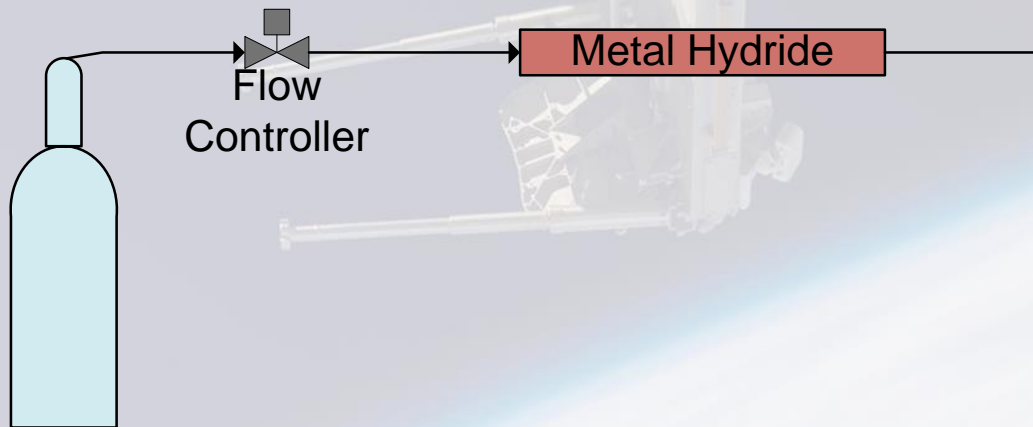
- Electrochemical hydrogen separation
 - H_2 electro-oxidized to protons and electrons
 - Protons are electro-reduced, recombined with electrons, in another chamber producing purified H_2
- Basic technology was well developed but not compatible with CO
 - CO would preferentially adsorb on catalytic electrodes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



Sustainable Innovations
Cell Stack

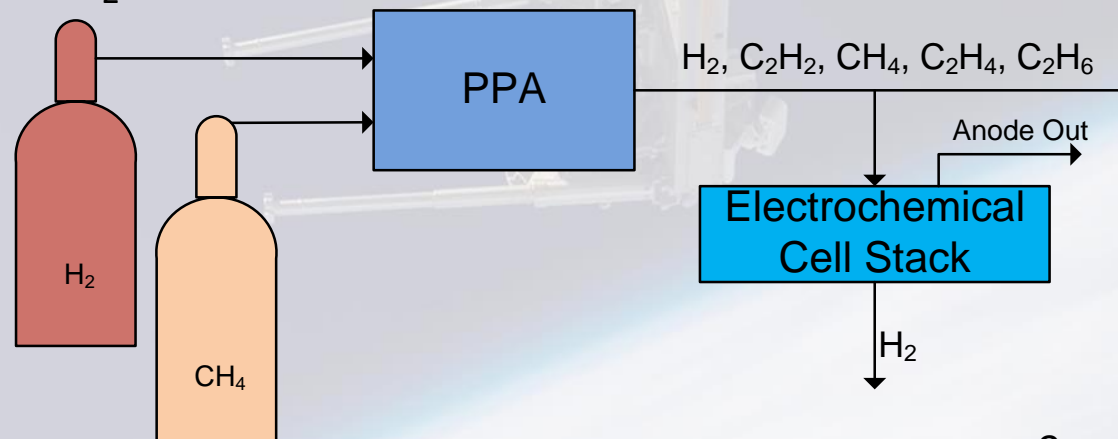
Test Configurations

- 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% C₂H₂, 1% CH₄, and 92% H₂
 - Tested in Marshall Space Flight Center's Component Development Area, usually used for rocket engine component testing



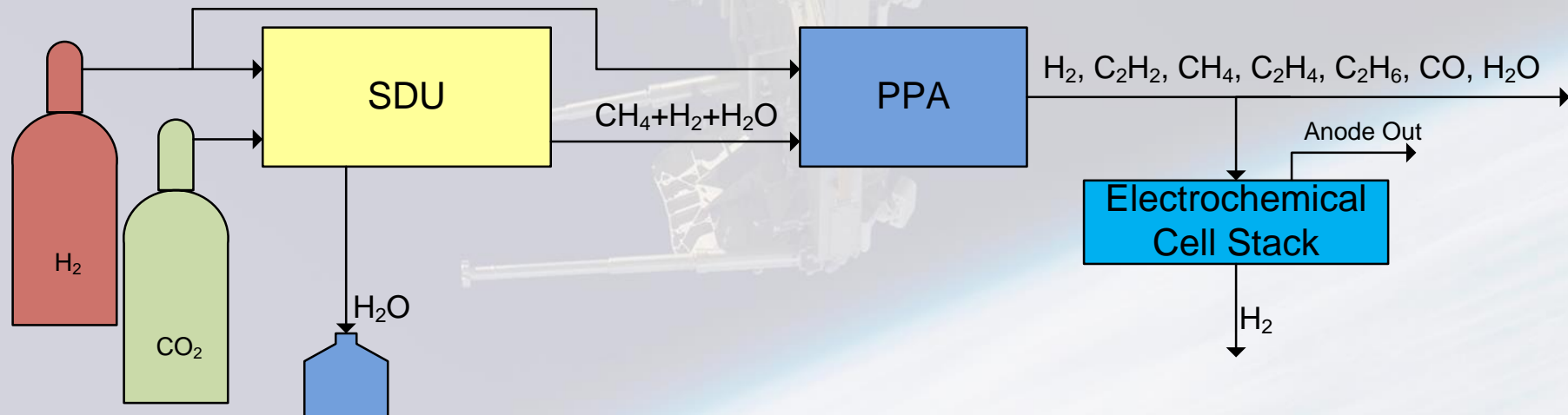
Test Configurations

- **PPA + H₂ Purification**
 - Cell stacks integrated with 2nd Gen. PPA
 - PPA operated with ultra-high purity H₂ and CH₄ bottles
 - 1 Crew Member processing rate
 - 4:1 ratio of H₂:CH₄
 - 52 torr
 - 550 W microwave power
- PPA products contained H₂, C₂H₂, unreacted CH₄, C₂H₄, and C₂H₆
- No CO
- 100 standard milliliters per minute (SmLPM) to cell stack
- Evaluated H₂ product and process effluent



