Microscale Enhancement of Heat and Mass Transfer for Hydrogen Energy Storage

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June 9th 2015

Hydrogen Storage Engineering

CENTER OF EXCELLENCE

ST 046

ONAMI

OREGON NANOSCIENCE AND MICROTECHNOLOGIES INSTITUTE

μicroproducts Breakthrough Institute

Overview

Timeline

- Project Start Date: 2/1/09
- Project End Date: 6/30/15
- 82% Complete

Barriers

- Barriers addressed
  - **A**) System Weight and Volume
  - **E**) Charging and Discharging Rates
  - **H**) Balance of Plant

Budget

- Total Funding Spent*: $2,649,224
- Total DOE Project Value: $2,111,935
- Total Cost Share: $600,400

Partners

- **HSECoE Partners** - SNRL, PNNL, LANL, NREL, JPL, United Technologies, GM, Ford, BASF, Hexagon Lincoln, UM, UQTR
- **Center Lead** - SNRL

* As of 1/31/15

Relevance -Objectives

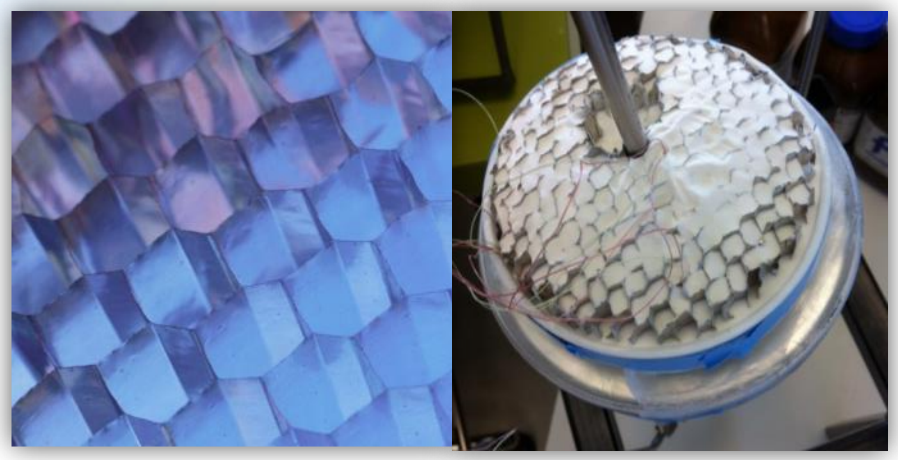
- **Phase 3 Objective** – Use enhanced heat and mass transfer available from arrayed microchannel processing technology to design, fabricate and test a modular adsorption task insert (MATI) prototype. The objective of phase 3 is to *demonstrate fundamental technical feasibility and validate simulations*. Smart goals include:
  - **June 30th 2015 Smart Goal** - Demonstrate performance of subscale system evaluations and model validation of a 2L adsorbent system utilizing a MATI thermal management system having 54 g available hydrogen, internal densities of 0.10g/g gravimetric, and 27 g/L volumetric.
- **Barriers Addressed**
  - Reduce system size and weight (Barrier A)
  - Charging and Discharging rates (Barrier E)

Relevance – Modular Adsorption Tank Insert (MATI)

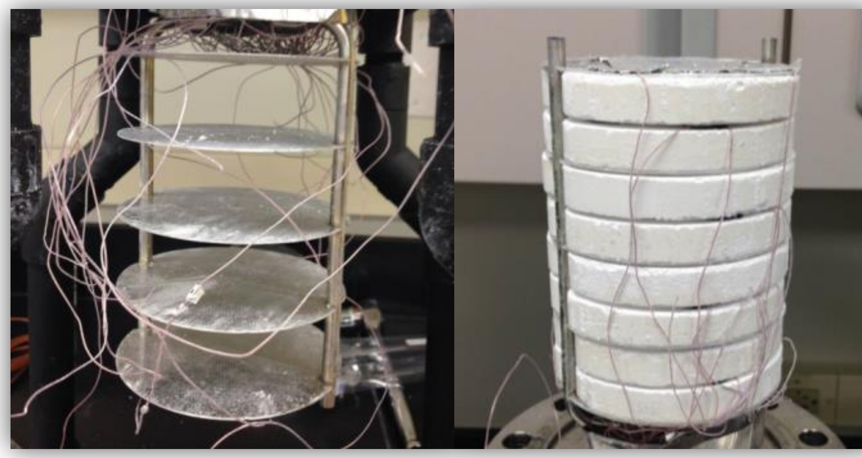
- Optimized for use with densified adsorbent media
  - Low void faction (<5%)
  - Insensitive to mechanical failure of the media
- Facilitates use of fuel cell waste heat for storage discharge improving onboard efficiency from 90% for resistance heating to 98%
- Separates cooling function from adsorption material allowing a wider range of cooling strategies
- Attractive high volume, low cost manufacturing options exist.

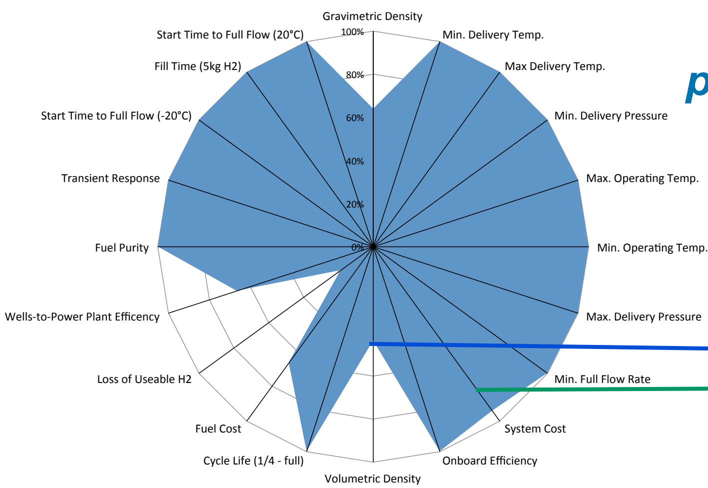
Adsorbent Heat Exchanger Types

HexCell
Flow Through Chilled H_2 Cooling

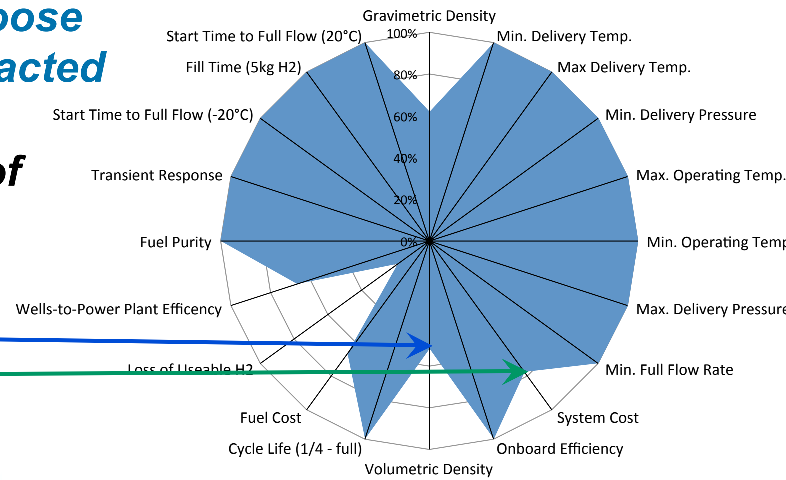


MATI
Isolated LN2 Flow Cooling





Gain Volumetric Density in going from loose powder to compacted pucks
at expense of Cost



