Microscale Enhancement of Heat and Mass Transfer for Hydrogen Energy Storage

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Hydrogen Storage Engineering
CENTER OF EXCELLENCE

ST 046

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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 Project Value: $2,570,509
- Cost Share: $521,685
- DOE Share: $2,048,824
- Total DOE Funds Spent*: $1,797,598
  - * As of 3/31/14

Partners

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

ONAMI
OREGON NANOSCIENCE AND
MICROTECHNOLOGIES INSTITUTE

μICROPRODUCTS BREAKTHROUGH INSTITUTE

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:

  – **Dec 31st 2013 Smart Goal** - Design a 2L adsorbent subscale prototype utilizing a MATI thermal management system having 54 g available hydrogen, internal densities of 0.10g/g gravimetric, and 27g/L volumetric.

  – **June 30th 2014 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)
  – Balance of Plant (Barrier H in Phase 1 and 2)

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.

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
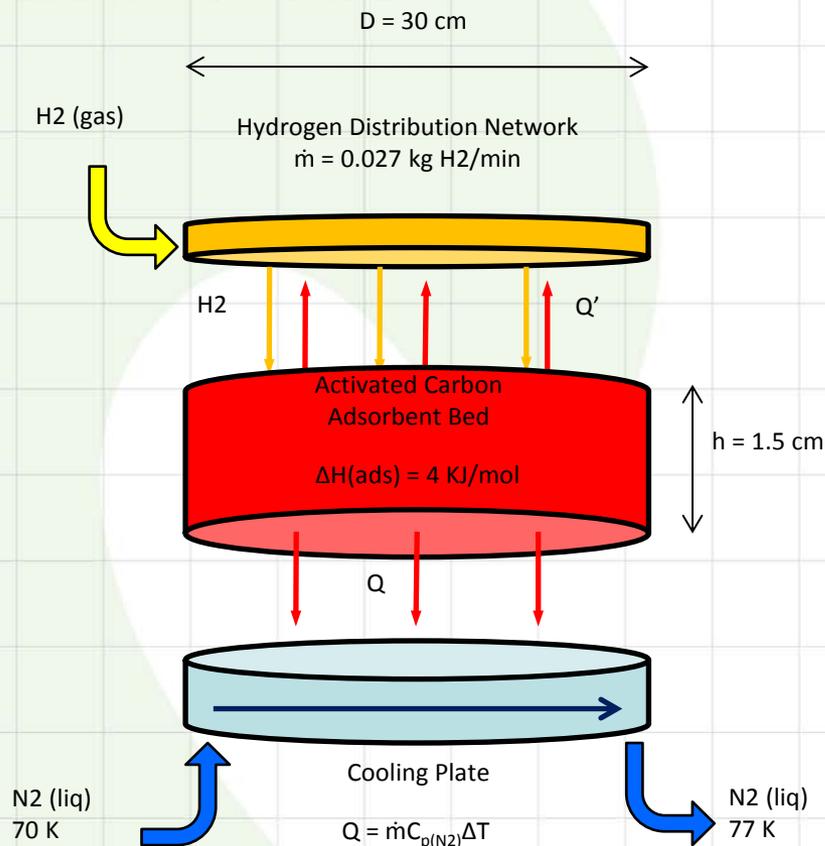
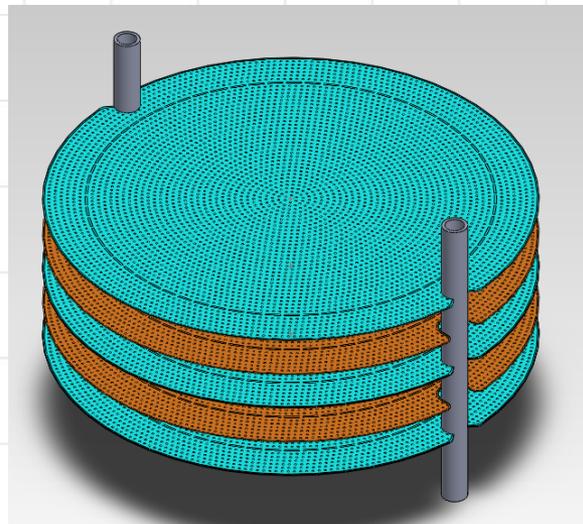# Approach - Project Gantt Chart
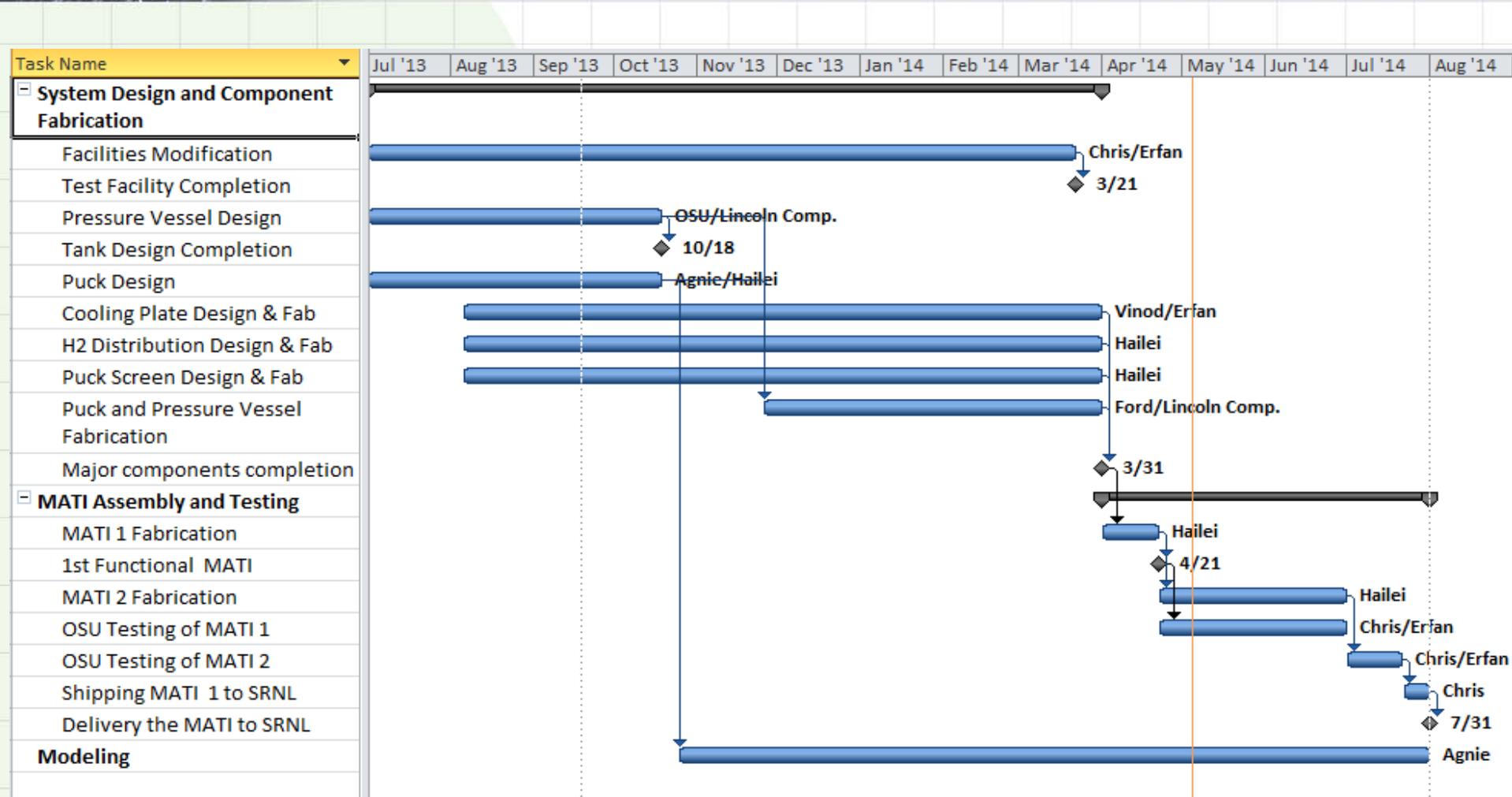
Technical Accomplishments

- **Technical Progress Relative to 2013/2014 Milestones –** Completed the design of the MATI prototype meeting our 12/31/2013 smart goal requirements;