Test Configurations

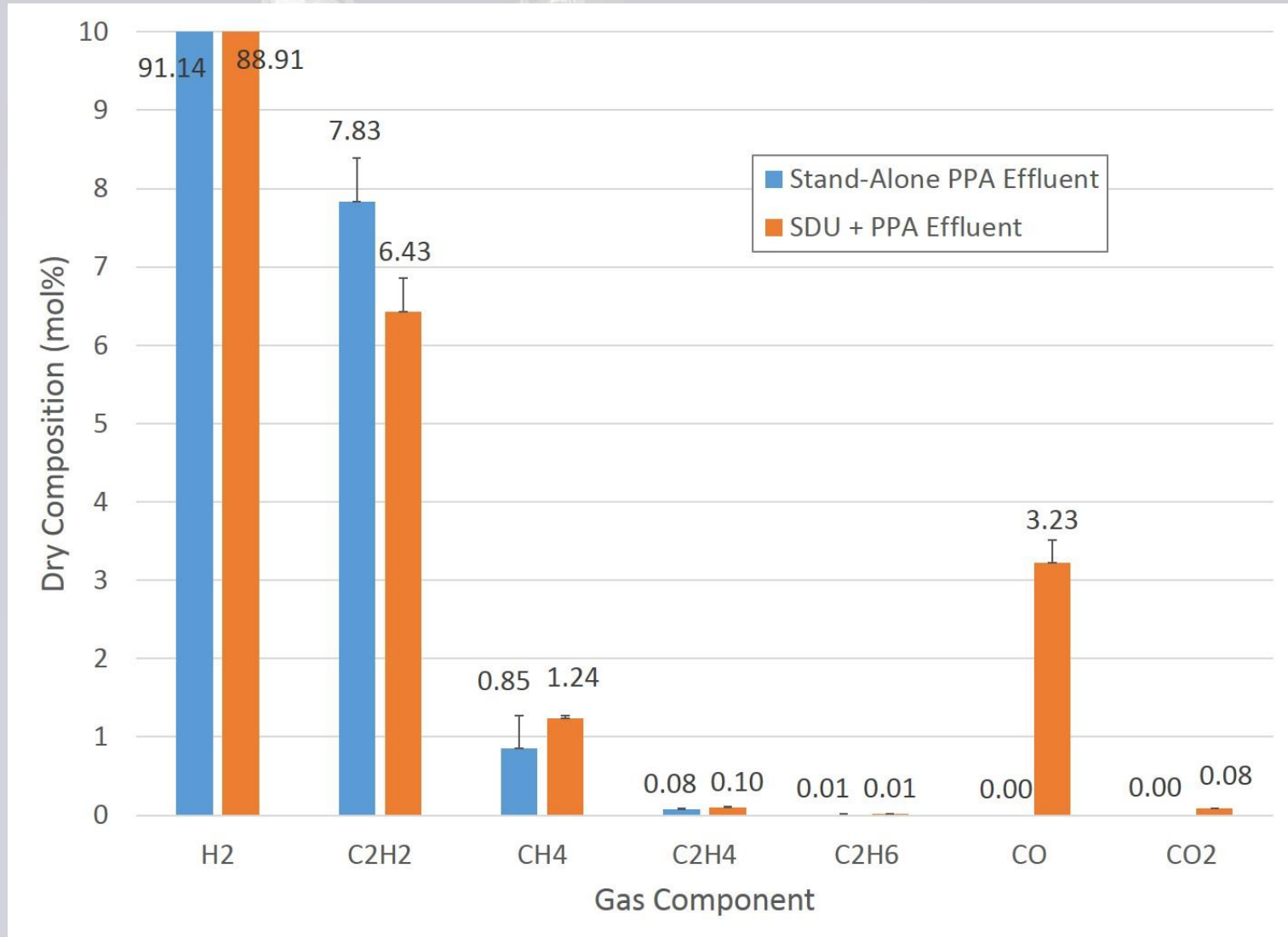
- **Sabatier Development Unit (SDU) + PPA + H₂ Purification**
 - Precision Combustion, Inc. SDU integrated upstream of PPA
 - SDU operated to produce 350 SmLPM CH₄ with no unreacted CO₂
 - Methane product containing 80 mol% hydrogen
 - Water vapor content dew point of 31°C
- PPA operated identically to PPA + H₂ testing
- PPA products contained all previously indicated components and CO and H₂O