Approach – Technical and Programmatic

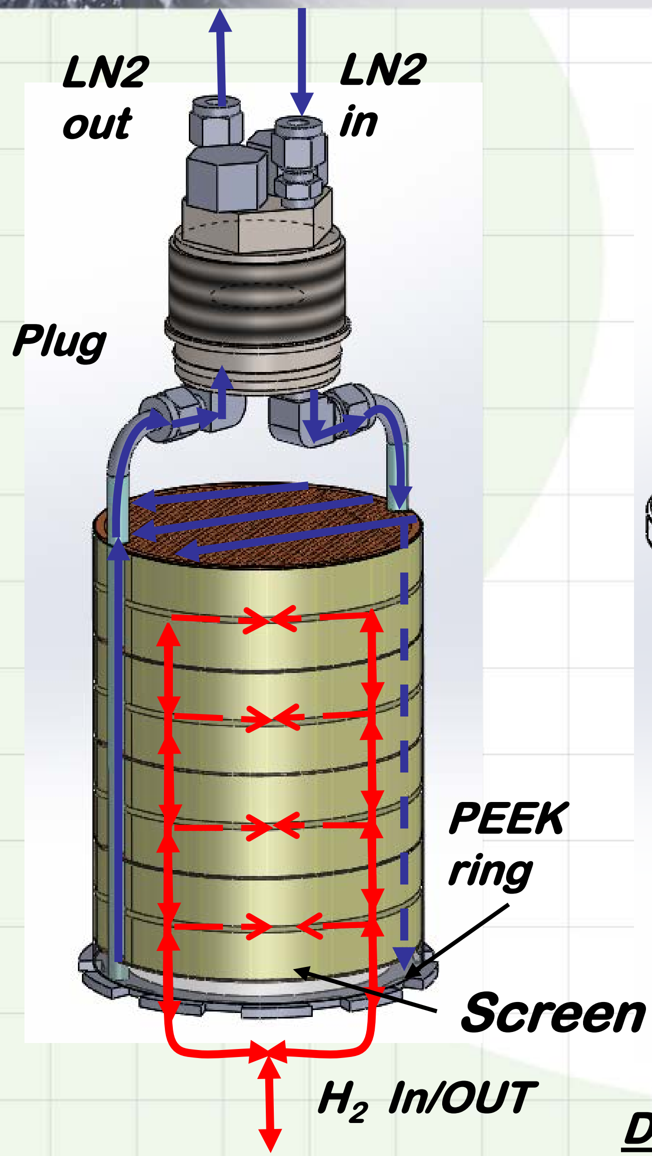
- **Phase 3: MATI Subsystem Prototype Construction, Testing, and Evaluation:**
  - Using simulation and previous experience from *Phase 1* and 2, develop a design for the MATI that achieves the performance included in our first smart goal.
  - Fabricate several MATI prototypes.
  - Conduct acceptance testing at OSU.
  - Conduct performance testing at SRNL to demonstrate our second smart goal.
  - Validate simulations against performance results.
  - Demonstrate a variant with conduction enhanced pucks

Technical Accomplishments

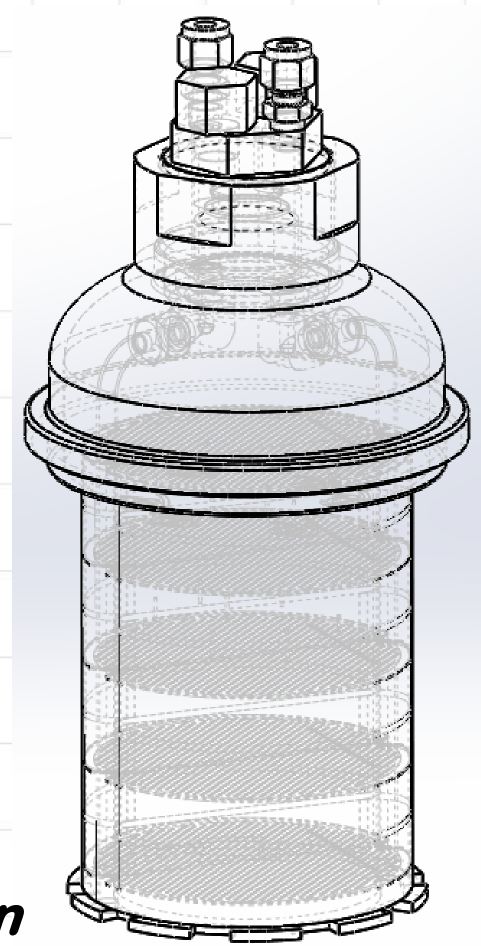
- **Technical Progress Relative to 2015 Milestone –** Completed the design, assembly and pressure testing of two MATI prototypes which were delivered to SRNL for testing.

- **Technical Progress relative to Objectives**: Reduce the size and weight of storage and Improve charging and discharging rate of storage – **MATI**
  - Completed assembly and pressure testing of three prototype MATI's
    - ✓ Two have been shipped to SRNL for performance testing
    - ✓ One will be used at OSU for conduction enhancement
  - Conduction enhanced pucks have been designed and are being fabricated
  - Completed assembly of OSU test apparatus for acceptance testing and testing of conduction enhancements
  - In collaboration with SRNL, completed modifications to simulation to allow model validation