- **Technical Progress relative to Objectives**: Reduce the size and weight of storage and Improve charging and discharging rate of storage – **MATI**
    - Completed design of MATI prototype which meets smart goals requirements
        - Pressure vessel and plug in collaboration with Hexagon Lincoln
        - Media puck in collaboration with Ford and University of Michigan
        - Design of cooling plates
        - Design of headers
    - Completed center design reviews of MATI prototype design, test apparatus design and test plan
    - Completed design of modified test apparatus for MATI acceptance testing
    - In collaboration with SRNL, completed modifications to simulation to allow model validation
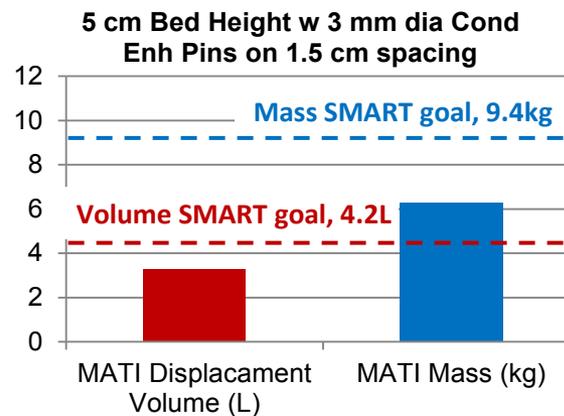
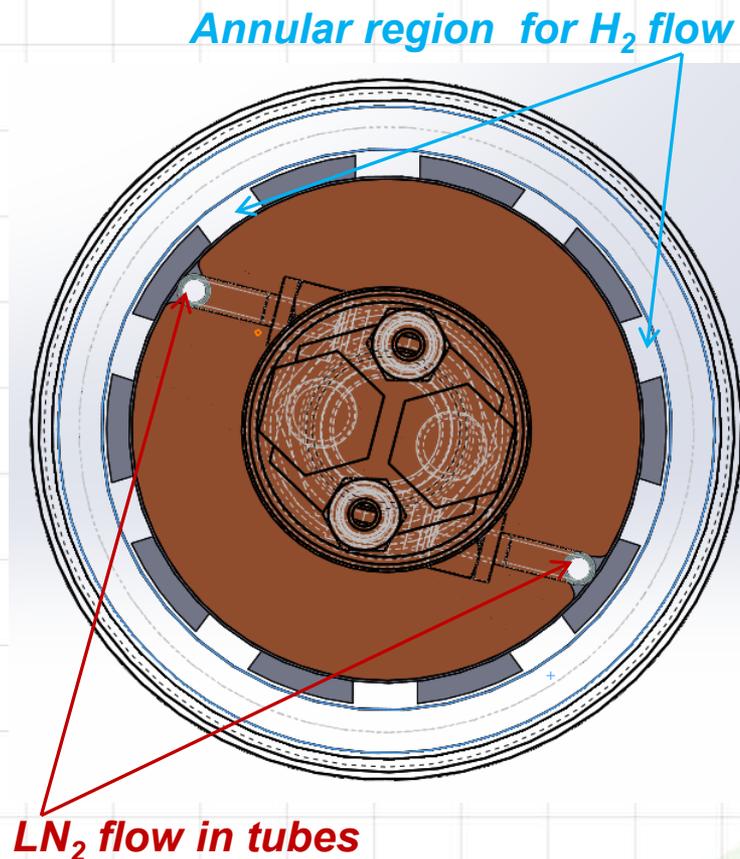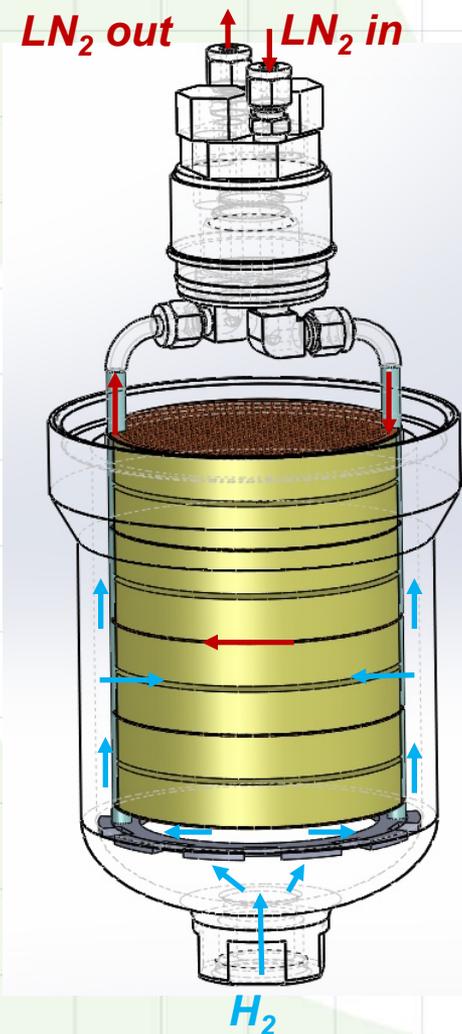# Barriers A and E - Modular Adsorption Tank Insert (MATI) Design Concept and Status

D = 30 cm

H2 (gas)

Hydrogen Distribution Network
$\dot{m}$ = 0.027 kg H2/min

H2   Q'

Activated Carbon
Adsorbent Bed   h = 1.5 cm

ΔH(ads) = 4 KJ/mol

Q

Cooling Plate

N2 (liq)
70 K   N2 (liq)
77 K

$Q = \dot{m}C_{p(N2)}\Delta T$

MATI Concept: can be enclosed to contain media

- Heat of adsorption removed by LN_2 and H_2
- Axial distribution of H_2 to adsorption bed;

5 cm Bed Height w 3 mm dia Cond Enh Pins on 1.5 cm spacing

Mass SMART goal, 9.4kg

Volume SMART goal, 4.2L

MATI Displacament Volume (L)	MATI Mass (kg)

