Steam Methane Reformation Testing for Air-Independent Solid Oxide Fuel Cell Systems

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Fuel Cell Seminar 2015

Recently, NASA has been looking into utilizing landers that can be propelled by LOX-CH_4, to be used for long duration missions. Using landers that utilize such propellants, also provides the opportunity to use solid oxide fuel cells as a power option, especially since they are able to process methane into a reactant through fuel reformation. One type of reformation, called steam methane reformation, is a process to reform methane into a hydrogen-rich product by reacting methane and steam (fuel cell exhaust) over a catalyst. A steam methane reformation system could potentially use the fuel cell's own exhaust to create a reactant stream that is hydrogen-rich, and requires less internal reforming of the incoming methane. Also, steam reformation may hold some advantages over other types of reforming, such as partial oxidation (PROX) reformation. Steam reformation does not require oxygen, while up to 25% can be lost in PROX reformation due to unusable CO2 reformation. NASA's Johnson Space Center has conducted various phases of steam methane reformation testing, as a viable solution for in-space reformation. This has included using two different types of catalysts, developing a custom reformer, and optimizing the test system to find the optimal performance parameters and operating conditions.

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2015 Fuel Cell Seminar

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Fuel Cells at NASA

- Gemini, Apollo, and Space Shuttle used fuel cells as main power source for vehicle and water source for life support and thermal

  PEM (Gemini) and Alkaline (Apollo, Shuttle) fuel cells were used

  Ideal for short (less than 3 weeks) missions when the complete mission load of O_2 and H_2 can be launched with the vehicle

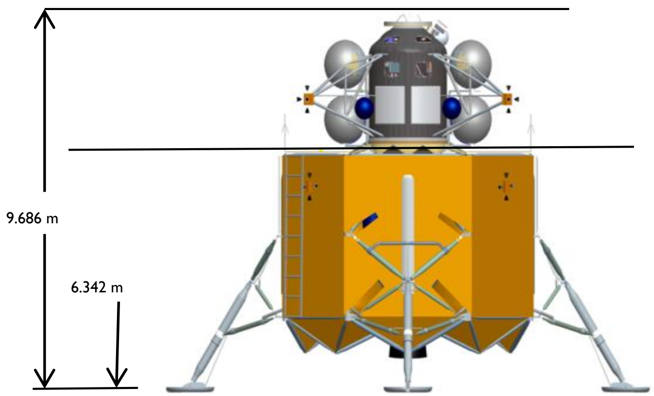
- New missions that might require long-duration stays in orbit or at a habitat, cannot rely on the availability of *pure* reactants and should aim to be sun-independent – a problem for which Solid Oxide Fuel Cells might be the answer

LOX/LH2 vs. LOX/CH_4 Usage

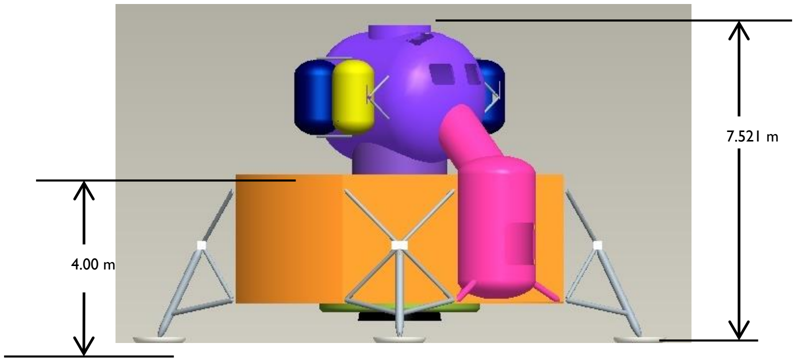
- Recently, NASA has investigated & developed LOX/CH4-propelled landers (e.g. Morpheus). In order to preserve mission flexibility, fuel cells should be studied as a potential power source.

- Previous work at JSC has identified the volumetric and mass benefits of LOX/CH_4 propelled vehicles vs LH_2/LO_2

- The availability of LOx/CH_4 introduces solid oxide fuel cells (SOFCs) as an option, due to their ability to efficiently utilize those reactants.