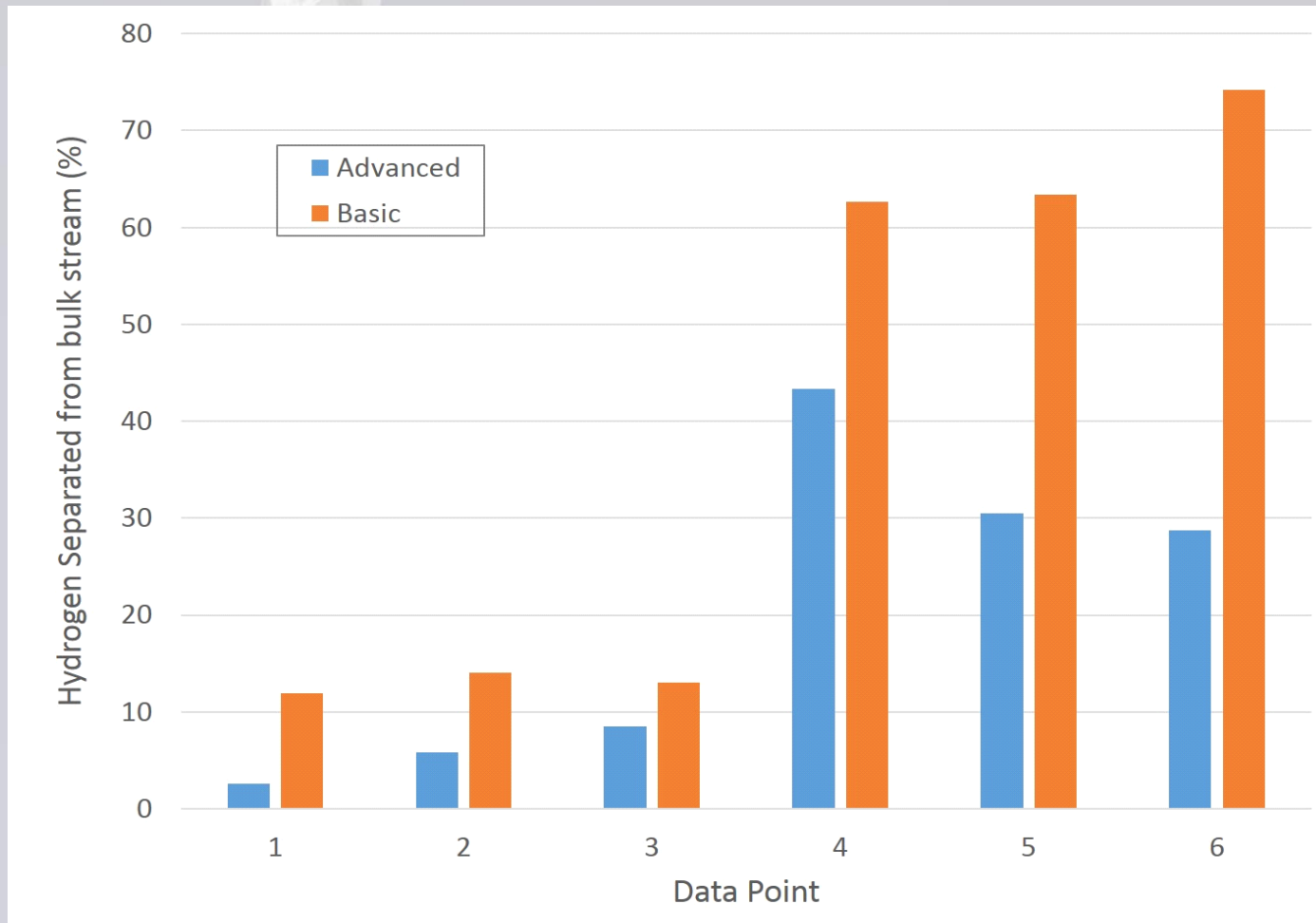
Metal Hydride Performance

- No measurable pressure or temperature difference between pure H₂ runs and acetylene mixed gas runs
- No safety risk under expected operating conditions

PPA effluent composition as a function of configuration



H₂ separation performance comparison between Basic and Advanced cell stacks



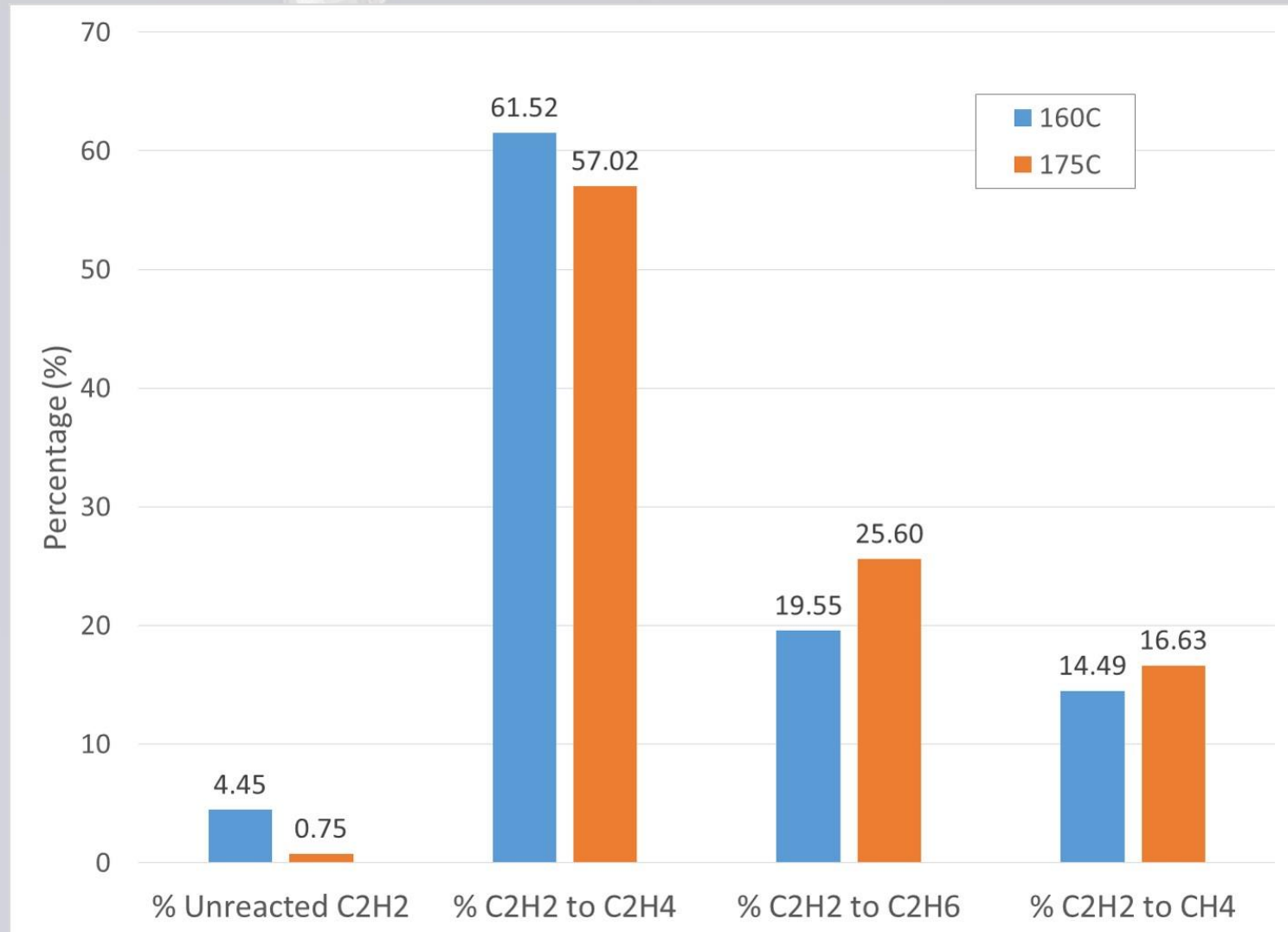
- Varied gas feed from PPA, stack temperature, inlet composition, and applied voltage
 - Conditions for each data point were identical
- **All recovered H₂ pure within measurable limits of μ GC**