LN$_2$ out   LN$_2$ in

Annular region for H_2 flow
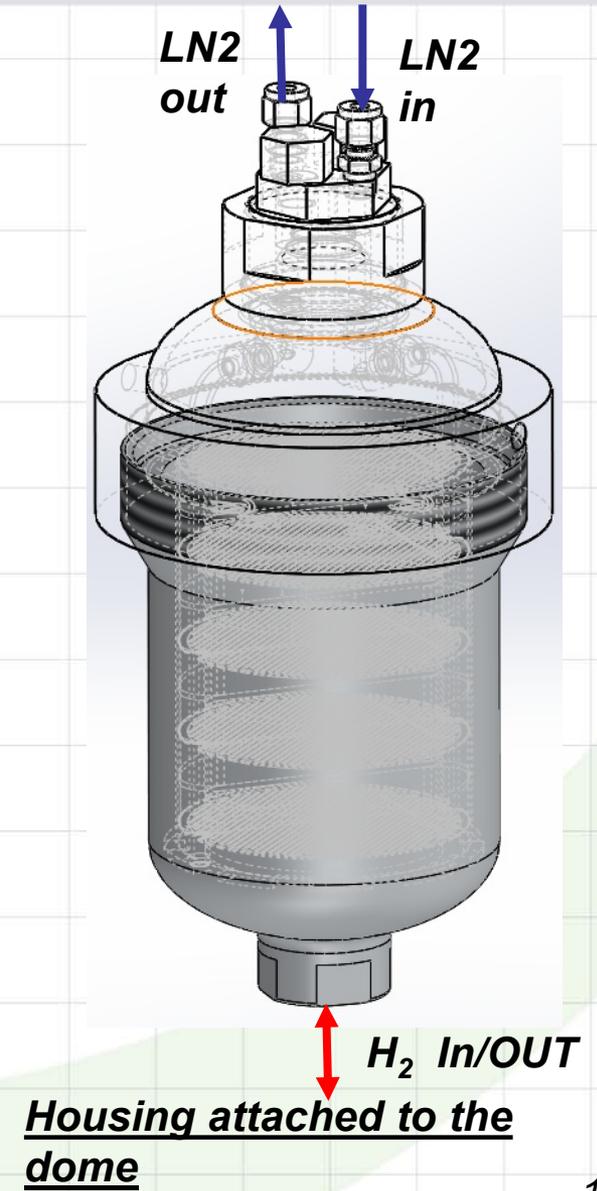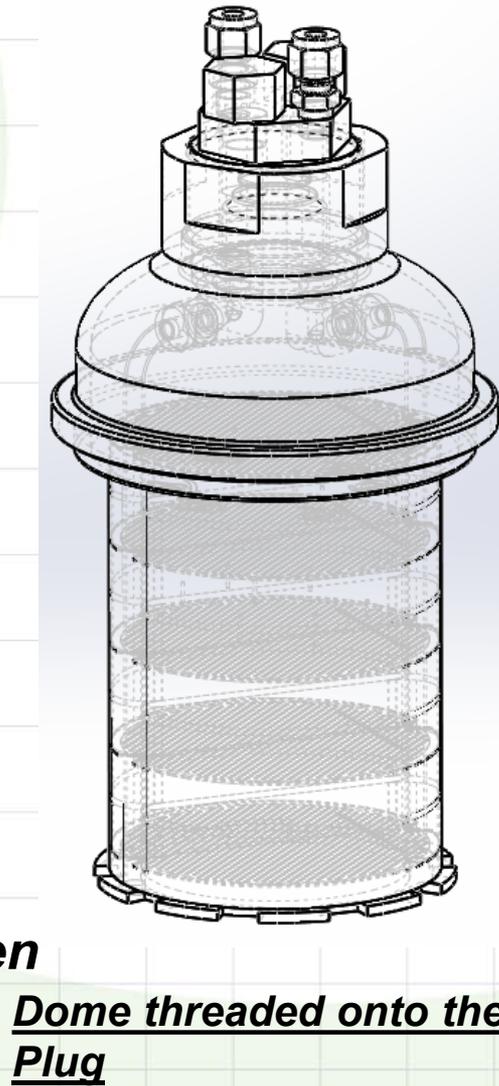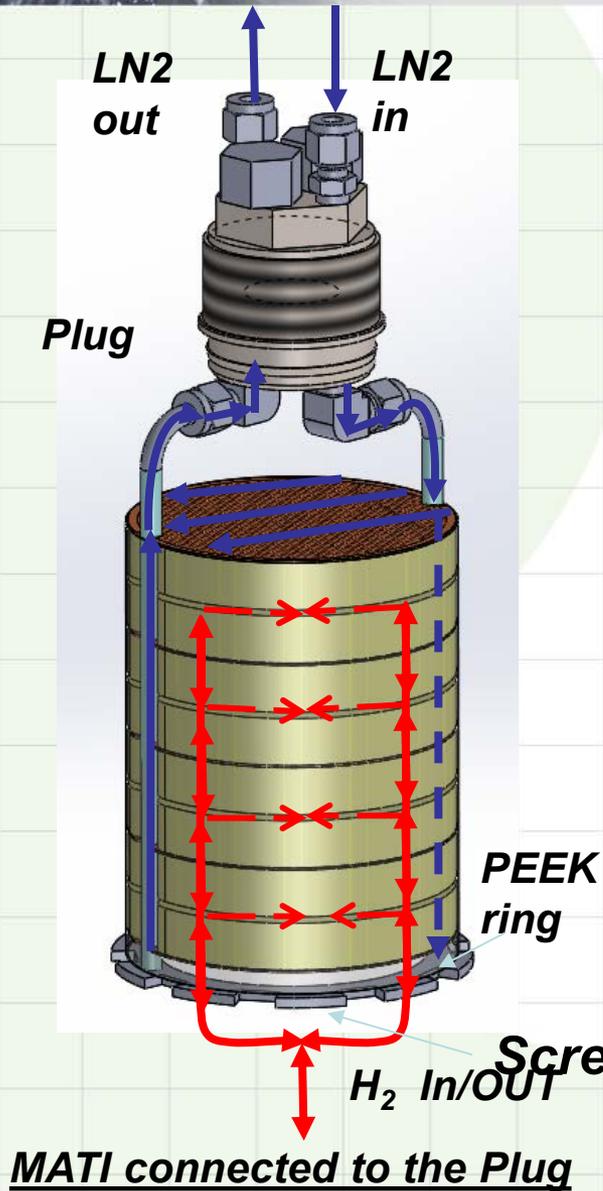
LN_2 flow in tubes

H_2

Barriers A and E –MATI Functional Criteria and Design Specifications

- **_Phase 3 MATI Functional Criteria_:**
  - 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

- **_Key MATI Design Specifications_**
  - 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 – Design Overview

LN2 out   LN2 in

Plug

PEEK ring

Screen

H_2 In/OUT

MATI connected to the Plug

Dome threaded onto the Plug

LN2 out   LN2 in

H_2 In/OUT

Housing attached to the dome

10

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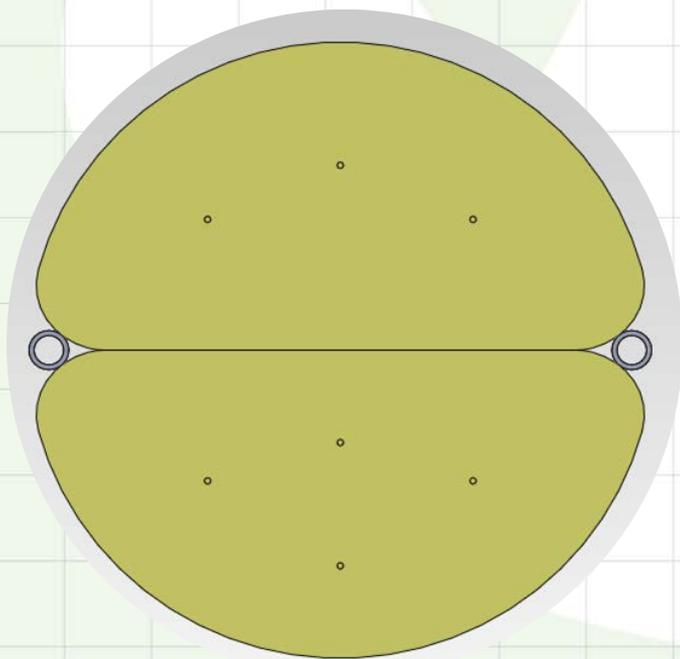
Half-bed design is to maximize MOF-5 volume while enabling the assembly process

TCs at Different Depth
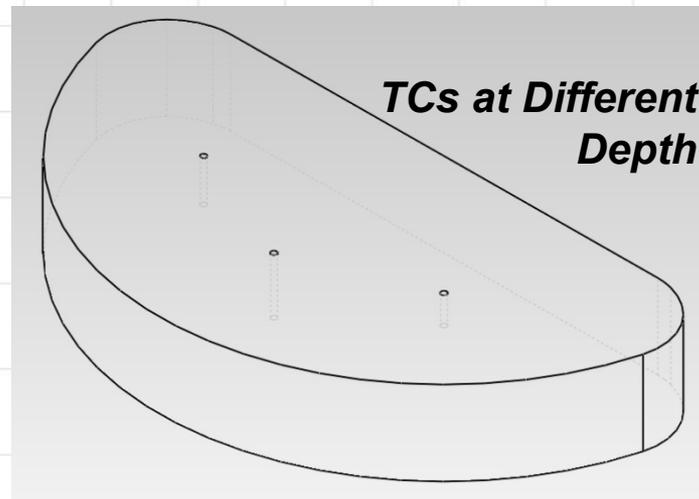
## Current Half-bed Design

TCs at Different Radius
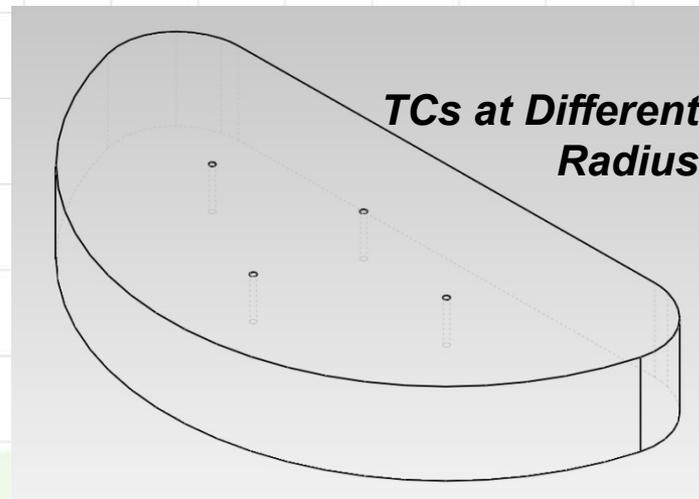
Barriers A and E – Instrumentation

Measurements of Interest:

- At least 1 embedded TC in each bed
- Heavily instrument at least 1 bed with TC's at multiple depths and radial dimension
- External MOF-5 surfaces
- Cooling plate surfaces
- Tank internal wall
- Internal System Pressure
- MATI heat transfer fluid inlet/outlet temperature
- H2 (dis)charging rate
- Heat transfer fluid pressure and ΔP