LH2/LO2 Lander Size



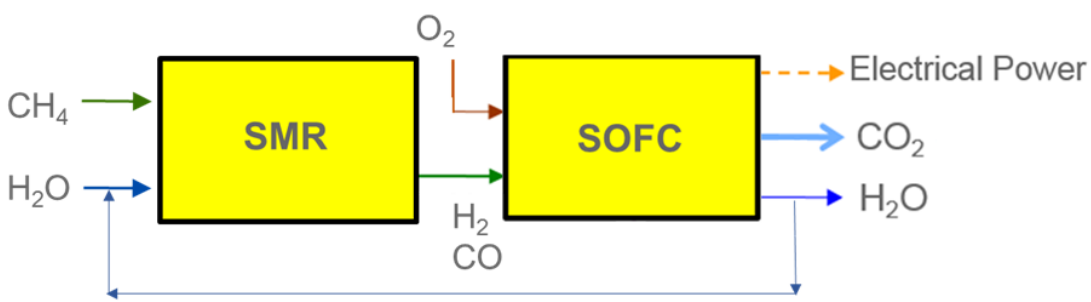
LOX/Methane Lander Size

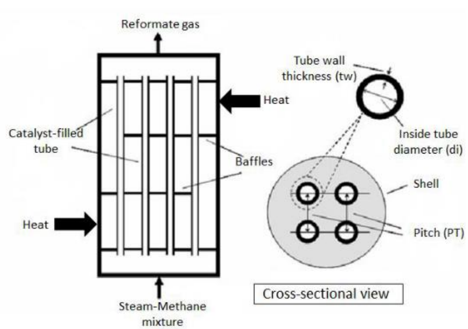
Steam Reforming Introduction

- SOFCs allow internal reforming to convert CH_4 into H_2 for fuel
  - Some external reforming of the fuel stream is optimal

- Utilizing an external steam methane reformer (SMR) would be the first step to creating a more efficient SOFC system

Predicted typical output gas concentrations:
~48% H2
~27% CH4
~22% CO
~3% CO2





SMR Primary Chemical Reactions

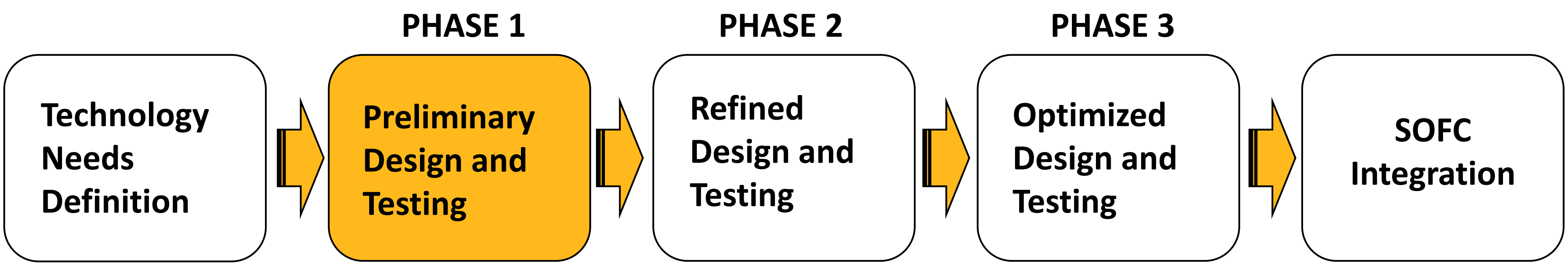
FUEL FOR SOFC

Oxygenolysis	$CH_4 + H_2O \leftrightarrow CO + 3H_2$	$\Delta H^o_{rxn} = 206\ kJ/mol$
Water-gas shift	$CO + H_2O \leftrightarrow CO_2 + H_2$	$\Delta H^o_{rxn} = -41\ kJ/mol$

SMR – Phase 1

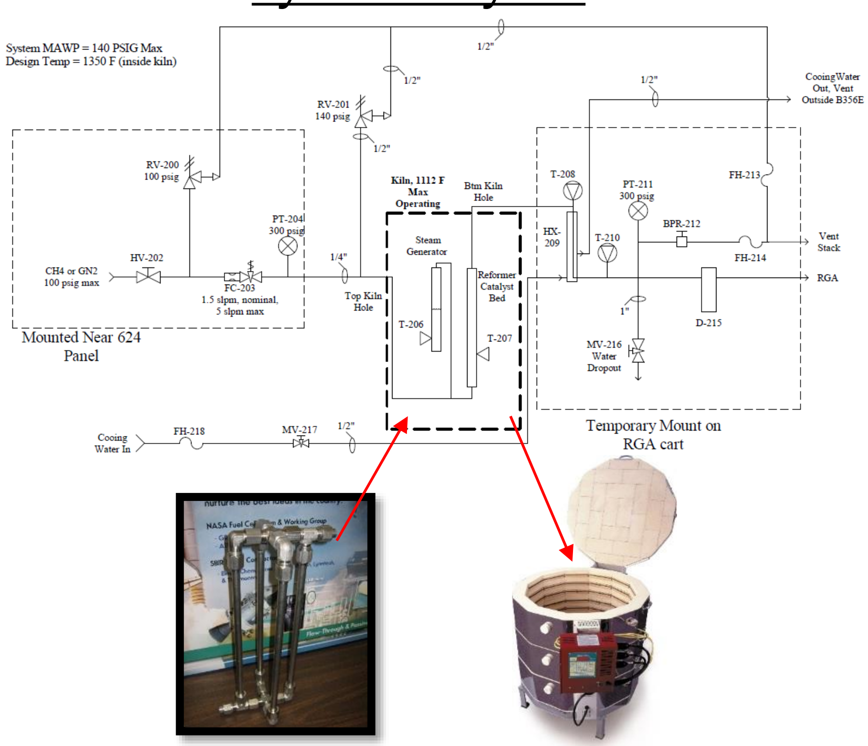
Objective:

- Determine optimal operating conditions, for 4 cylinder SMR reactor system design
  - Based on theoretical Matlab model



SMR – Phase 1

System Layout



Catalyst

Nickel Oxide Catalyst

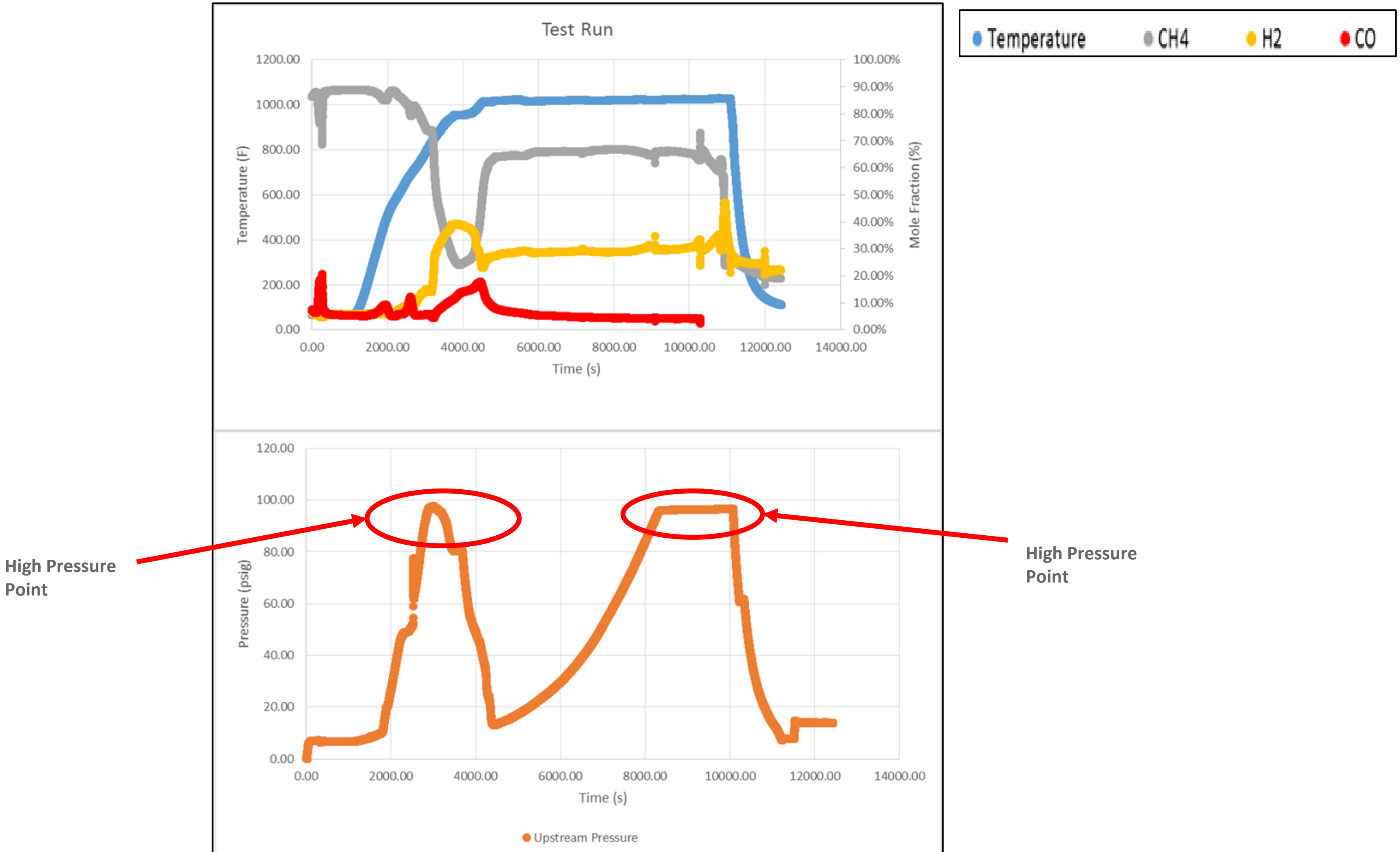
Reasons for selection:

- Low cost
- Reasonably good efficiency

Hot Flow Test (With Preheating of Gases)					
Test #	CH_4 Mass Flow Rate [g/min]	H_2O Mass Flow Rate [g/min]	Steam to Methane Ratio [mol/mol]	SMR Temperature [˚F]	System Pressure [psia]
1	0.982	1.2	1	1020	14.7
2	0.982	2.3	2	1020	14.7
3	0.982	3.3	3	1020	14.7

SMR – Phase 1

Generated H_2 production, and also high build-up of upstream pressure



SMR – Phase 1

Test results:
- Initial production of Hydrogen
- Carbon deposition blockage in the catalyst bed

Conclusions:
- Use of a kiln to produce steam
  - No steam flow regulation (steam-to-methane ratio)
- Catalyst was not reduced:

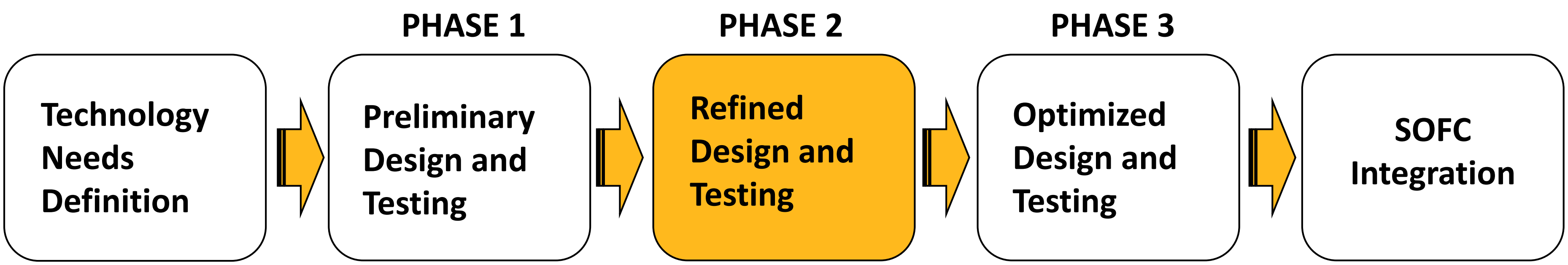
$$NiO + H_2 \leftrightarrow Ni + H_2O$$

Phase 2 recommendations:
- Use syringe pumps to regulate water flow
- Use a separate heater to generate steam
- Reduce catalyst

SMR – Phase 2

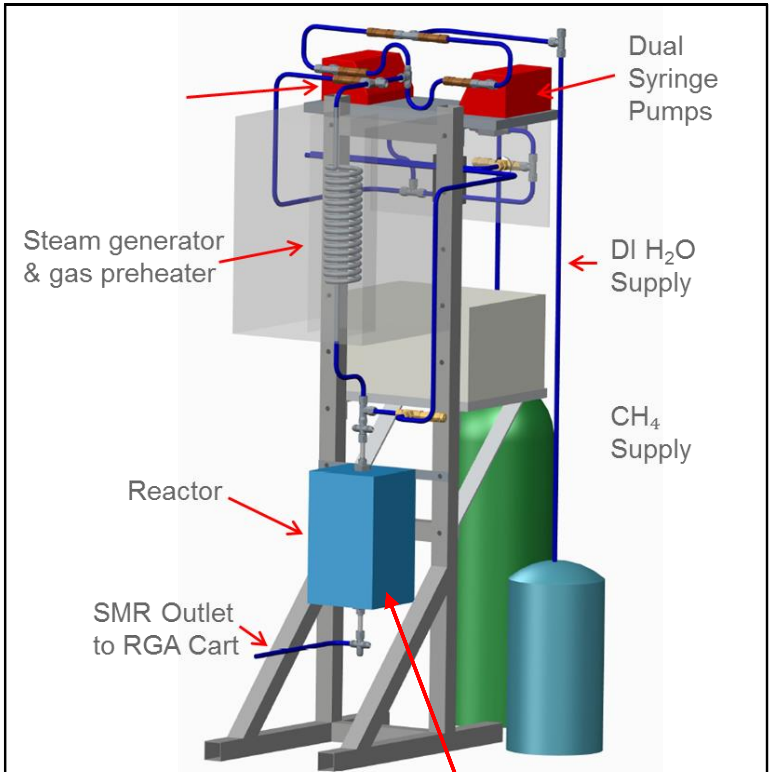
Objective:

- Determine optimal operating conditions, for 1 cylinder SMR reactor system design
  - Based on updated theoretical Matlab model
- Incorporate recommendations from Phase 1 test results