Hydrogenation

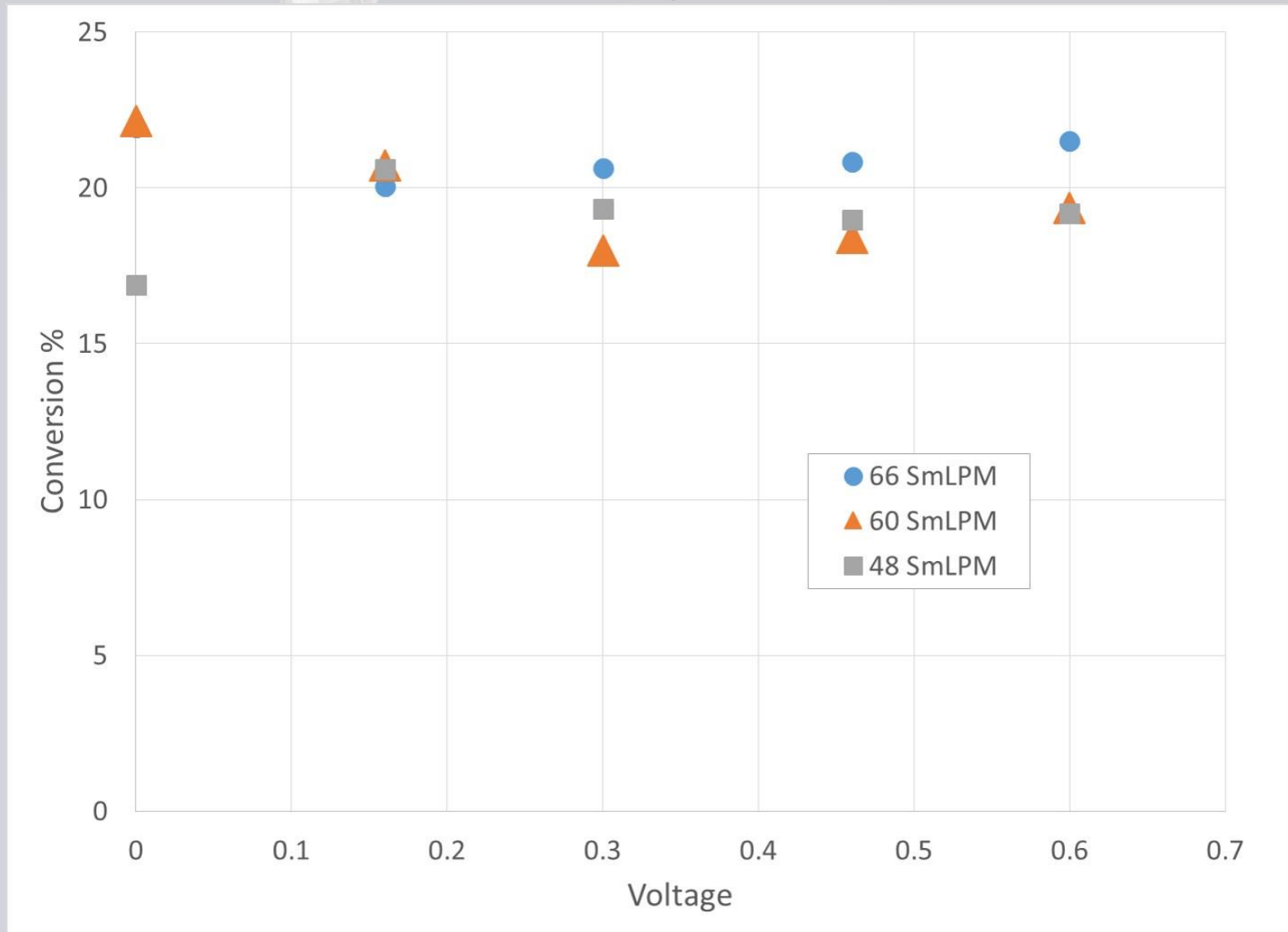
- Expected similar gas mix (minus H₂) leaving anode as entering
- High levels of C₂H₄ and C₂H₆ were observed with minimal or no C₂H₂
- Overall chemical equations:
 - CH₄ Conversion to Ethane $2\text{CH}_4 \leftrightarrow \text{H}_2 + \text{C}_2\text{H}_6$
 - CH₄ Conversion to Ethylene $2\text{CH}_4 \leftrightarrow 2\text{H}_2 + \text{C}_2\text{H}_4$
- Ethane Formation from CH₄ with free radical intermediates:

$$\text{CH}_4 + \text{CH}_4 \leftrightarrow \text{CH}_3^* + \text{CH}_3^* + \text{H}^* + \text{H}^* \leftrightarrow \text{C}_2\text{H}_6 + \text{H}_2$$
 - CH₄ forms CH₃^{*} free radicals which then recombine to form C₂H₆
 - C₂H₆ is converted to C₂H₄ and C₂H₄ is converted to C₂H₂
 - Reverse reactions also occur providing a mechanism for C₂H₂ hydrogenation to the other hydrocarbons