Thermocouples:

- Interior tank wall
- MOF5 puck surface
- At depth within MOF5 puck
- MATI plate surface

Heavily instrumented "puck"
(5 to 10 TCs)
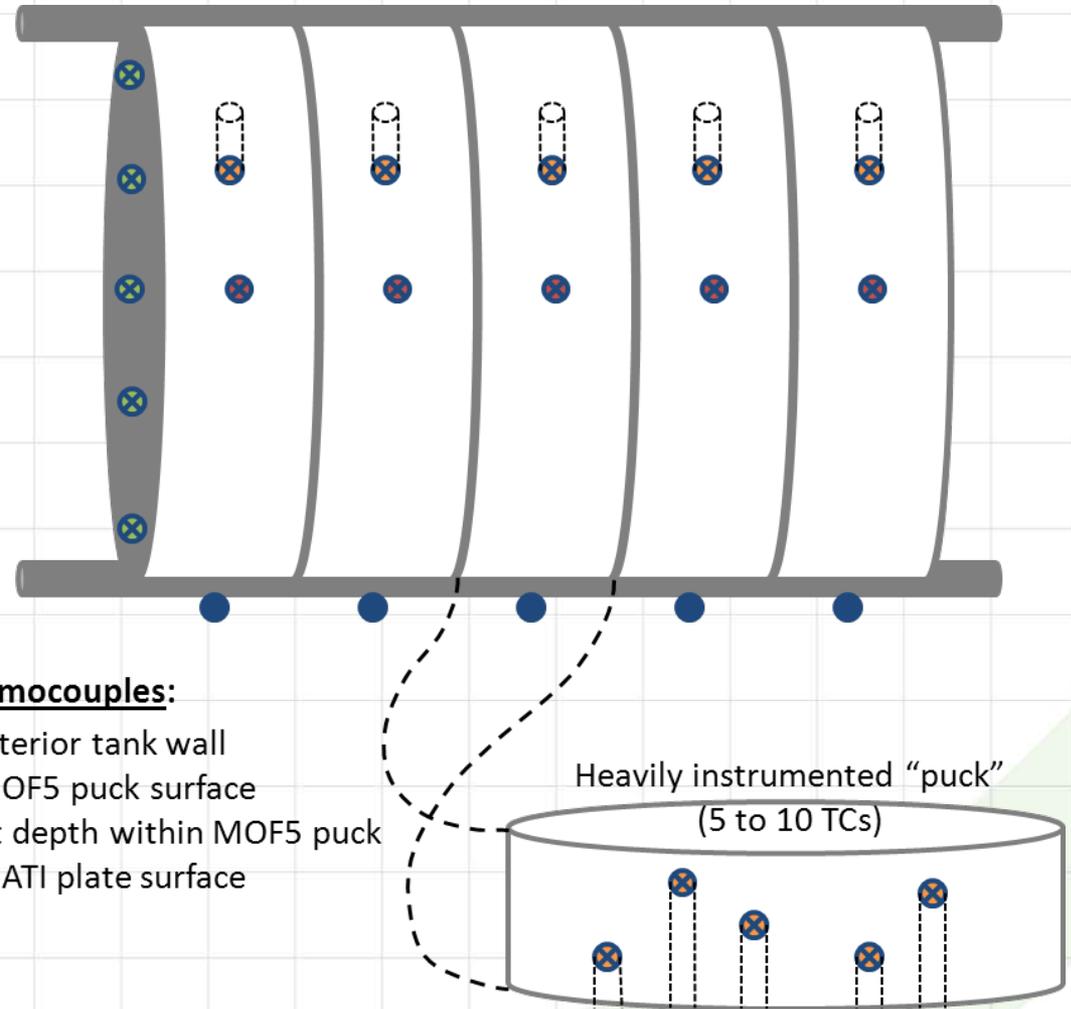
Barriers A and E – MATI Cooling Plate Design

Design considerations for the Phase III MATI prototype:

➢Withstand external compressive pressure of 100 bar and an internal pressure of 35 bar

➢Have uniform flow distribution (Phase 2 design was not optimal in this regard-Local velocity and temperature contours indicate room for improvement of performance by more uniform flow distribution, see fig.)

➢Meet all the fabrication constraints associated with chemical etching and diffusion bonding.

Non-uniform surface temperature
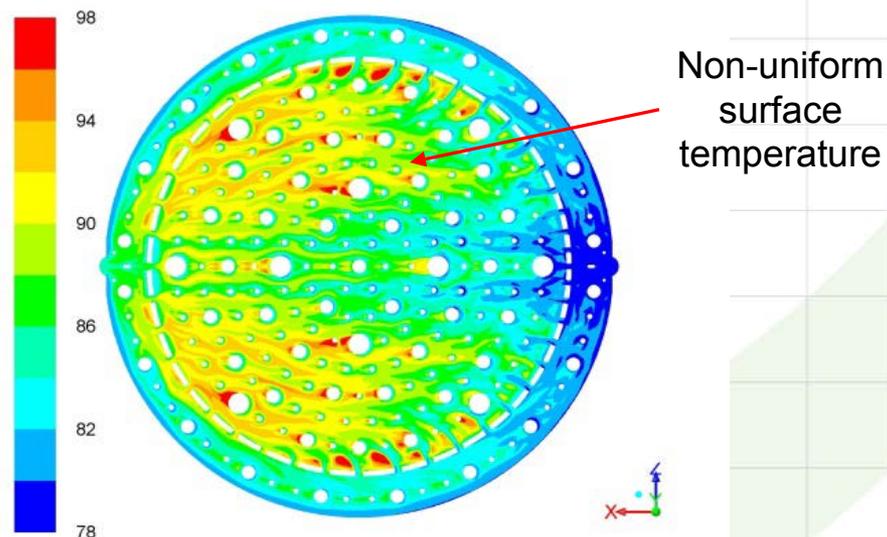
Phase II MATI. Contours of wall temperature for a mass flow rate of 3 g/s and an applied heat flux to the bottom wall of 17600 W/m^2. The flow is from right to left.

➢ CFD simulations were performed on the selected designs to compare flow distribution and pressure drop.
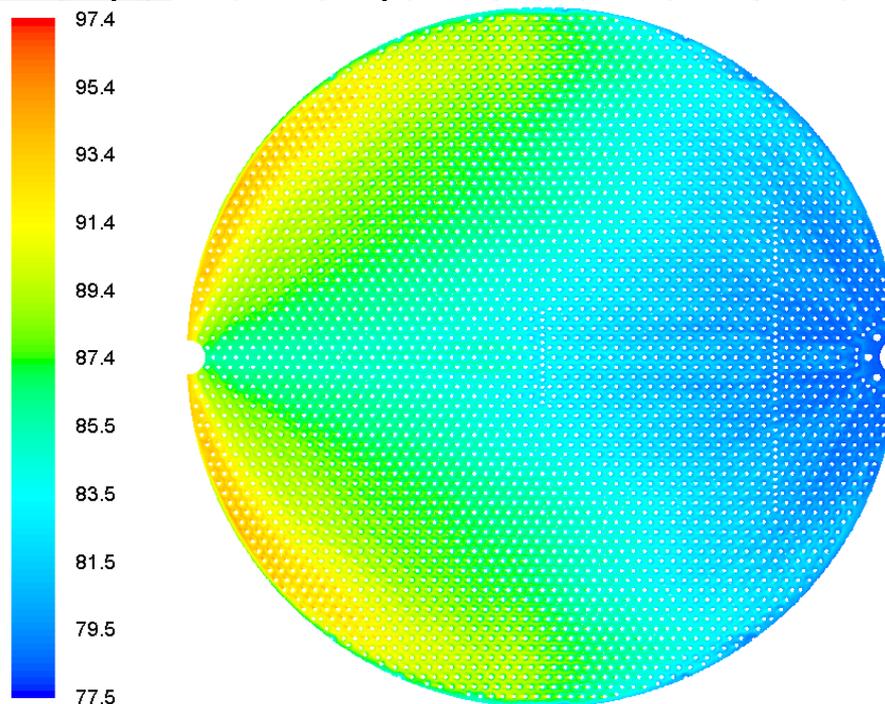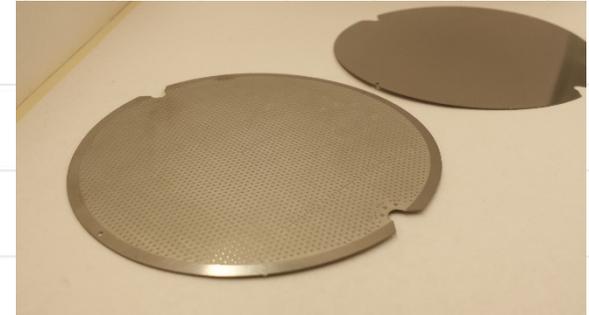