SMR – Phase 2

System Layout



Improvements

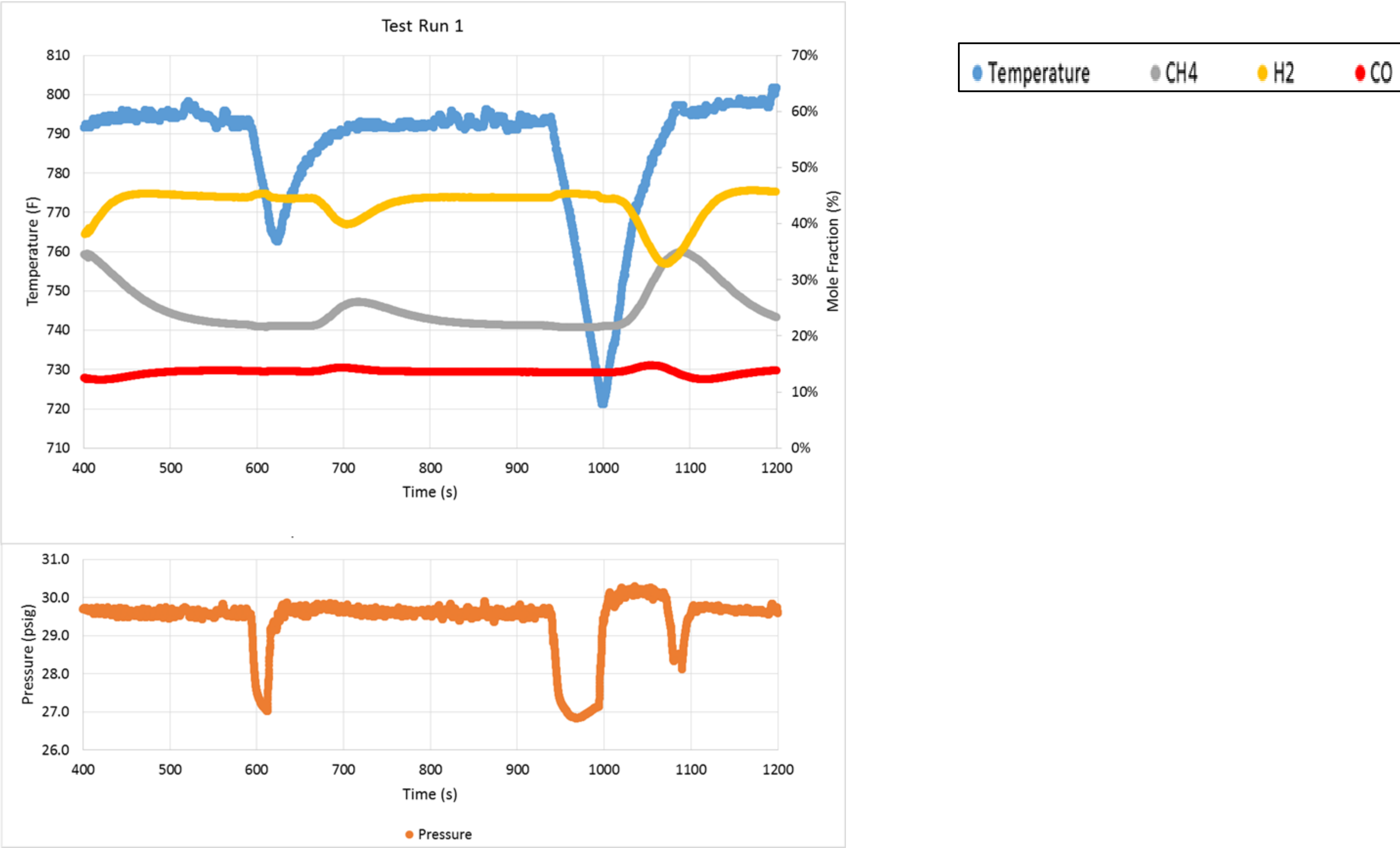
Before	After
Passive Water Control • Tubes filled with H_2O and heated; no control over amount of steam generation and delivery	Active water control • Syringe pumps deliver specific flow rate of H_2O to steam generator
Steam generator & reformer placed in kiln	Separate tube furnaces for steam generator and reformer • Better heating control
Catalyst was not reduced	Catalyst reduced by hydrogen, before test runs
Four tube reactor design	One tube reactor design

Hot Flow Test (With Preheating of Gases)

Test #	CH_4 Mass Flow Rate [g/min]	H_2O Mass Flow Rate [g/min]	Steam to Methane Ratio [mol/mol]	SMR Temperature [˚F]	System Pressure [psia]
1	1.637	5.516	3	930	14.7
2	1.637	6.435	3.5	930	14.7
3	1.637	7.354	4	930	14.7

SMR – Phase 2

Observed temperature and pressure transients – Affected H_2 production at certain points



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SMR – Phase 2

Test Results:

- Higher overall amount of hydrogen being produced
- Carbon deposition still being generated, though at lower rate

Carbon deposition in test system

Conclusions:

- Catalyst was not sufficiently reduced
- Need to increase thermal mass to heat up fluids to design temperatures
- Need to minimize hotspots that promote carbon deposition
  - Essential to maintain consistent heating profile for input stream going into the reactor

Recommendations:

- Conduct one more round of testing, with system modifications

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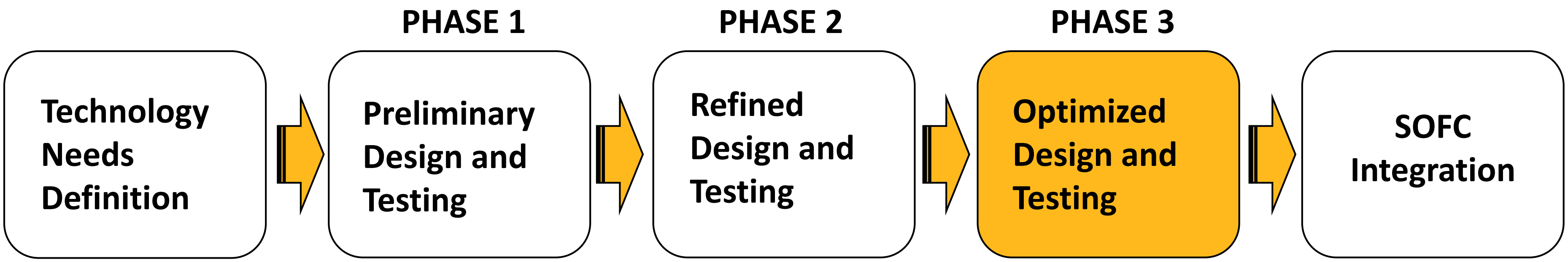
SMR – Phase 3

