Magnetocaloric Cryogenic System for High Efficiency Air Separation

(aka "Magnetocaloric Oxygen Liquefaction System"- MOLS)

NETL FY20-FY21 Gasification
Annual Project Review

FWP-73143

Pacific Northwest National Laboratory

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Enable gasification of high-carbon materials with O_2 for syngas and easier CO_2 capture

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- Overall Strategic Objective of this project is to develop efficient, small-scale, cryogenic air separation units (ASU)
  - Produce 10-90 metric ton/day of LOX near gasification plants
    - ✓ ~50-450 metric ton/day of liquid air supply into distillation columns
  - Use magnetocaloric liquefier (MCL) for air to replace traditional turbo-Brayton cycle air liquefier
    - ✓ Active Magnetic Regenerator Design (AMRL) to increase ASU energy efficiency by ~50%
    - ✓ Leverage ~6 years of experience with AMRL prototypes at PNNL (+ Ames Lab)
  - Achieve modular designs for "make local, use local" applications
    - ✓ Learn how to cost-effectively scale to ~100 times smaller than large ASU plants
    - ✓ Techno-economic analyses indicate flexible operation and excellent scaling of MCLs achieved by parallel connection of number of parallel optimized modules

Magnetocaloric Cryogenic System for High Efficiency Air Separation

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Timeline:

- Project Start Date: 12/1/2018
  - Delayed in FY19 due to new magnet
  - Delayed in FY20 due to pandemic
- Project End Date: 9/30/2021*

  * FWP-FY21 pending by DOE/FE/NETL

Budget:

- FY21-NCTE from FY20 to 9/30/2021
- Total DOE funds received to date: $2.0 MM

Technology needs addressed:

- Small-scale, efficient cryogenic ASU
  - 10-90 tonne/day of LOX
  - Efficient magnetic liquefier for air
  - Microchannel air distillation column
  - Scalable with parallel modules

Technical Background

Highly efficient MOLS design focuses on three key subsystems of MCLs

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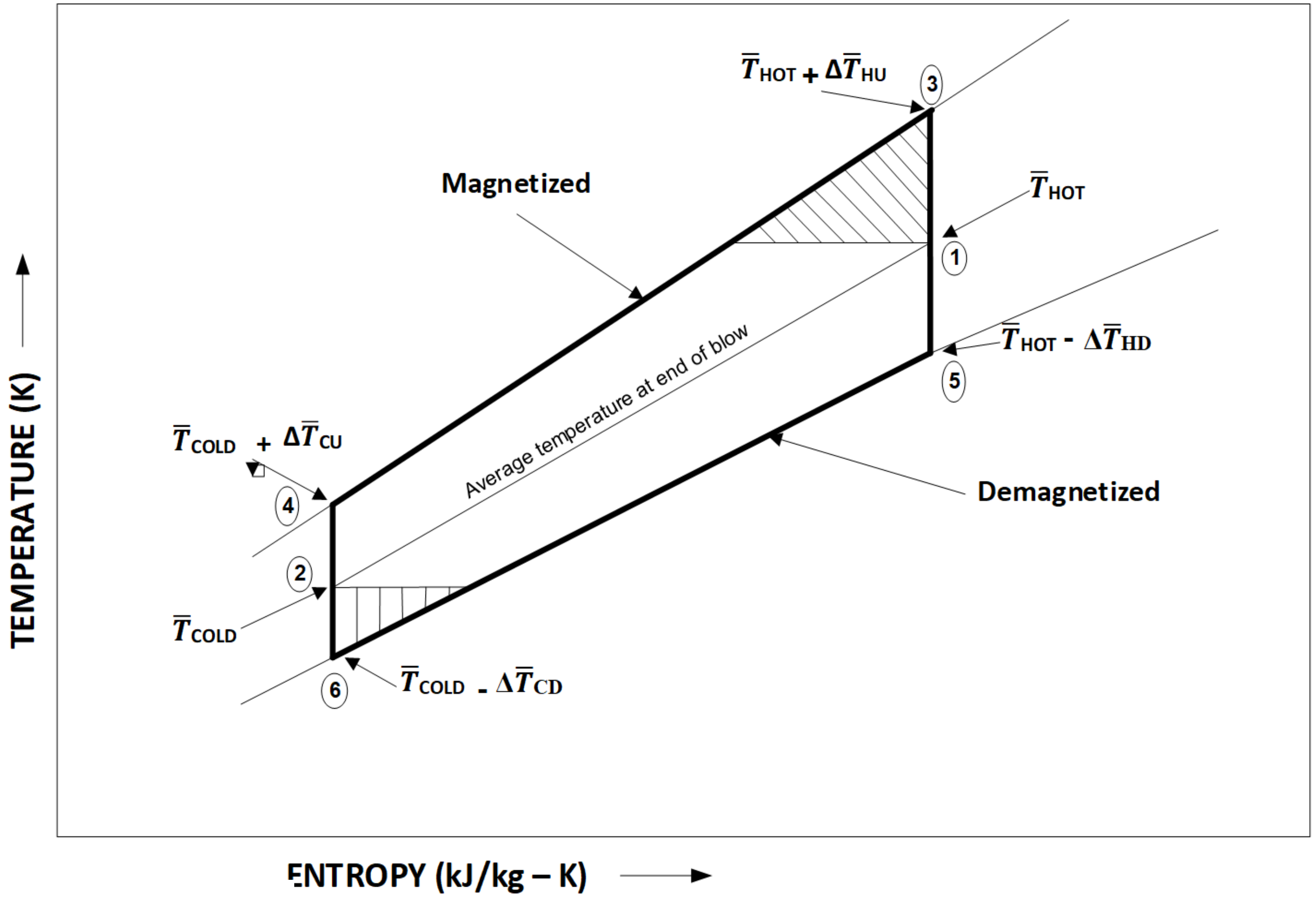
- Core subsystem – high-performance magnetic regenerators
- Plus - heat transfer fluid flows to efficiently couple refrigerants to loads and sinks in AMR cycle
- Plus – High field NbTi superconducting magnets for large ΔB during AMR cycle
- Integrate all subsystems to execute AMR cycle illustrated in T-S diagram
- Efficiency measured by figure of merit (FOM)
  - FOM = Ratio of ideal to real work rates (see eqn.)
  - Regenerators designed to minimize all sources of ΔS_{IRR} and auxiliary work rate input
  - FOM ~0.60 achievable