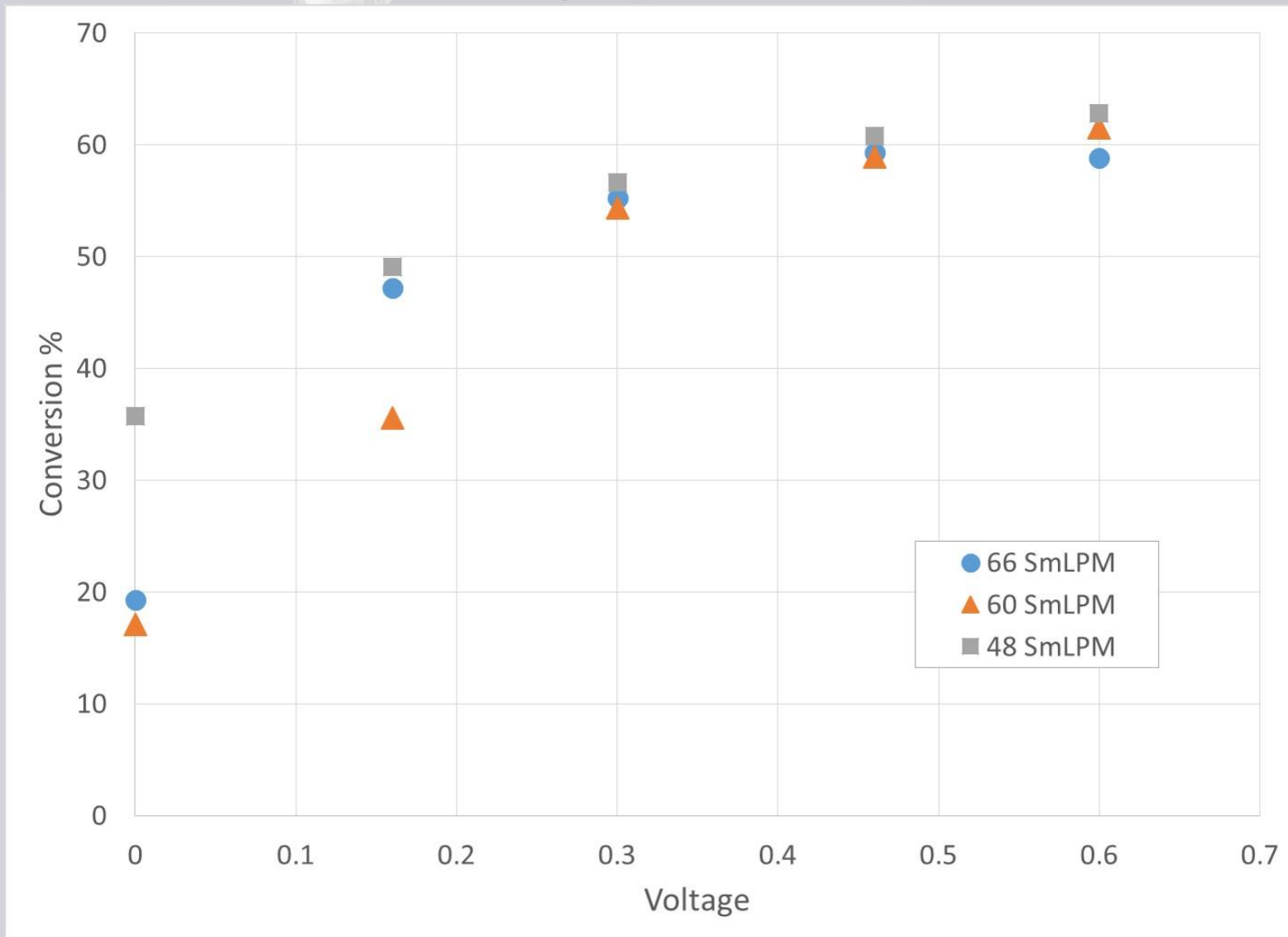
Effect of temperature on C₂H₂ hydrogenation, Advanced Cell Stack