Objective:

- Determine optimal operating conditions, for 1 cylinder SMR reactor design
  - Based on updated theoretical Matlab model
- Incorporate recommendations from Phase 2 test results



SMR – Phase 3

System modifications:

Switched to new metal foam catalyst – for increased chemical stability and potential conversion efficiency

Heat Transfer	Mass Transfer
Higher thermal conductivity minimizes temperature gradients & hot spots	Porous structure provides more tortuous path for gas molecules
Helps favor the reactions we want and prevent those we don't	Better dispersion of the active metals coated on the metal foam structure

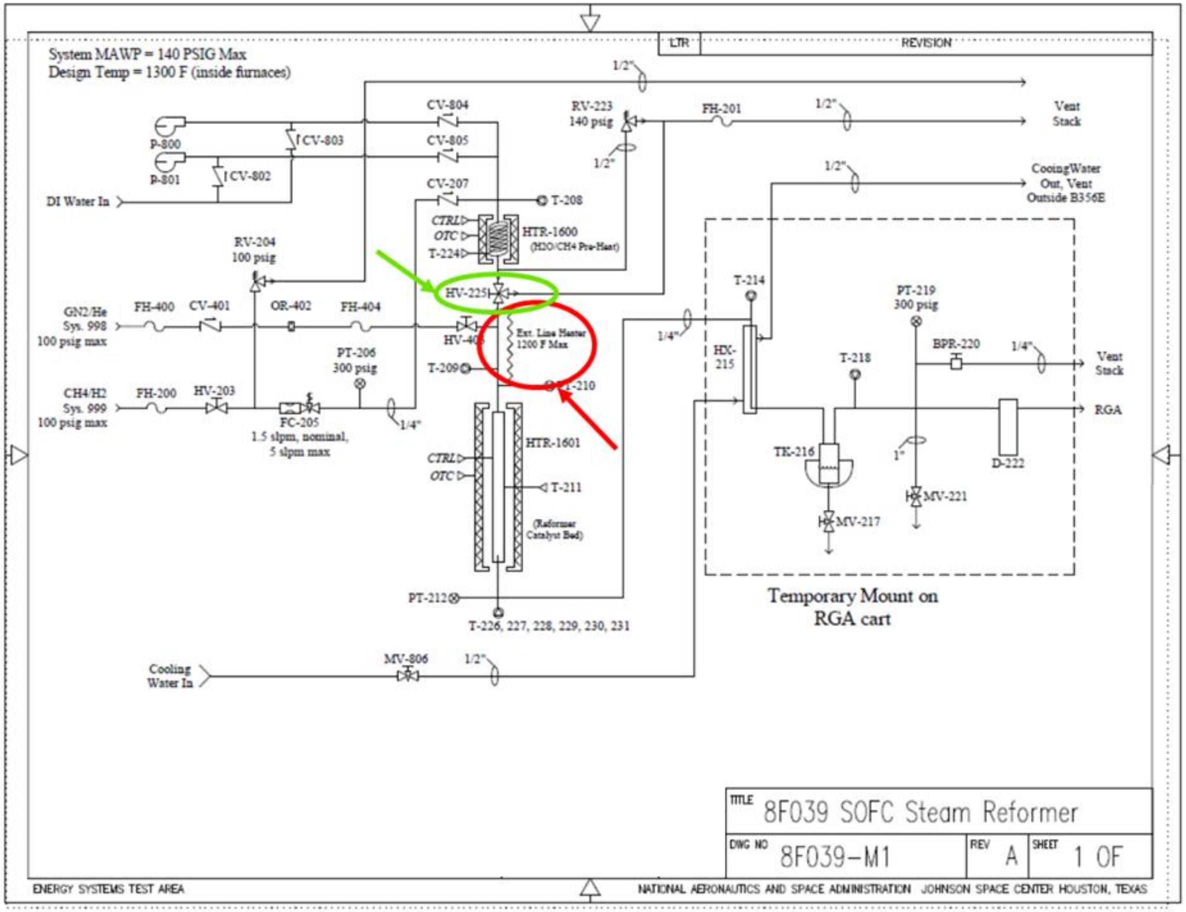
Ordered Pd/Rh coated SIC metal foam catalyst, machined to SMR physical dimensions with through-hole for high temp temperature probe

Hot Flow Test (With Preheating of Gases)					
Test #	CH_4 Mass Flow Rate [g/min]	H_2O Mass Flow Rate [g/min]	Steam to Methane Ratio [mol/mol]	SMR Temperature [˚F]	System Pressure [psia]
1	1.637	7.354	4	930	14.7
2	1.637	7.354	4	1020	14.7
3	1.637	7.354	4	1110	14.7
4	3.274	14.708	4	930	14.7
5	3.274	14.708	4	1020	14.7
6	3.274	14.708	4	1110	14.7