$$\dot{W}_{real_{Layer}} = \left(\dot{Q}_{CHEX} + \dot{Q}_{LC} + \dot{Q}_{Para}\right)\left(\frac{T_H}{T_C} - 1\right) + \frac{T_H \int_{T_C}^{T_H} \Delta\dot{S}_{IRR} dT}{\int_{T_C}^{T_H} dT}$$

MCL prototype test apparatus was upgraded with new superconducting magnet subsystem

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- New magnet system arrived 9/25/20; first lab run to 6.5 T -10/5/20
- Assembled/leak checked dual 5-layer magnetic regenerators–11/1/20
- Installed parallel pump to double flow capacity of helium gas system–11/15/20
- Tested pump for diversion flow system using passive ss regenerator–11/30/20
- Installed linear reciprocating 16” drive with controller on cold box-12/21/20
- LabVIEW for DAQ - 48 sensors/inputs
- Initial runs: mid-January 2021; encountered force balance issues; COMSOL-AC/DC modeling to guide fix

Measured magnetic forces for 5-layer dual regenerators differed from initial expectations

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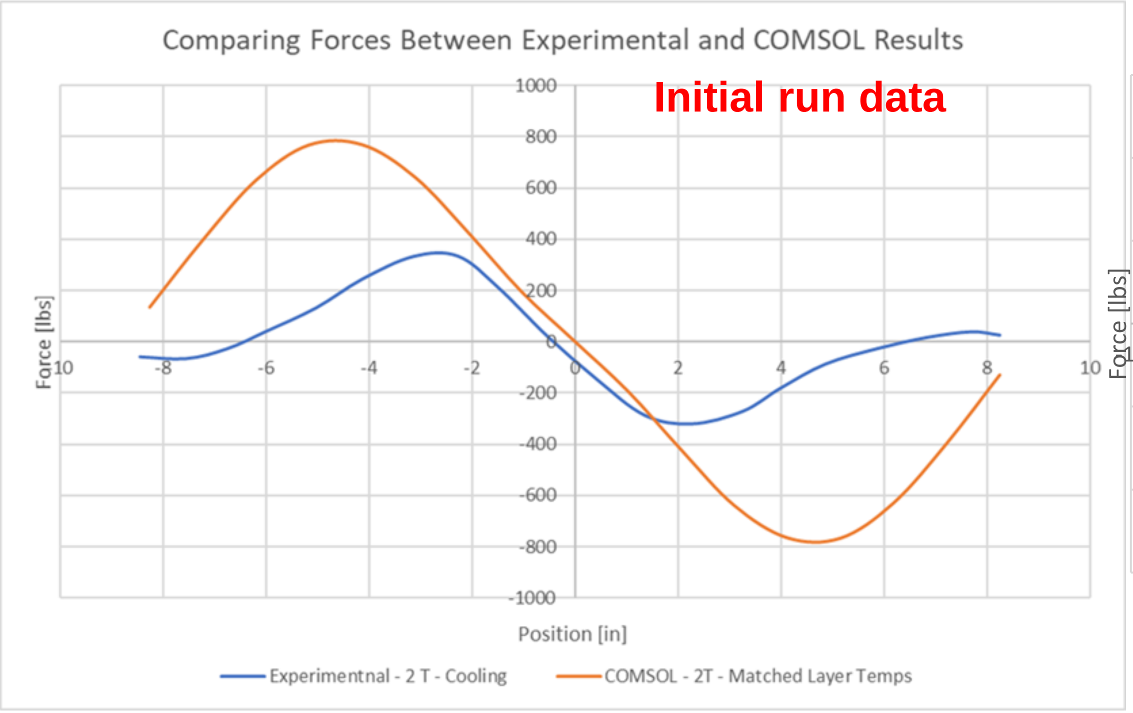