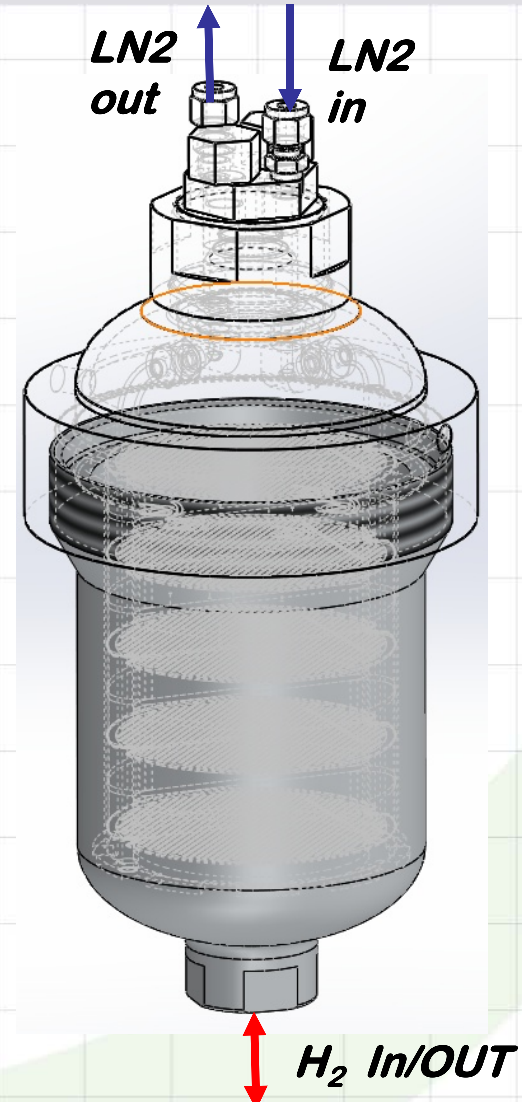
Barriers A and E – MATI Design Overview



MATI connected to the Plug



Dome threaded onto the Plug



Housing attached to the dome

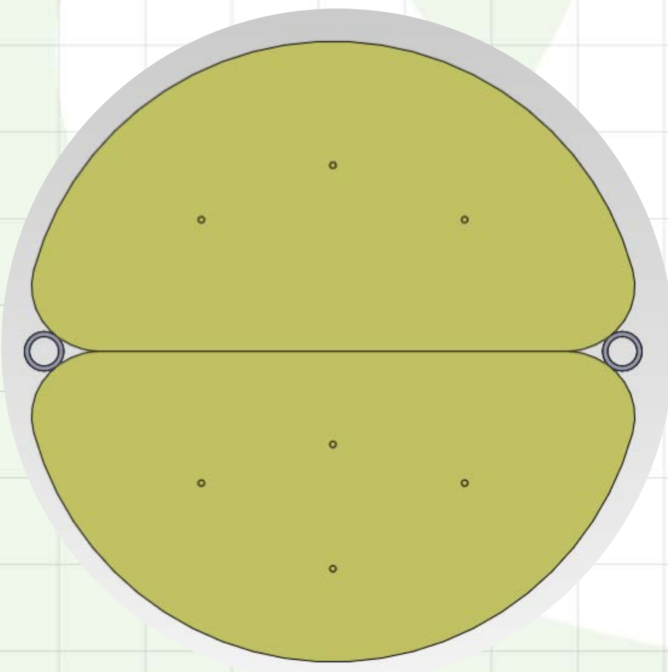
Barriers A and E –MATI Functional Criteria and Design Specifications

- ***<u>Phase 3 MATI Functional Criteria</u>*:**
  - Provide data for model validation instead of meeting specific DOE goals
  - Sufficient temperature measurements within the MOF beds, cooling plates and tank interior surface
  - Fit inside a 2 liter aluminum tank with minimal thermal communication
  - Withstand 100 bars external pressure during adsorption and up to 35 bars internal pressure during desorption
  - Durable puck design for both testing and transportation
  - Sizable H2 storage capacity within 3 min charge cycle

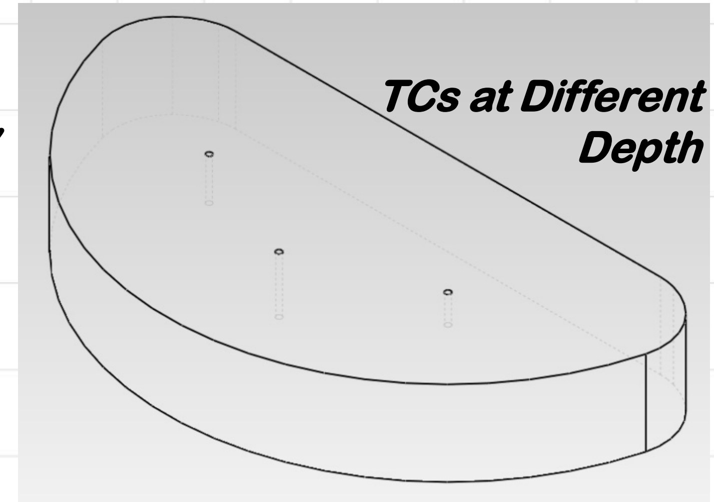
- ***<u>Key MATI Design Specifications</u>***
  - Simple baseline MATI design
  - Stainless steel construction for low fabrication risk
  - Relatively high MOF bed density (e.g. ≥ 0.4 g/cc) for puck integrity

Barriers A and E – Puck Design (Ford and University of Michigan)

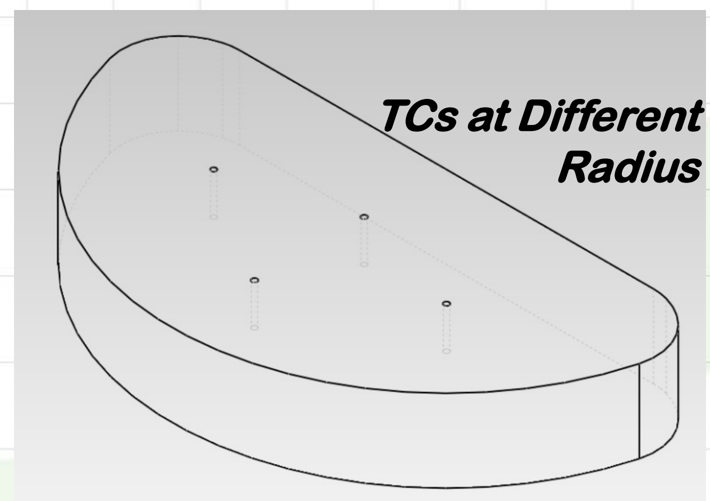
- ***10 cm diameter to be fit inside the 2 liter tank***
- ***1.5 cm bed height based on adsorption simulation and consideration of bed durability***
- ***Relatively high bed density (e.g. 0.4 g/cc) for puck integrity***
- ***Rounded design with approximate TC locations shown (details to be decided)***