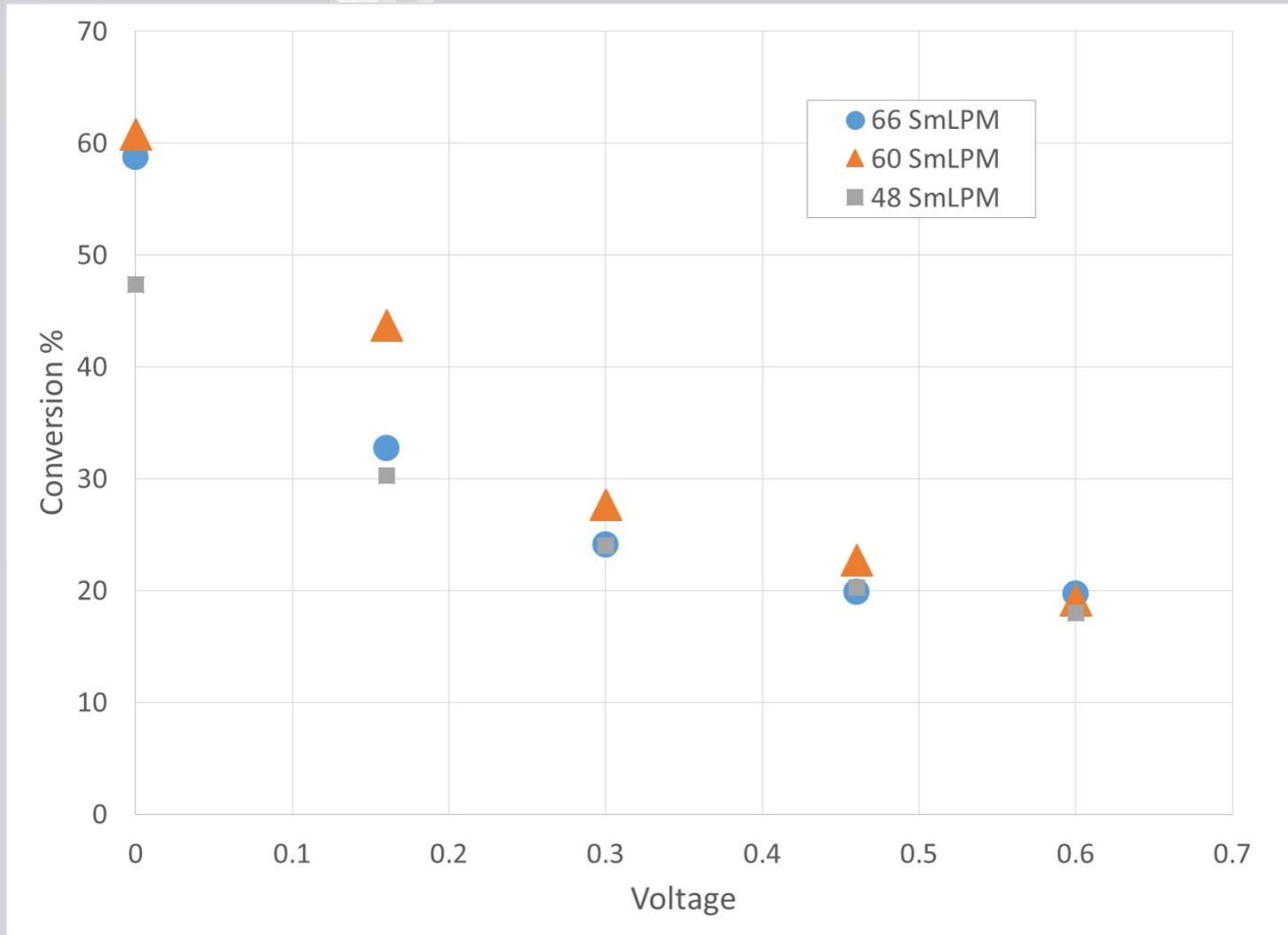
Acetylene conversion to methane in Advanced cell stack as a function of voltage and anode feed rate.



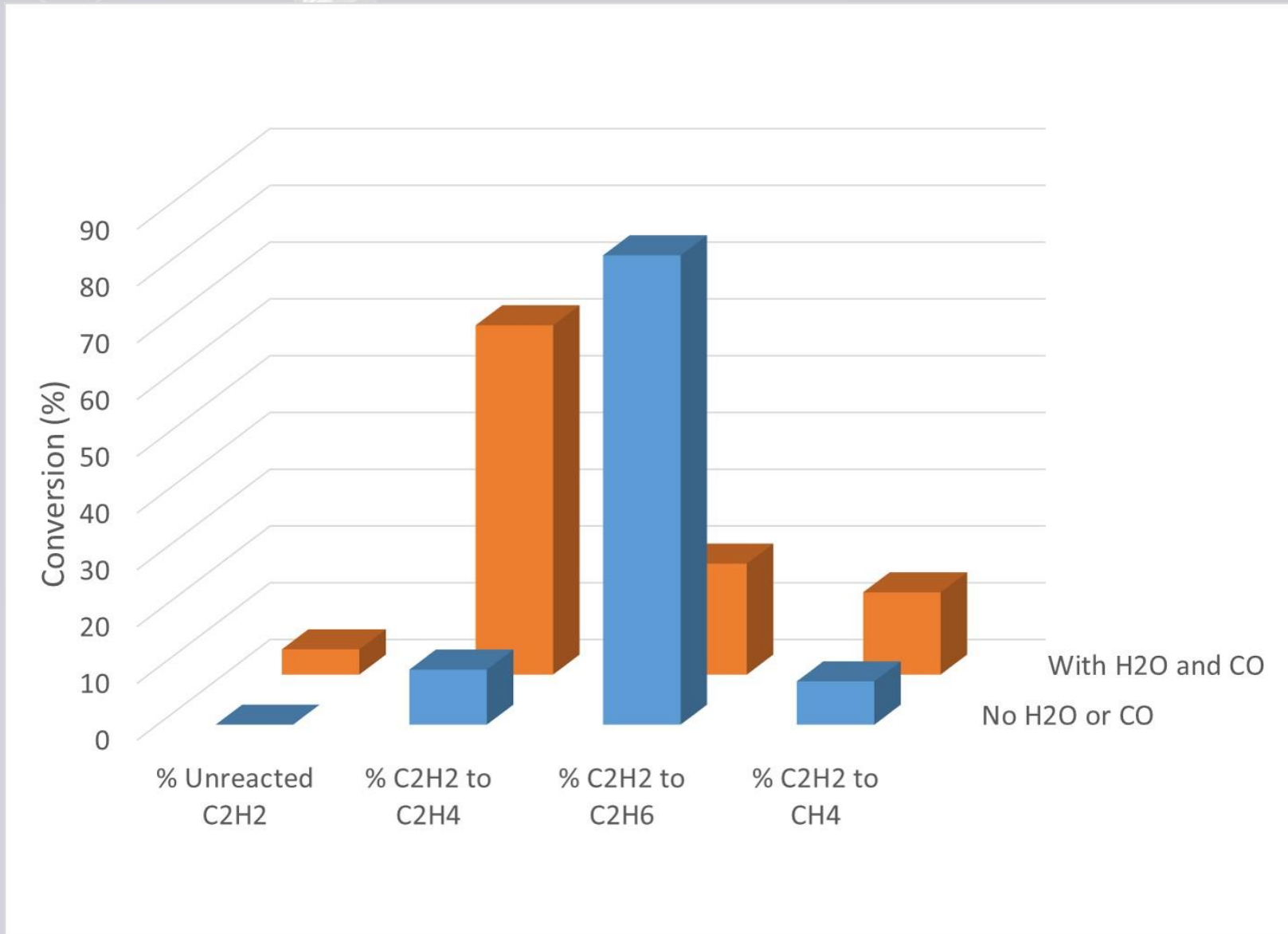
Acetylene conversion to ethylene in Advanced cell stack as a function of voltage and anode feed rate.