Contour maps of wall temperature (K) within the design # 4 of heat sink for a mass flow rate of 6.9 g/s and an applied heat flux to both top and bottom walls of 11268 W/m² with system pressure of 8 bar. 10 million cells were used in this simulation.
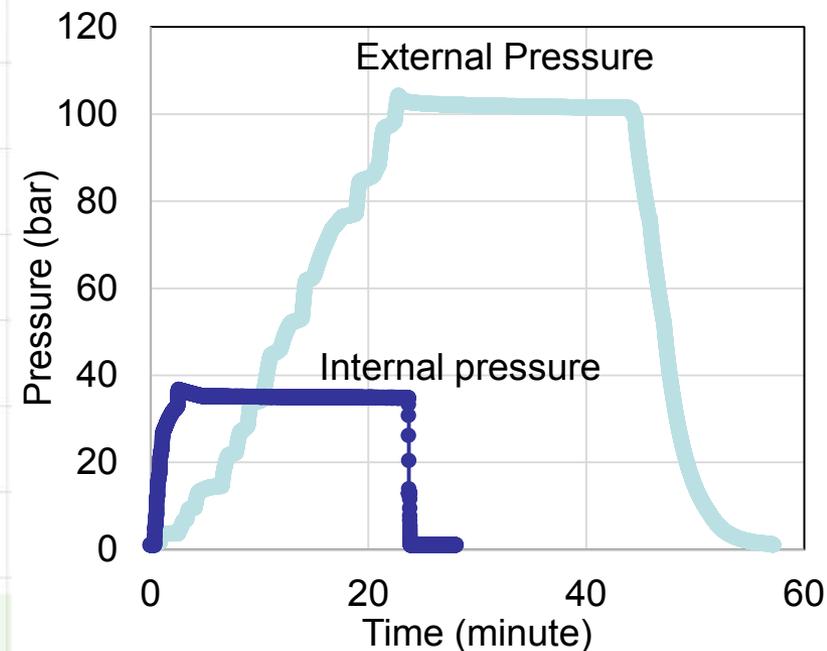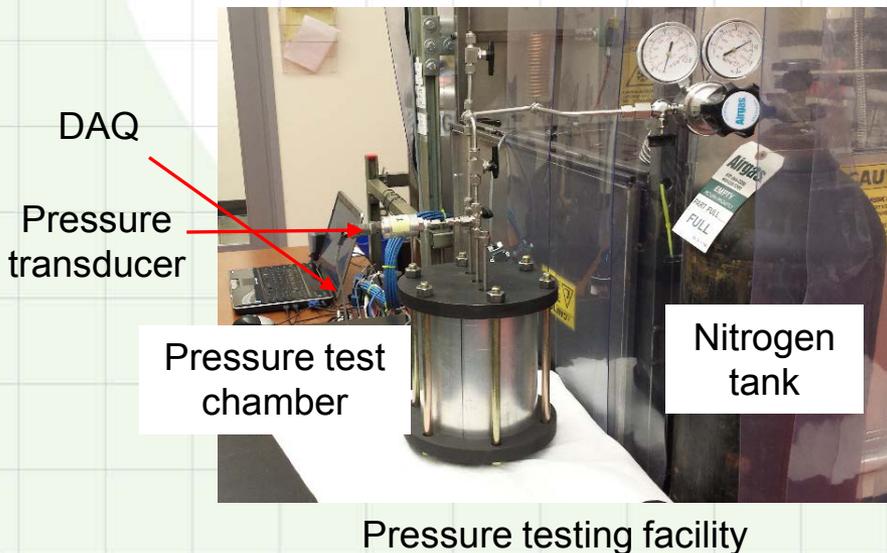
➢ The fluid flow conditions such as mass flow rate, heat flux, and system pressure were defined according to the operational conditions that MATI prototype would have based on 3 minutes charging time.

Barriers A and E – MATI Cooling Plate Design, Pressure Testing

- The finalized design was fabricated on 10 cm stainless steel 316 shims by chemical etching process by an external vendor.
- Plates were diffusion bonded and pressure tested according to the defined constraints.
- The profilamtery scan comparison on the surface of plates before and after each pressure test, showed neither deformation nor leakage.
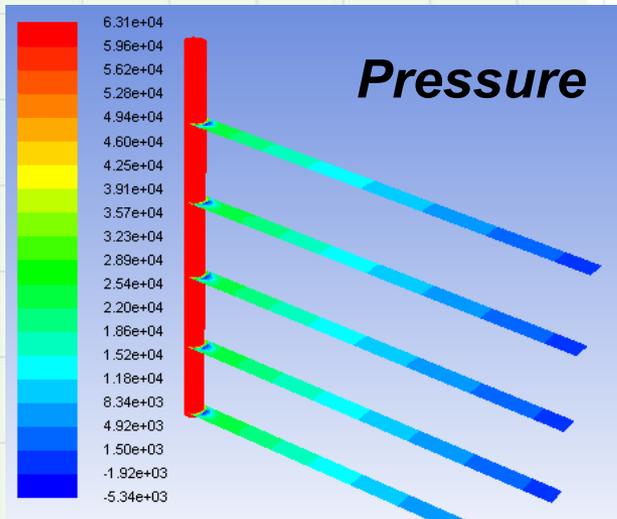

Chemically etched plates before diffusion bonding

DAQ

Pressure transducer

Pressure test chamber

Nitrogen tank

Pressure testing facility

External Pressure

Internal pressure

# Barriers A and E – Header Flow Simulation

Pressure

- The estimated pressure drop inside the header tube is insignificant
- The pressure drop across the small slot into the MATI cooling plate is on the order of 0.2 bars
- The flow inside the header is modeled using standard k-ε turbulence model with standard wall function
- Issues with brazing cooling plates to the header

Pressure

velocity
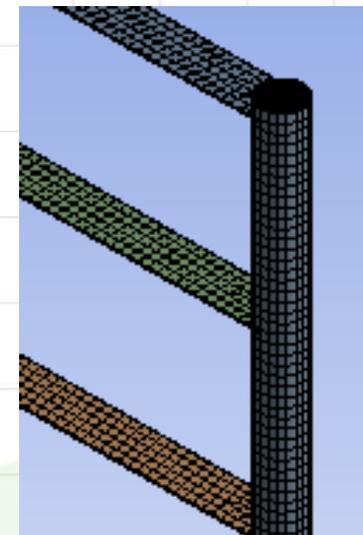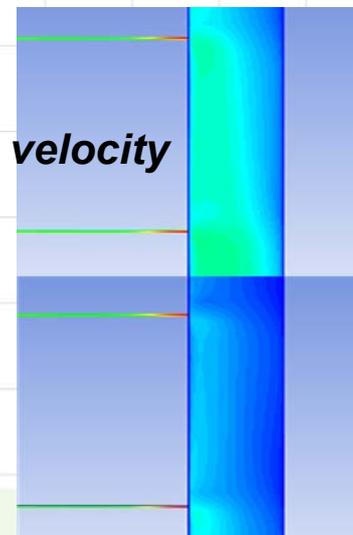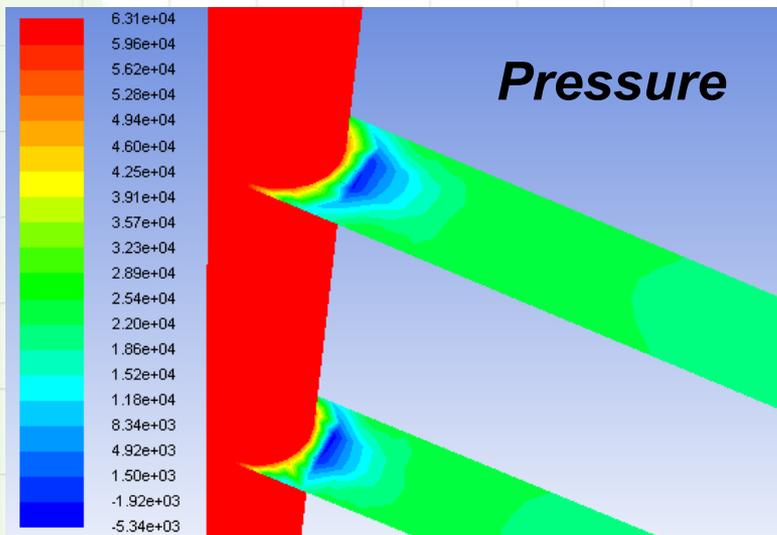
Barriers A and E – Prototype Unit Cell