Half-bed design is to maximize MOF-5 volume while enabling the assembly process

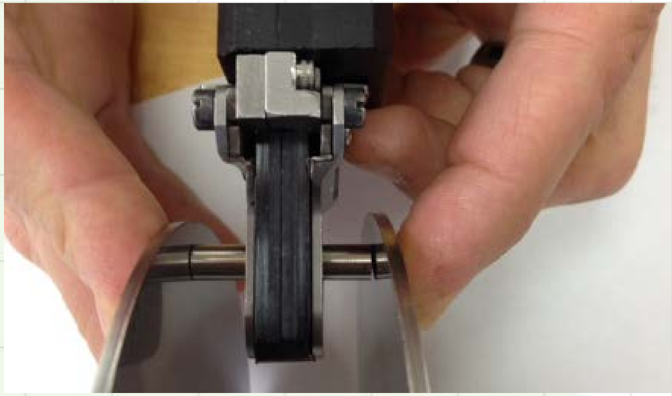


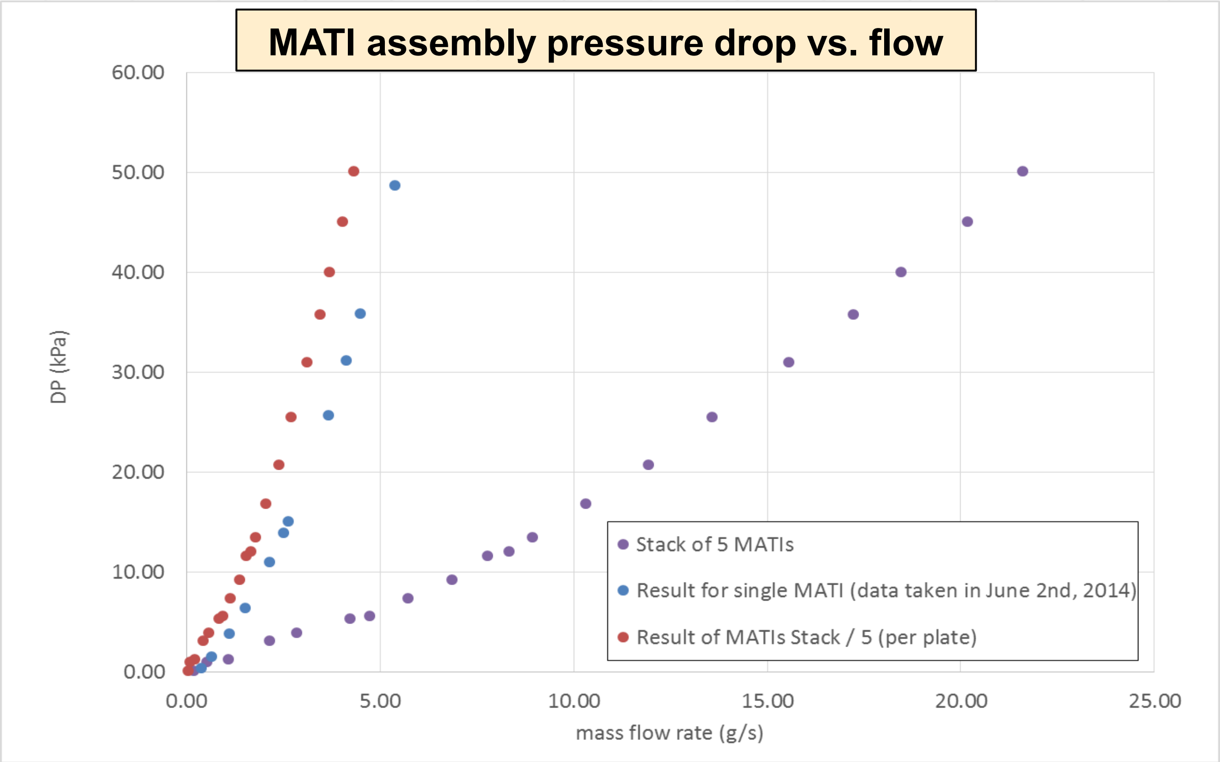
Current Half-bed Design



Barriers A and E – MATI Test Article Assembly and testing

- ***Cooling plates were photochemically machined & diffusion bonded.***
- ***Two header tubes were vacuum brazed onto each cooling plate, allowing each cooling plate flow examination.***
- ***Cooling plates were stacked together using orbital welding.***





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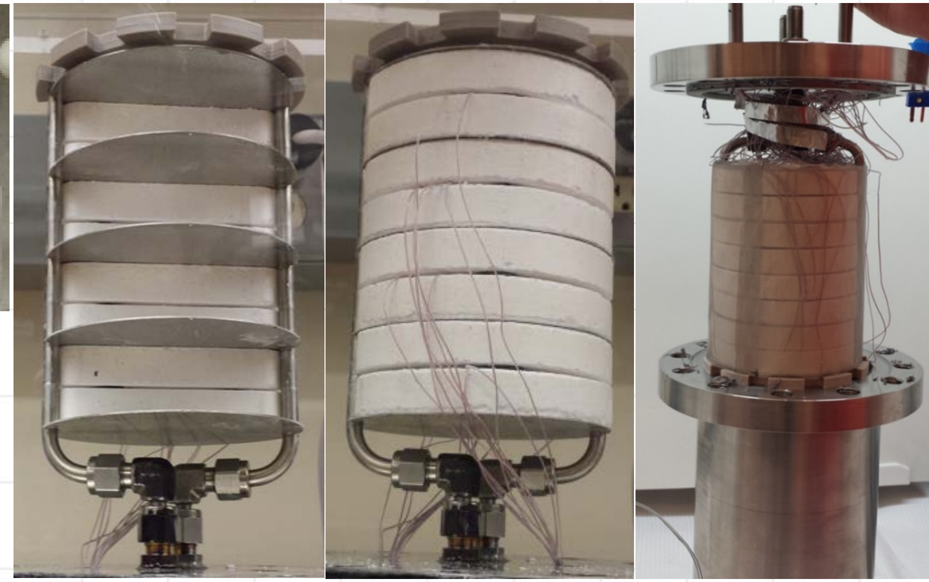
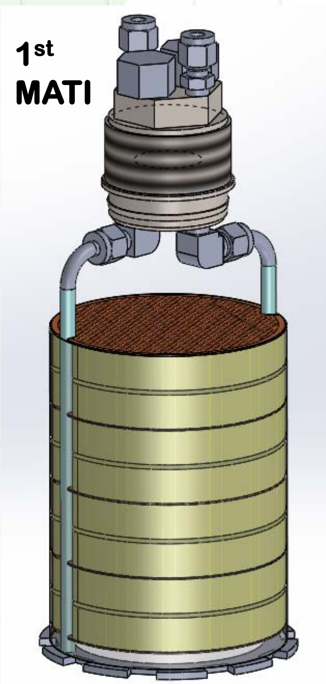
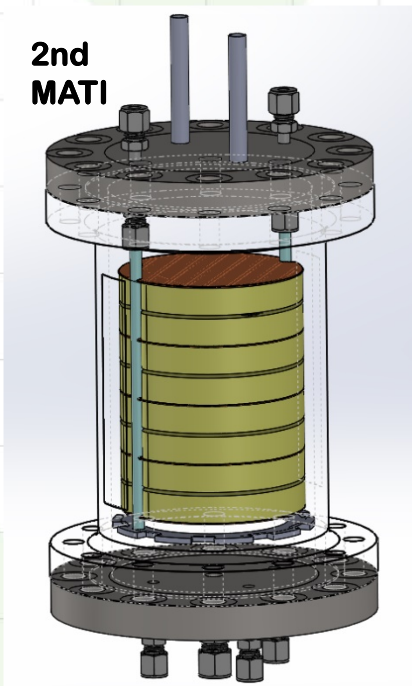






- *Original Aluminum tank with plug design failed to seal under cryogenic conditions.*
- *1st MATI assembly was installed in flanged tank and included 32 unsheathed fine gage Thermocouples.*
- *Puck assembly took place inside a glove box to minimize exposure to air.*
- *Three MATI prototypes have been assembled.*
- *2nd and 3rd MATIs were made for flanged tank and LN_2 tubes do not require a bend.*

Barriers A and E – Experimental system Requirements

- Fabrication and Assembly of MATI and Integrated Storage Vessel
  - Individual MATI cooling plate was exposed to 100 bars external differential gas pressure and demonstrated no measurable deflection of cooling plate that could result in flow maldistribution.
  - Individual MATI cooling plate was exposed to 100 bars internal differential pressure and demonstrated no measurable deflection, delamination or leakage that would render it inoperable.
  - Fully assembled five plate MATI was installed in stainless steel flanged pressure vessel