SMR – Phase 3

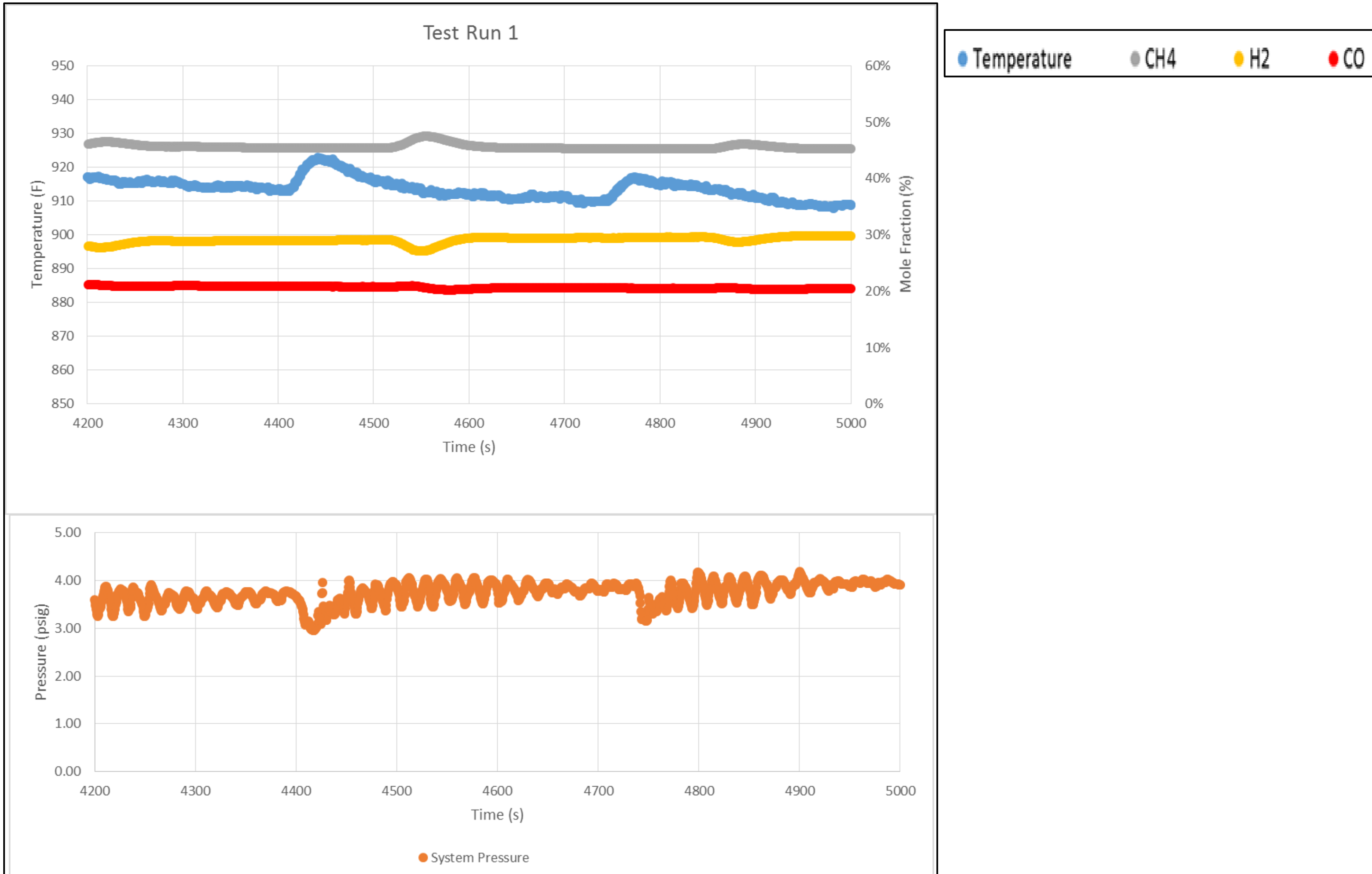
System modifications:

- Installed 3-way valve downstream of steam generator, to help avoid T and P transients (circled in green)
- Installed heating tape between steam generator and reactor, to maintain consistent heating profile (circled in red)