- *The half-pucks show good strength at density of 0.4 g/cc and good geometric tolerance*
- *No damages on the half-pucks during initial simulated assembly between cooling plates*
- *Includes proprietary design features that push the half-pucks against the plates, reducing contact thermal resistance*
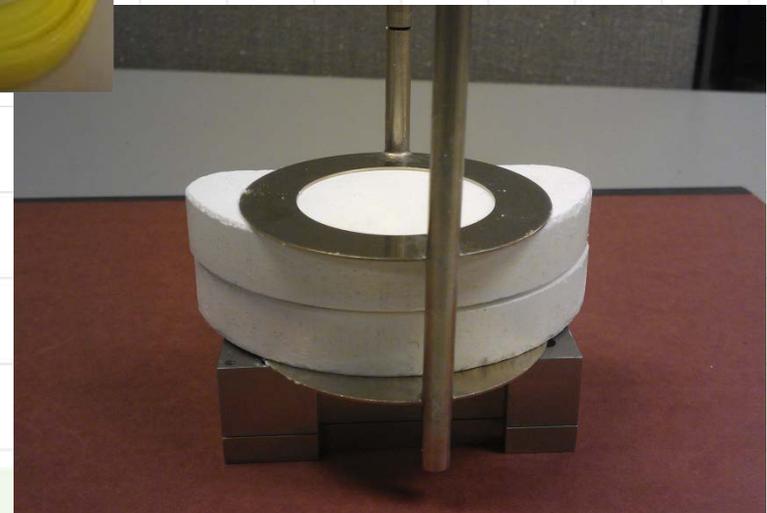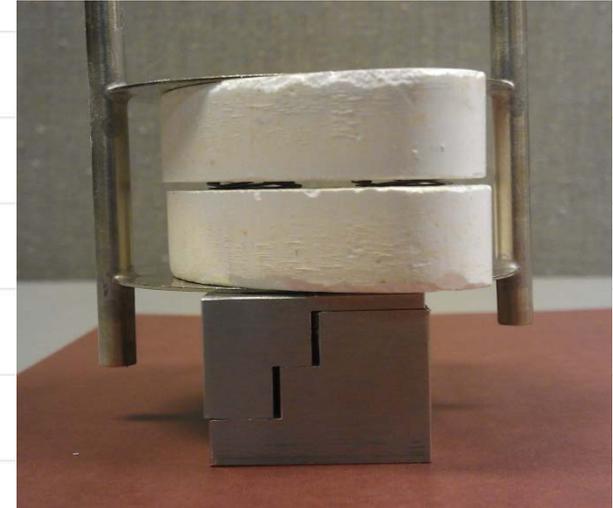
- ## Key Design Requirements
  - MATI must be fabricated and constructed to withstand 100 bars of external H_2 gas pressure and 35 bars of internal N_2 gas pressure with no detrimental signs of stress sagging or delamination
  - Aluminum pressure vessel must withstand 100 bars H_2 pressure
  - Constructed system must withstand temperatures of 130^oC required for activation and temperatures as low as 77 K for experimentation
  - Capable of running LN2 cooling during charging and heated N_2 gas for discharging experiments
  - Generate high quality data for model validation
    - Utilize high quality instruments and calibration to provide a ≤5% experimental error
  - Demonstrate safe and controllable operation

Barriers A and E – OSU Acceptance Testing

- Testing will serve to verify functionality of installed MATI inside Phase 3 pressure vessel

- Stepped testing procedure will be utilized to ensure safety
  - First full testing cycles will be performed at 20 bars storage pressure
  - Upon safe completion of cycled tests, storage pressure will be increased by an additional 20 bars and repeated up to 60 bars

- At 60 bar storage pressure, OSU will test:
  - Multiple LN2 flow rates
  - Multiple H_2 feed rates
  - Multiple Discharging rates

- Each experimental cycle will be performed at least 3 times at each operating pressure (up to 60) at OSU

- SRNL is planning to test up to 100 bars following the completion of rigorous cycling experiments up to 60 bars at both OSU and SRNL
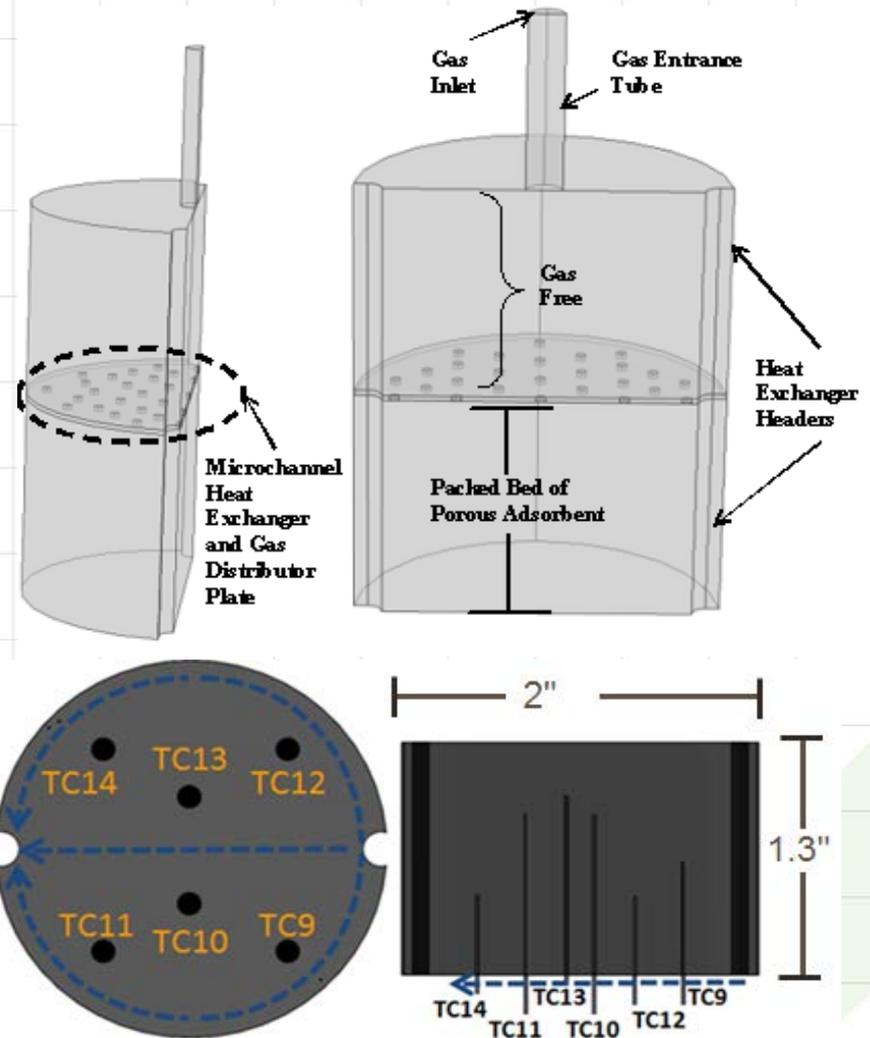
Barriers A and E - Modeling

- 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

- All eight domains participate in heat transfer simulation

- Domains 2, 4, 6, 7, and 8 participate in free and porous media flow solution of momentum transport and mass balance

- When a single unit cell simulation is optimized and meeting performance criteria, a larger full system simulation will be performed