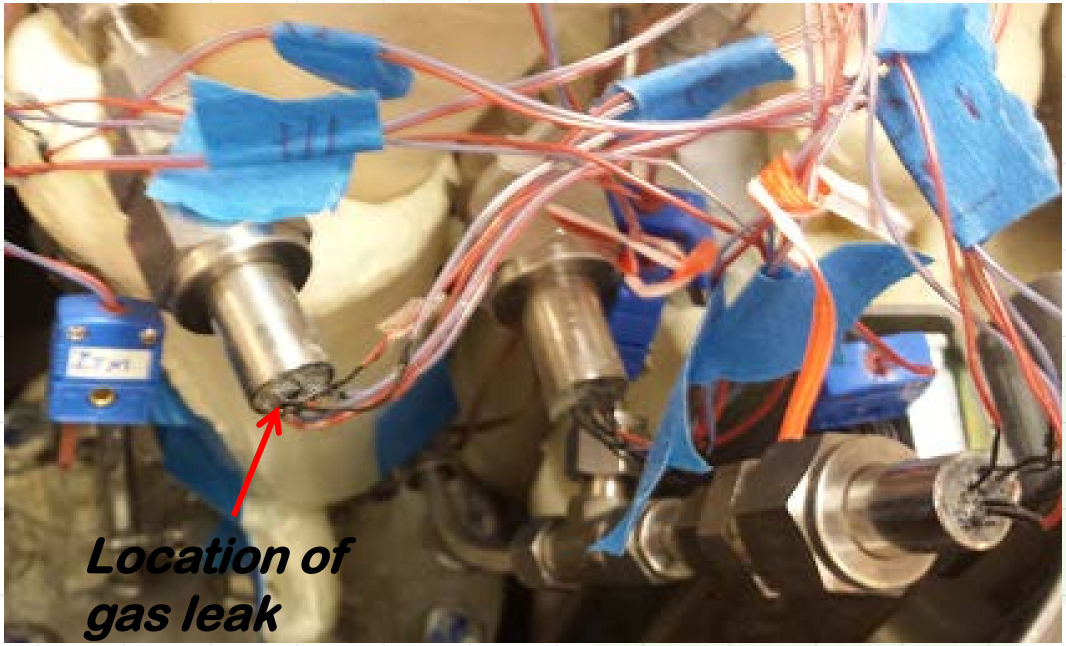
Barriers A and E – Integrated System Assembly

Barriers A and E – Integrated System Assembly

- OSU assembled and sealed an integrated pressure vessel containing 16 compacted MOF-5 beds, 30 fine gauge wire thermocouples (one thermocouple located in 14 of MOF-5 beds, one bed containing three thermocouples, one bed containing four thermocouples, two thermocouples on outside surface of MOF-5 beds, four located on non-contacting surface of MATI plate, three on inside surface of stainless steel vessel)
- Passed 48 hour pressure decay test at 100 bar using Helium
- In-situ activation of MOF-5 beds – held system under vacuum for 24 hours. Subsequently wrapped electrical heating tape around pressure vessel and heated to 115^{o}C under vacuum for 24 hours.
- Performed and passed second pressure decay test at 100 bar using He
- Upon first addition of LN_2 around the pressure vessel, three of eight thermocouple seals failed causing gas leaks out of vessel

Barriers A and E – OSU Acceptance Testing

- Upon cryogenic LN2 addition, approximately 32" below location of thermocouple pass-through, gas began to leak along thermocouple wire
- Attempted to fix using a secondary application of epoxy on outside of tube and extending ½" above top of tube end, however exposure to LN_2 cause cracking of epoxy
- After attempted OSU fix failed and a successful cryogenic sealing solution was developed by SRNL, the decision was made in conjunction with SRNL to ship non-functioning MATI system to SRNL for complete testing



Barriers A and E - Modeling

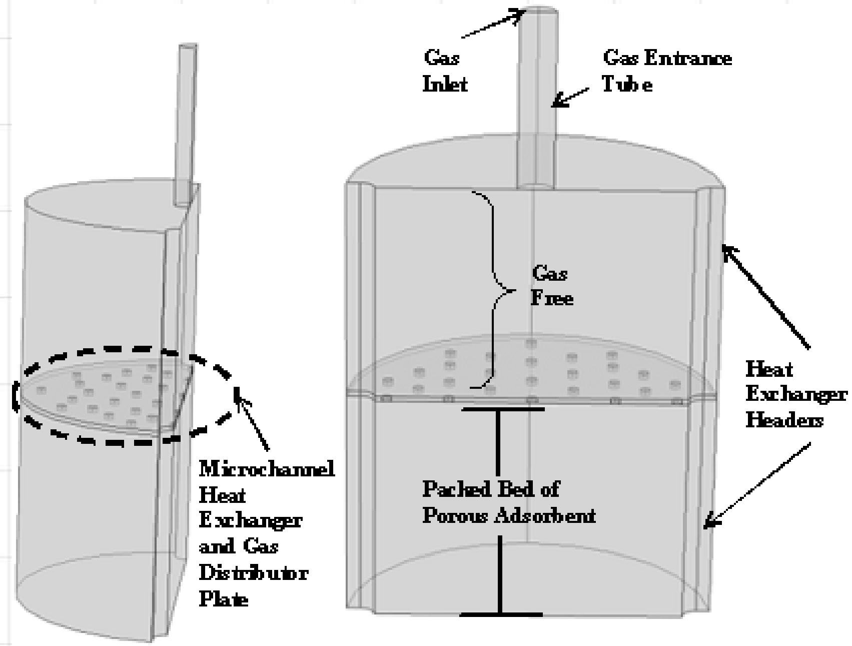
- Systematic enhancement of OSU – Phase II integrated COMSOL modeling effort
  - Improve simulation results for the adsorption of H_2 on compressed MOF-5 beds
  - Transition from ideal gas law concentration (density) calculation and double interpolation of remaining fluidic and thermal properties (Phase II) to polynomial calculation developed by Savannah River National Laboratory (Phase III)
  - Phase II simulation utilized variable isoteric heat of adsorption to determine the heat released due to adsorption of H_2 within compressed bed
  - Phase III utilizes internal energy and enthalpy to determine the energy change and heat released during the adsorption process

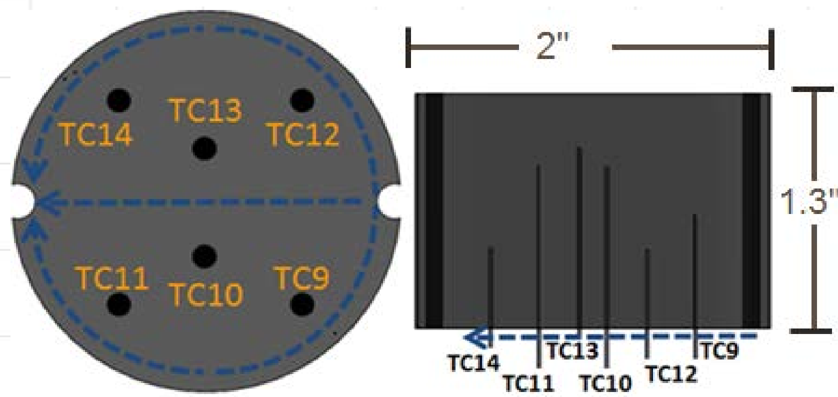
B. Hardy, C. Corgnale, R. Chahine, M.-A. Richard, S. Garrison, D. Tamburello, D. Cossement, and D. Anton, "Modeling of adsorbent based hydrogen storage systems," International Journal of Hydrogen Energy, vol. 37, no. 7, pp. 5691–5705, Apr. 2012.