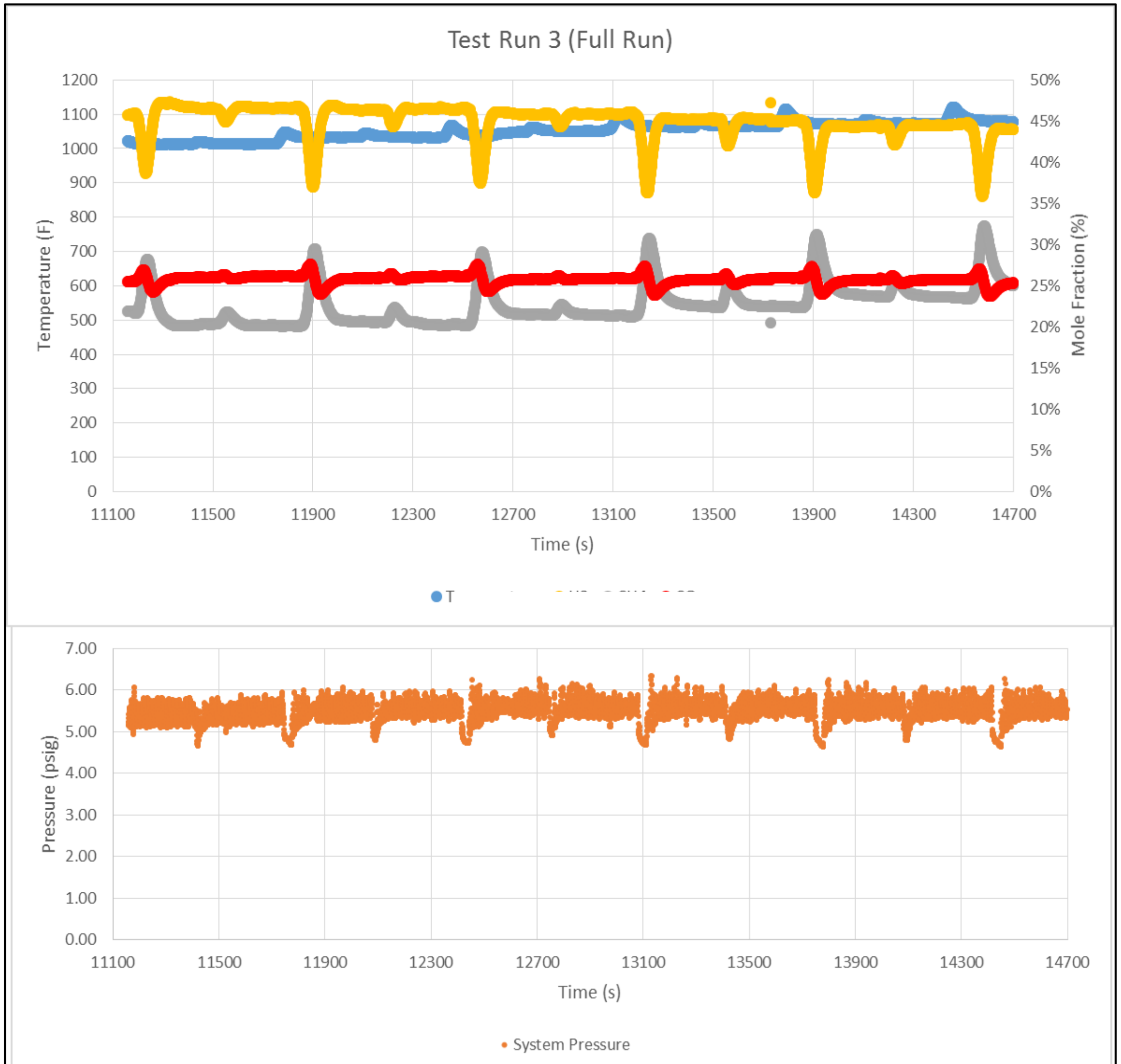
SMR - Phase 3

Initial test run – steadier temperature and pressure rates



SMR - Phase 3

Final test run – steady pressure flow, fluctuating H_2 production



Temperature CH4 H2 CO

SMR - Phase 3 & Test Summary

Testing Results:

- Test prematurely ended due to leaking relief valve
- Eliminated carbon deposition
- Similar H_2 conversion rates to Phase 2

Conclusions/Lessons Learned:

- Ensure relief valve has captured venting
- When possible, run higher flow rates through new catalyst to determine whether conversion efficiency increases
- Potentially better to use Coriolis flow controller instead of syringe pumps

SMR Overall Test Summary

	Phase 1	Phase 2	Phase 3
Reformer Design/Catalyst	4 cylinder reactor/Nickel Oxide Pellets	1 cylinder reactor/Nickel Oxide Pellets	1 cylinder reactor/Pd-Rh on SIC metal foam
Test Results	-Lower than expected production of H2 (30%-35%) -Carbon deposition blockage in the catalyst bed	-Higher H2 production (~45%) -Carbon deposition generated at lower rate	-Similar H_2 conversion rates to Phase 2 (~45%) -Eliminated carbon deposition -Test ended due to leaking relief valve
Conclusions	-Improper steam-to-methane ratio -No method of steam flow regulation -Catalyst was not reduced, as it should've been	-Catalyst was not sufficiently reduced -Needed to increase thermal mass to heat up fluids to design temperatures -Need to minimize hotspots	-Ensure relief valve has captured venting -Run higher test flow rates through new catalyst to determine whether conversion efficiency increases -Maybe use Coriolis flow controller

SMR – Future Plans

Future System Integration Path

Planned Activities	Time Period
Integrated test with SOFC	FY'16
Integrated test with SOFC, In-Situ Resource Utilization (ISRU), and LOX/CH_4 Cryogenic Fluid Management (CFM) -Feed CH_4 from boiloff	FY'16

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