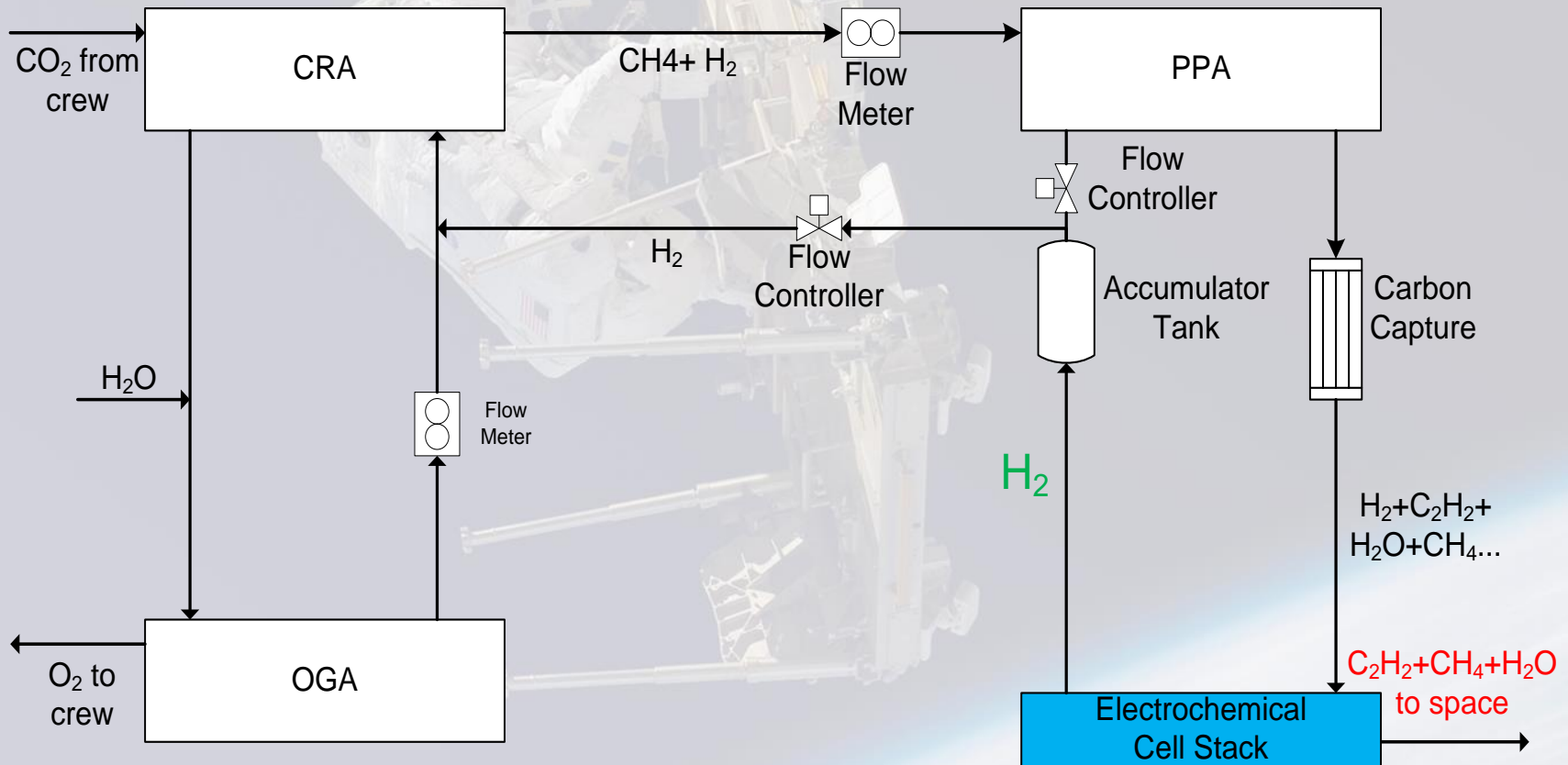
Acetylene conversion to ethane in Advanced cell stack as a function of voltage and anode feed rate.