Barriers A and E - Modeling

An improved representation of the Phase II experimental data from the simulation was achieved with help of SRNL and updates to H_2 properties and kinetic expression

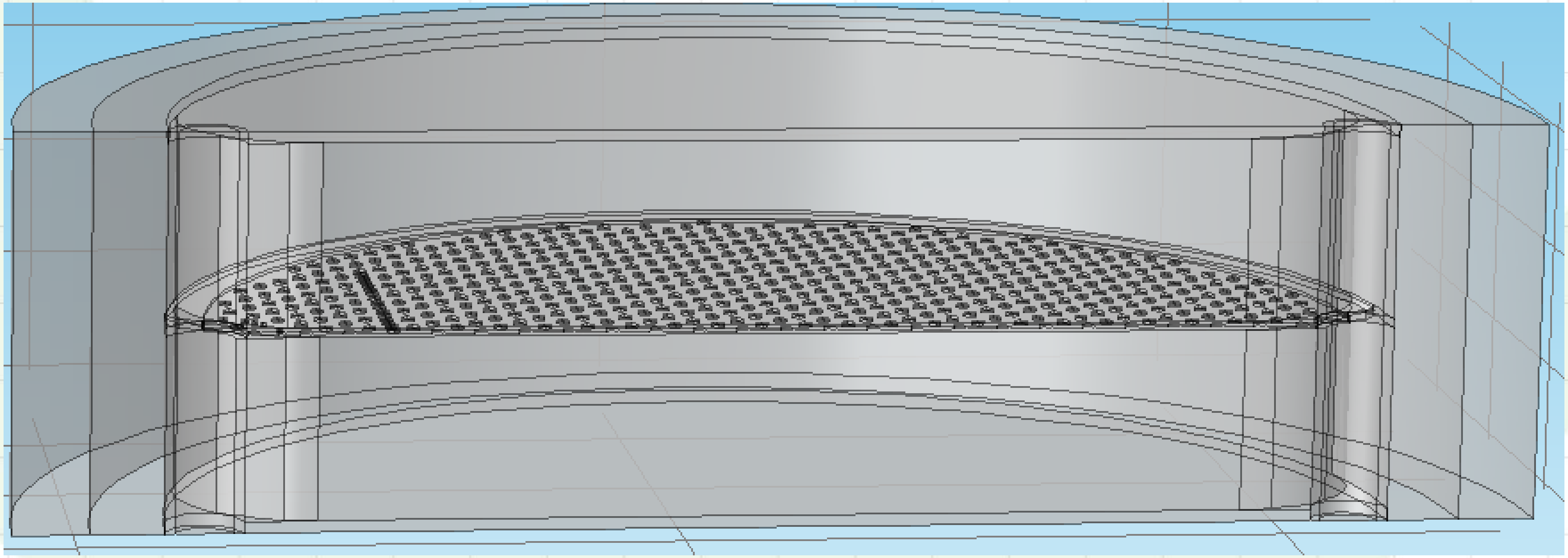
- Lowered maximum absolute and relative errors at all 6 thermocouples in porous bed;
- Reduced average relative error below 3% at all thermocouples;
  – Phase II resulted in 4 of 6 below 3%
- Reduced high average absolute error to 3.4 % from 5.8%



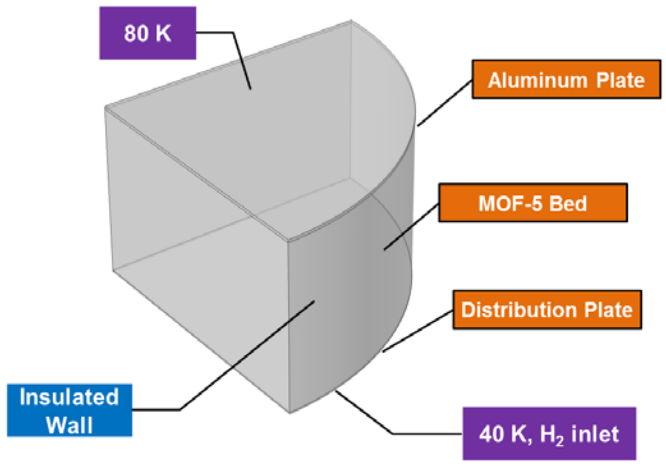


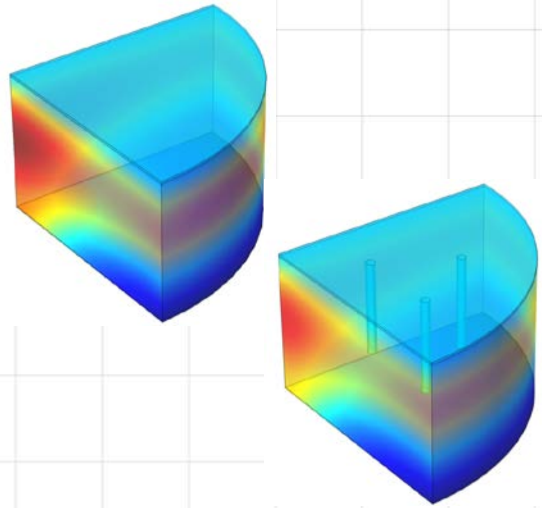
Barriers A and E - Modeling

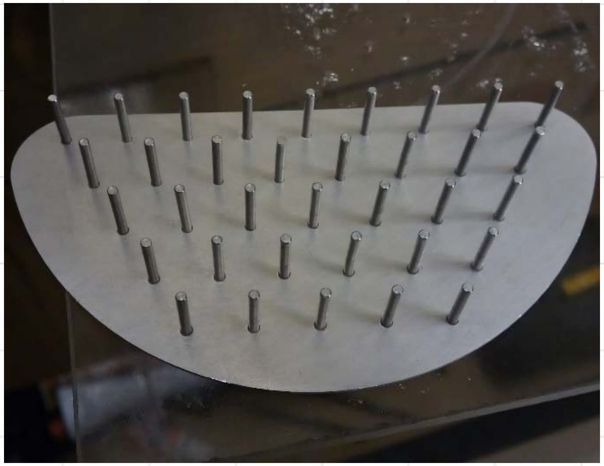
- Single unit cell consists of
  - One single cooling plate with headers
  - Heat transfer fluid flow path
  - Two half height H_2 distribution layers
  - Two half height MOF-5 beds
  - Annular region between MATI and tank
  - H_2 void spaces resulting from round bed design
  - Stainless steel storage tank
- To fully model all regions of H_2 gas flow, heat transfer fluid flow and the adsorbent material, a model geometry was created in SolidWorks and transferred within COMSOL using Livelink™ license.
- To model this complex geometry OSU has acquired a new server utilizing 24 cores and 256 GB ram.