1. *Steel vessel*
2. *H_2 annular region*
3. *Header tube*
4. *LN2 fluid in header*
5. *MATI Plate*
6. *LN2 fluid in plate*
7. *MOF-5 bed*
8. *H_2 distribution layer*
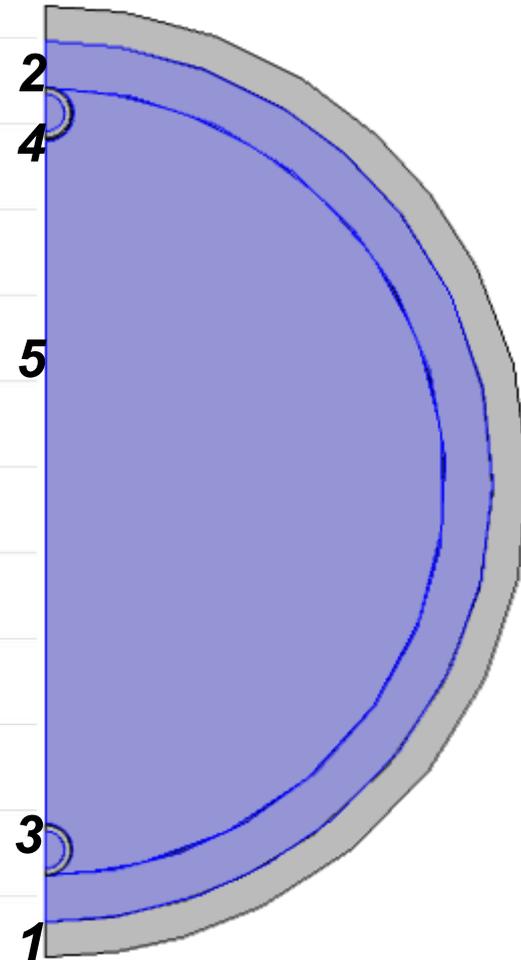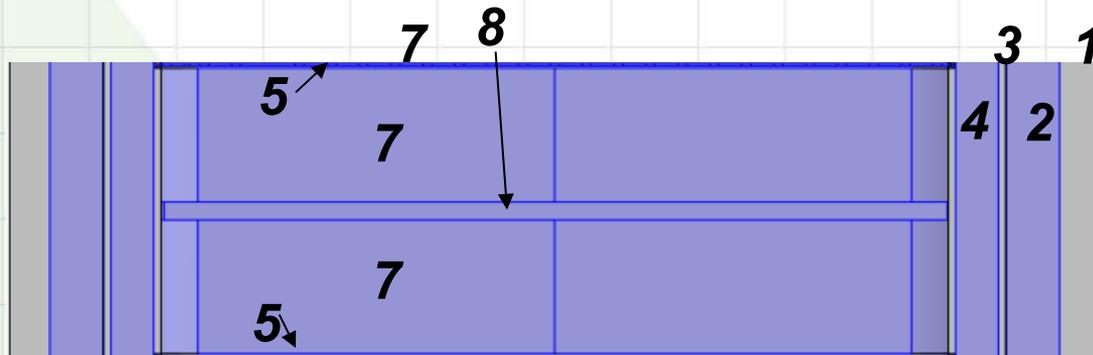
Response to Previous Year Reviewer Recommendations

- **"Experimental System tests need to demonstrate sustainable/robust microchannel device performance"** – While long term testing is not part of the centers mission, we will be conducting multiple performance tests on the MATI prototype as part of the acceptance testing at OSU and performance testing at SRNL.

- **"Verify conceptual design for MATI via experiments to demonstrate no internal or external leaks"** - While long term reliability testing is not part of the centers mission, limited investigating the durability of the pucks, cooling plates, headers and header/cooling plate connections as part of the acceptance testing at OSU and separate effects testing at OSU on the cooling plate and header/cooling plate connections

- **"Can the concepts and test assembly be extended to other densified materials"** – The focus on simulation validation is intended to provide a validated model that can be applied to other densified media assuming the densified media properties are available. The experimental apparatus can be used with any media that can be densified into pucks. The concept can tolerate some degree of puck degradation over time.

Remaining Challenges and Barriers

- Complete assembly and acceptance testing of MATI including a fabrication scheme for integrating the cooling plates and header;

- Simulation Validation;

- Demonstrate conduction enhancement;

Proposed FY 2015 Future Work

- Reduce Size and Weight of Storage and Improve Charge and Discharge Rates – **Modular Adsorption Tank Insert Development**

    – Complete assembly of MATI prototype

    – Complete acceptance testing of prototype

    – Deliver prototype to SRNL for performance testing

    – Complete model validation

    – Complete acceptance testing using conduction enhanced pucks

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.

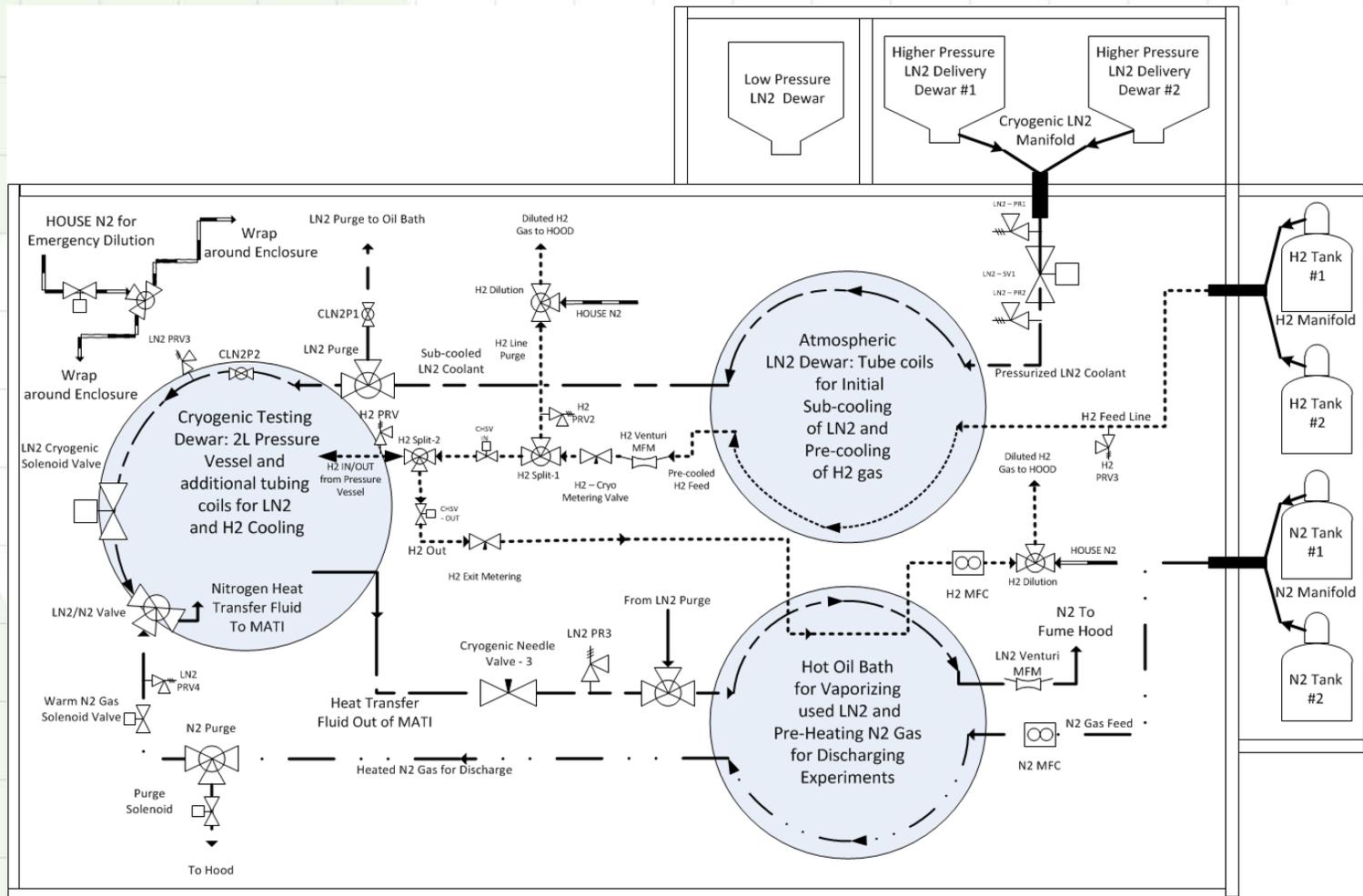- Developed simulation for code validation in collaboration with SRNL.