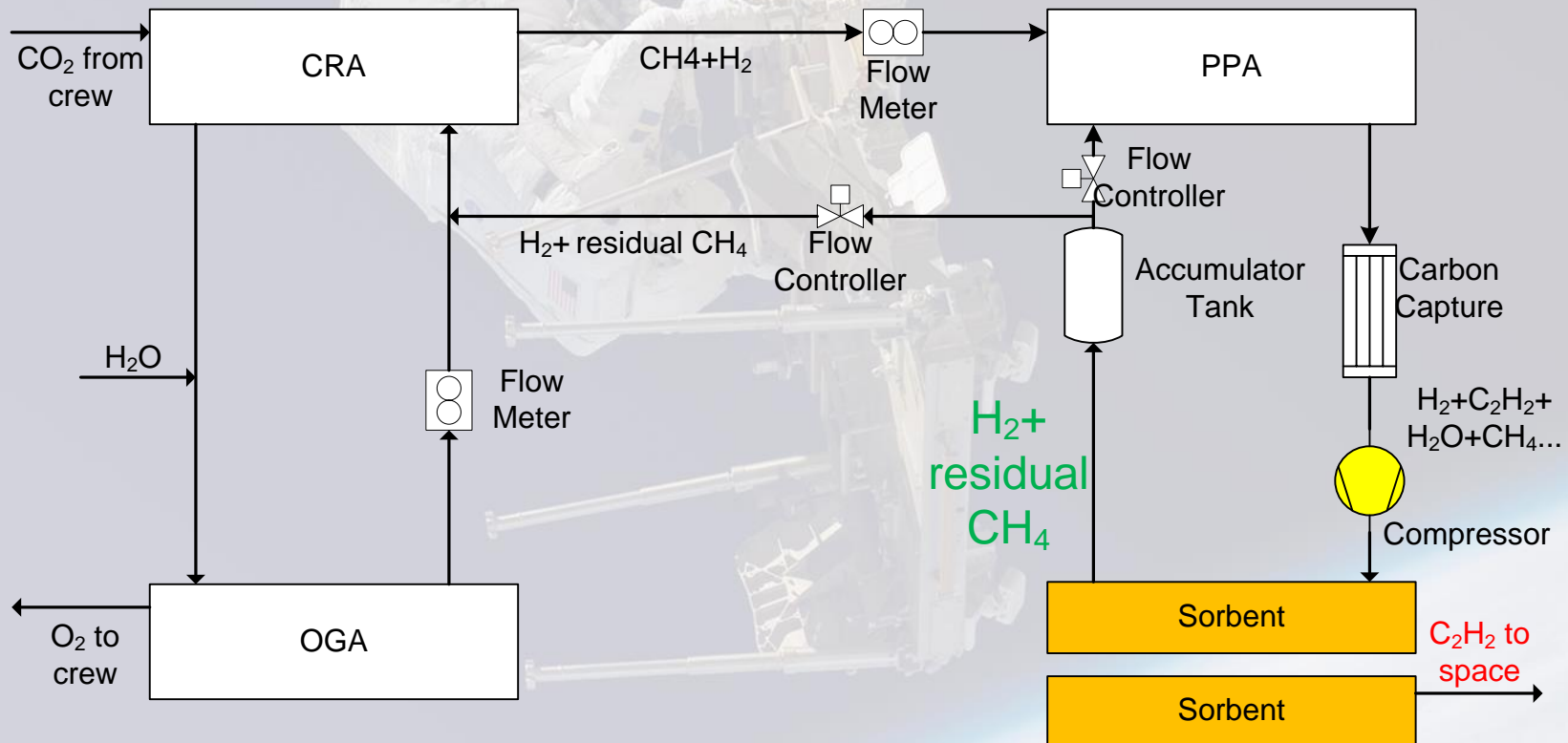
Effect of water vapor and CO on hydrogenation of C_2H_2 .



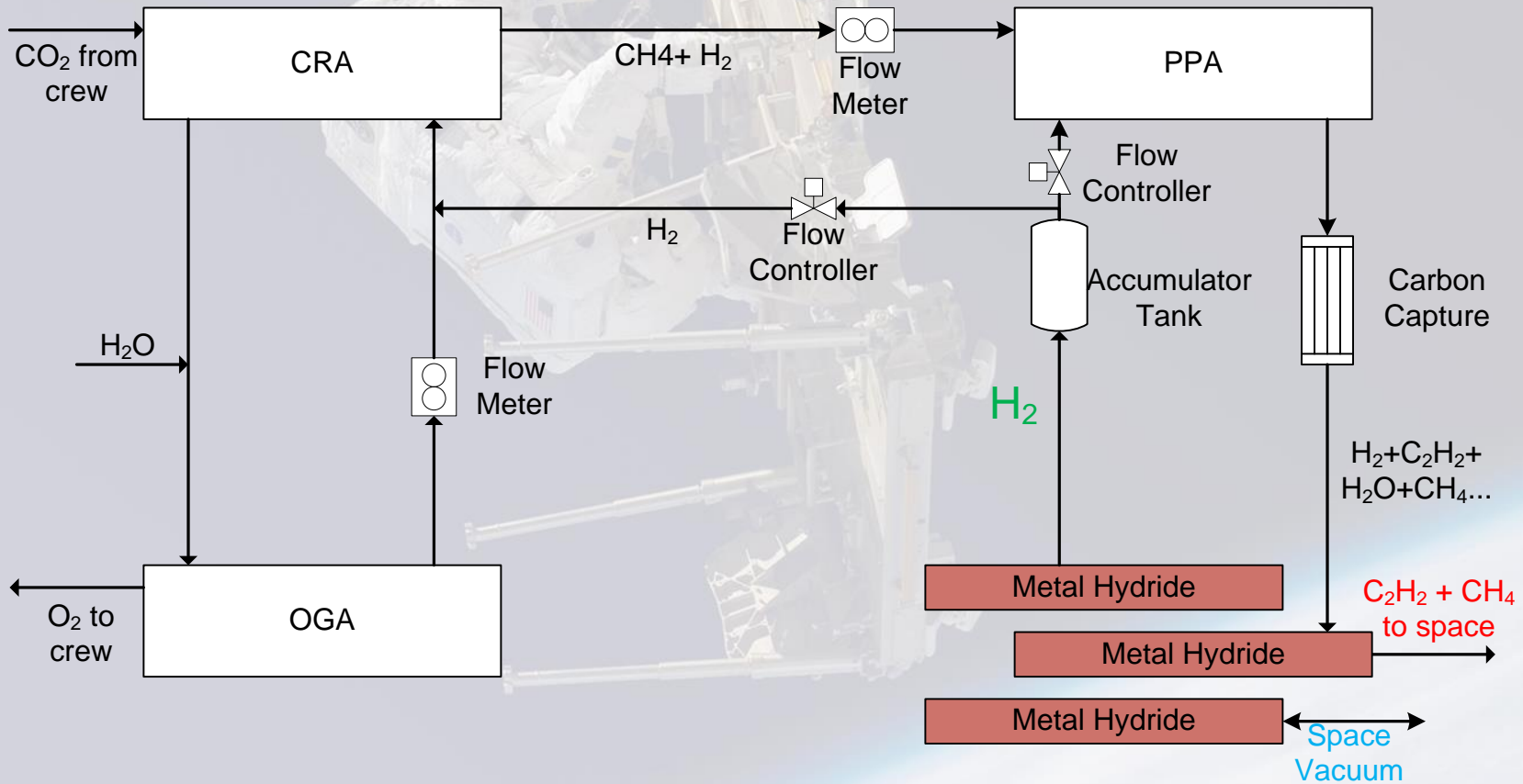
SI Cell Stack Architecture



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?