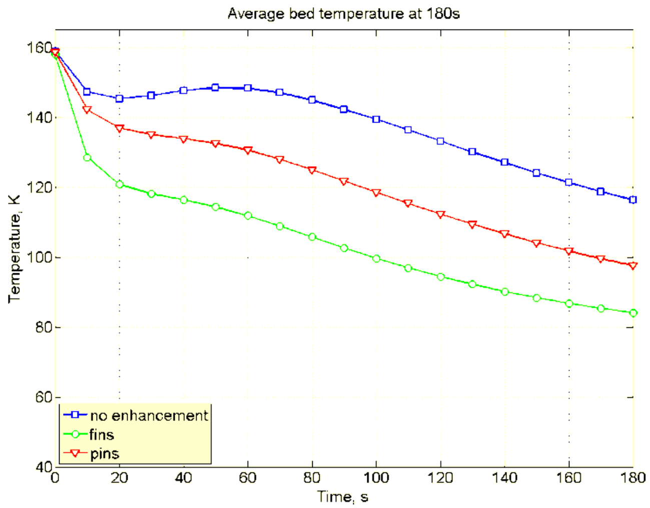


Barriers A and E – Conduction Enhancement









- ***Adsorption and desorption are heat transfer limited due to poor thermal conductivity of MOF***
- ***Aluminum pins can greatly reduce charge and discharge time***
- ***Tests will involve 1) Al pin enhanced pucks, 2) Al pin + ENG enhanced pucks and 3) ENG enhanced pucks***

Response to Previous Year Reviewer Recommendations

- **“A useful addition would be the development and testing of a method for enhancing the thermal conduction within the media puck”** – Conduction enhancements have been fabricated and are being tested as part of our Phase 3 scope of work.

- **“OSU should confirm the conceptual design for the MATI device via experiment and confirm reliable separation of fluids” –** The MATI has been successfully pressure tested at OSU and repeatedly tested at SRNL.

- **“Fabricate Multiple MATI devices”-** Three MATI prototypes have been fabricated at OSU and we have parts for a fourth MATI.

Remaining Challenges and Barriers

- Complete testing of conduction enhancements
- Complete model validation
- Complete final reporting

Proposed FY 2016 Future Work

- This project is completed as of June 30th. No work is planned for FY 2016.

Collaboration

- Oregon State University is a member of the Hydrogen Storage Engineering Center of Excellence (HSECoE) collaborating with five federal laboratories, one university and six companies.
- Development of the Modular Adsorption Tank Insert Pressure Vessel is in collaboration with Hexagon Lincoln.
- Development of densified MOF-5 puck in collaboration with Ford and University of Michigan.
- Developed design of acceptance test apparatus and test plan in collaboration with SRNL.
- Developed simulation for code validation in collaboration with SRNL.

Technology Transfer Activities

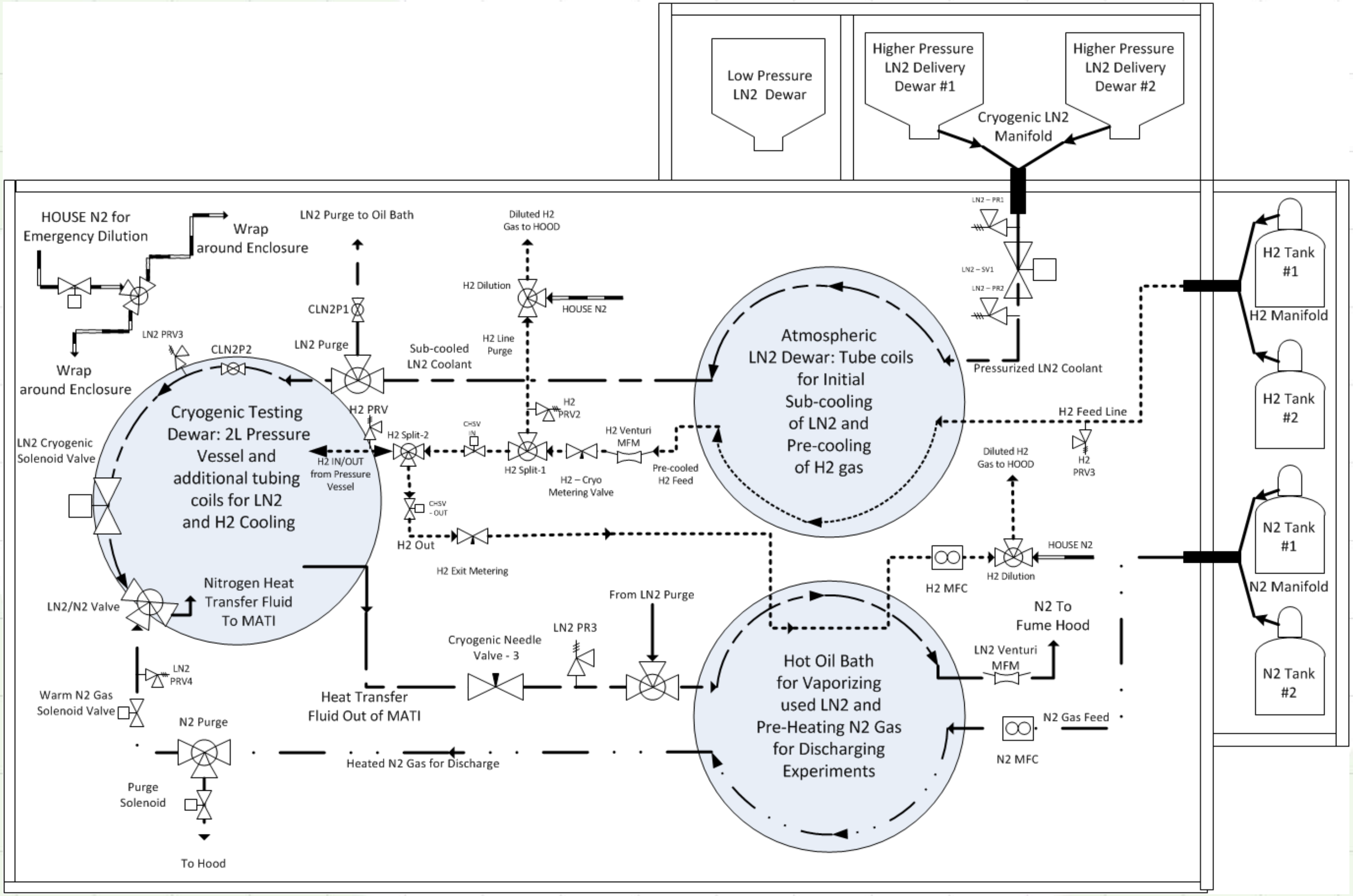
- A invention disclosure for the MATI has been filed with OSU and is being evaluated

Project Summary

- **Relevance**: The Modular Adsorption Tank Insert (MATI) can reduce size, weight and charging time of hydrogen storage.
- **Approach**:
  - Using simulation and previous experience from Phase 1 and 2, develop a design for the MATI that achieves the performance included in our first smart goal.
  - Fabricate several MATI prototypes.
  - Conduct performance testing at SRNL to demonstrate our second smart goal.
  - Validate simulations against performance results.
  - Test conduction enhanced pucks supplied by Ford
- **Technical Accomplishments**:
  - Completed assembly and pressure testing of three prototype MATI’s
    - ✓ Two have been shipped to SRNL for performance testing;
    - ✓ One is used at OSU for conduction enhancement;
  - Conduction enhanced pucks have been designed and are being fabricated
  - Completed assembly of OSU test apparatus for acceptance testing and testing of conduction enhancements
  - With SRNL, completed modifications to simulation to allow model validation;
- **Collaboration**: Member of HSECoE team.
- **Proposed Future Research**:
  - i) Complete tests with conduction enhanced pucks; ii) Complete final reports