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 acceptance testing at OSU.
  - Conduct performance testing at SRNL to demonstrate our second smart goal.
  - Validate simulations against performance results.
- **Technical Accomplishments**:
  - Completed design of MATI prototype which meets smart goals requirements
    - Pressure vessel and plug in collaboration with Hexagon Lincoln
    - Media puck in collaboration with Ford and University of Michigan
    - Design of cooling plates
    - Design of headers
  - Completed center design review of MATI prototype, test apparatus design and test plan
  - Completed design of modified test apparatus for MATI acceptance testing
  - In collaboration with SRNL, completed modifications to simulation to allow model validation
- **Collaboration**: Member of HSECoE team.
- **Proposed Future Research**:
  - Complete assembly of MATI prototype
  - Complete acceptance testing of prototype
  - Deliver prototype to SRNL for performance testing
  - Complete model validation
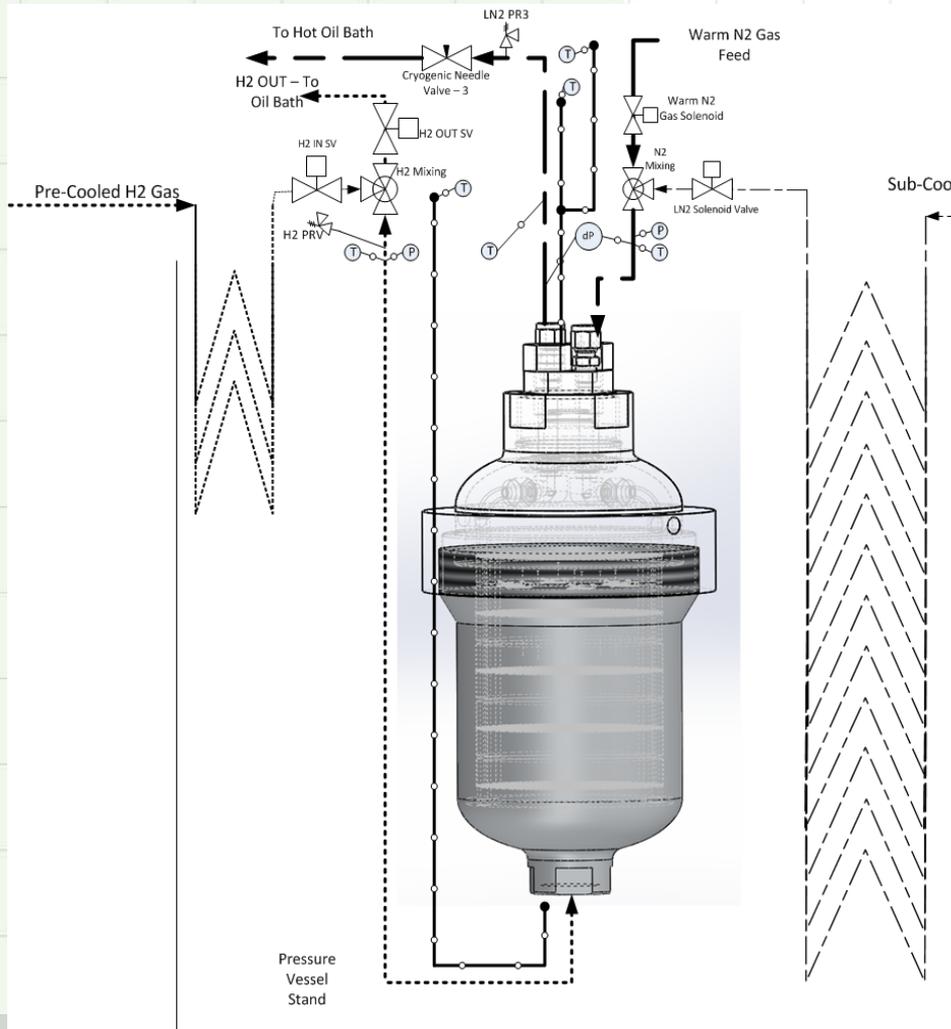 - Complete acceptance testing using conduction enhanced pucks
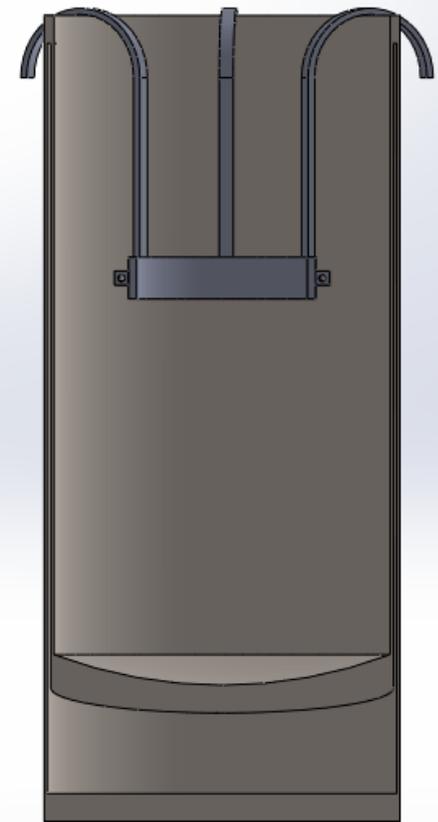
Supplemental Slides

Two-piece hanging stand to secure pressure vessel in place

MATI Cooling Plate (on top)

Axially aligned Fin and Tube
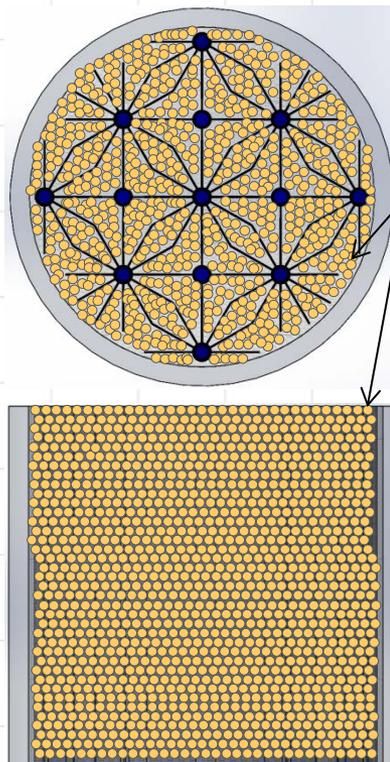
Circumferential Cooling tubes and Fins

Packed MOF-5 pellets or particles required

Al Tank

Compressed Monolithic MOF-5 Beds 3.0 cm between cooling plates

MATI Cooling Plates
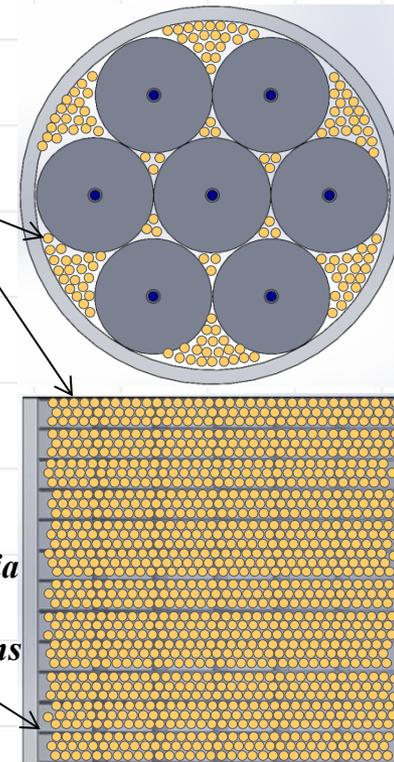
Circumferential Cooling Tubes and Fins (1 cm fin spacing)

-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**
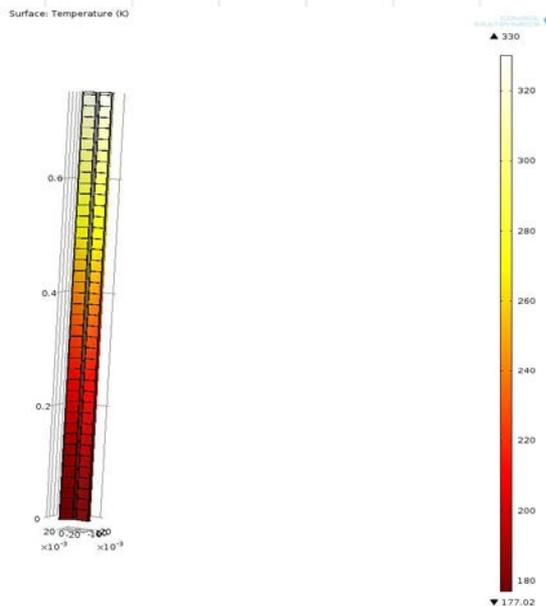
Simulation of Axial Fin Tube (SRNL)

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

  o **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**

  o **8% of the volume is metal or flow path and is unavailable for media, in the MATI, 5% is metal or flow path**

  o **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