Supplemental Slides

Barriers A and E - Overall Integrated System Flow Sheet



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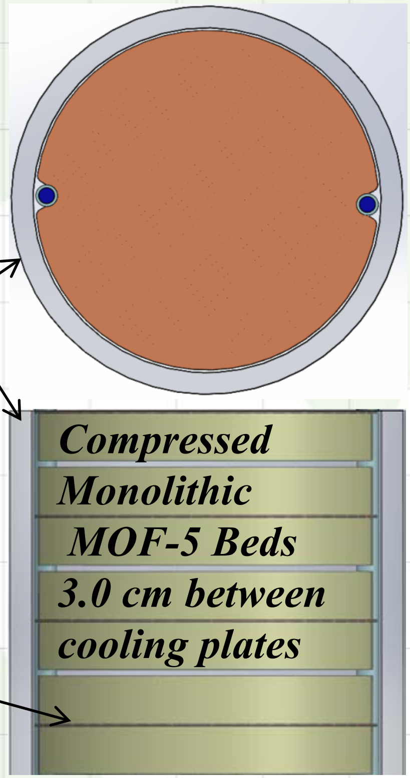
Barriers A and E - Integration with Monolithic Densified Media

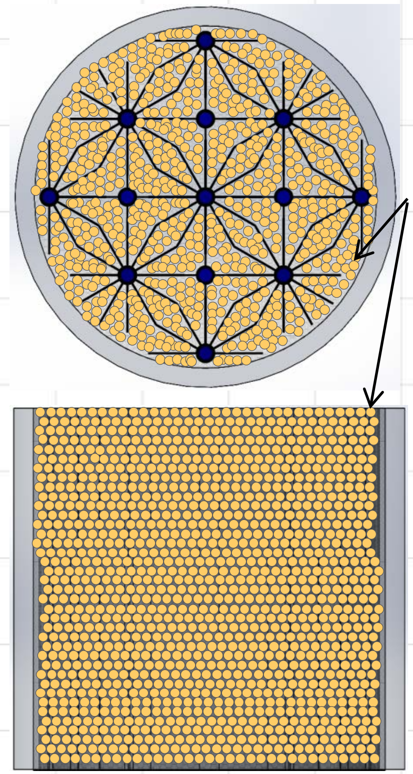
MATI Cooling Plate (on top)

Al Tank



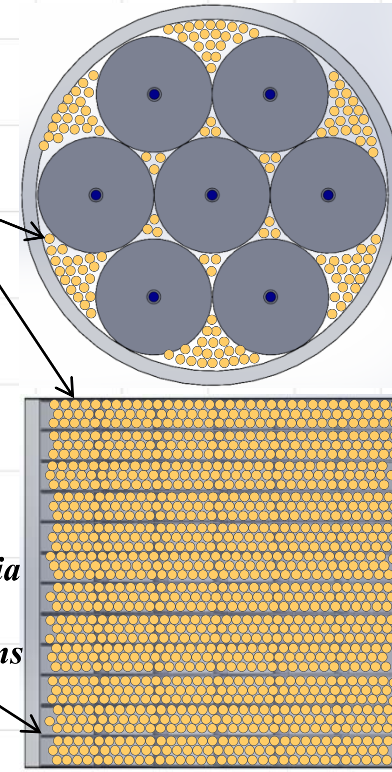
MATI Cooling Plates

Axially aligned Fin and Tube



Packed MOF-5 pellets or particles required

Circumferential Cooling tubes and Fins



Circumferential Cooling Tubes and Fins (1 cm fin spacing)

-Convenient Integration of Monolithic densified media
-95% densified media

-No known way to integrate monolithic densified media, requires pellets or powers
-60 to 80% media

-No known way to integrate monolithic densified media, requires pellets or powers
-60 to 80% media

Conclusion – MATI allow more media in a given volume than do finned tubes

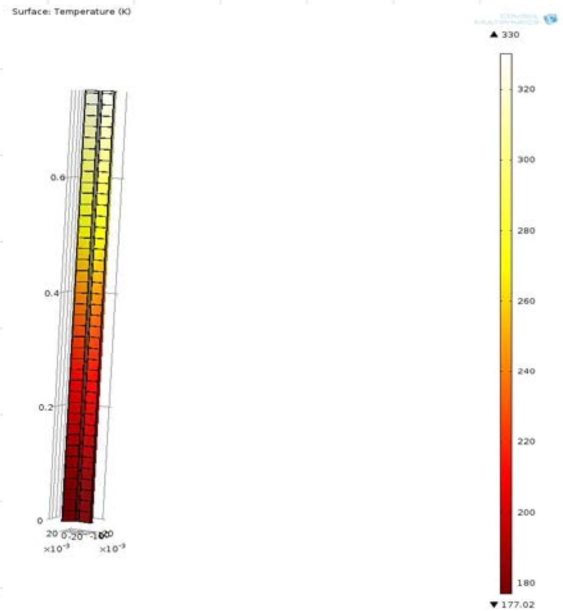
Barriers A and E - Simulation of Axial Fin tube for MOF-5 Adsorption (conducted by SRNL)

Modeling Assumptions

- ***Length of Cylinder = 0.75m***
- ***Diameter = 3.635 cm***
- ***0.25 inch OD tubing***
  - ***Yields 2 cm of MOF surrounding the tubing***
- ***80 Aluminum axial fins***
  - ***0.4 mm thick***
  - ***Yields approximately 1 cm spacing between plates.***
  - ***8% of volume is metal or flow path***
- ***Would require on the order of 60 tubes with 120 welds***

Results for H_2 supply of 1.6g/s)

- ***Supply power of 3600 W is needed.***
- ***H_2 Max supply power (highest Δ T and flow rate) is 3000 W***
- ***H_2 supply power decreases to 1000W during 1.25 hour desorption.***
- ***Combustion of hydrogen must supply more 50% of discharge heat***



Simulation of Axial Fin Tube (SRNL)

OSU Conclusions –

1) ***The performance of the axial fin tube is seriously degraded by axial conduction this results in the axial fin tube requiring 50% of discharge heating to come from hydrogen combustion as compared to 15% in MATI***
2) ***8% of the volume is metal or flow path and is unavailable for media, in the MATI, 5% is metal or flow path***
3) ***Would require 120 welded joints as compared to 30 in a MATI***

Barriers A and E - Conclusions

- We do not know of a method for integrating a densified monolithic media with either an axial fin or circumferential fins. The use of pellets and powers will result in a significant increase in volumetric density (i.e. a larger tank will be required for the same energy storage capacity)
- Based on the one design the center has produced for a MOF-5 fin-tube heat exchanger
  - **The performance of the axial fin tube is seriously degraded by axial conduction this results in the axial fin tube requiring 50% of discharge heating to come from hydrogen combustion as compared to 15% in MATI**
  - **8% of the volume is metal or flow path and is unavailable for media, in the MATI, 5% is metal or flow path**
  - **This design would require 120 welded joints as compared to 30 in a MATI, however this depends on tank aspect ratio and a longer and more narrow tank would have fewer welded joints for the axial fin tube design and more for the MATI.**
- Based on these results we do not plan on spending any additional time on evaluating fin tube heat exchanger for this application