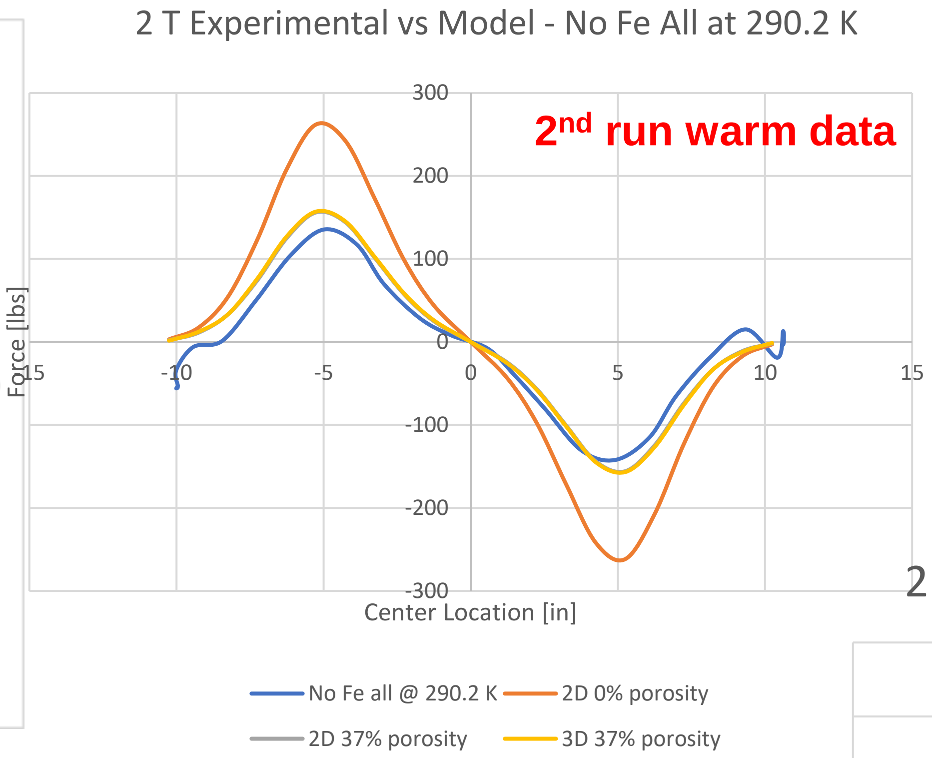

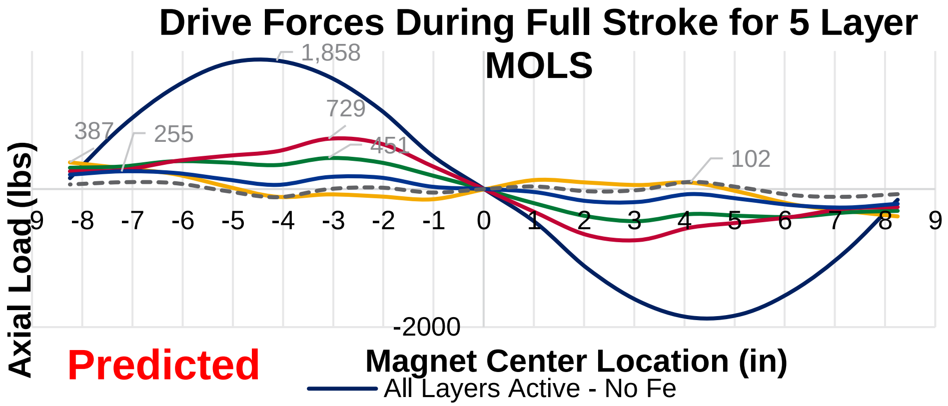

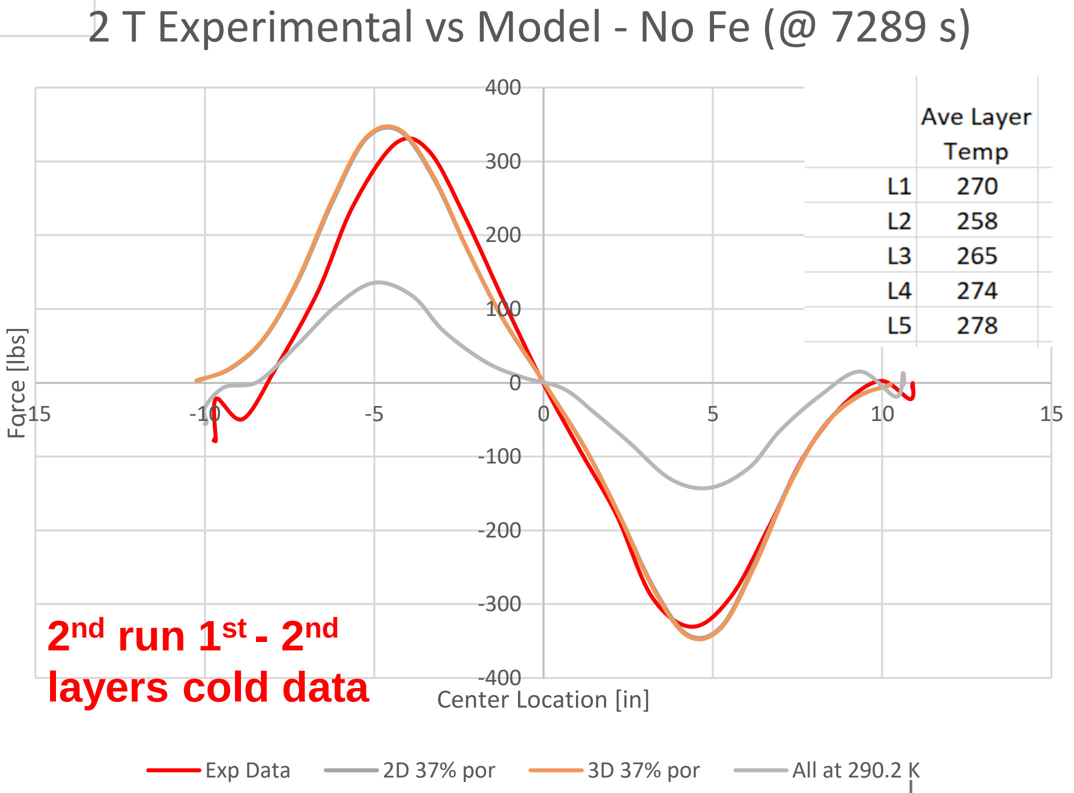

- Used COMSOL Multiphysics with AC/DC module (2-D & 3-D)
- Plot 1 – force differs in magnitude and position – error in locations of field gradients and Fe pieces
- Corrected magnet geometry and magnetic materials inputs for COMSOL; $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M}\ (\mathbf{T}, \mathbf{B}))$ for agreement between 2-D/3-D
- Experiments at end of March showed great improvement
- $\phi = \int \mathbf{B}*\mathbf{A}$ =constant in s/c magnet requires **H** to change as **M** changes - causes heating in magnet so cool down challenging.

HTF flow rates per layer vary with the mass of refrigerant in each layer

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- Mass flow of helium should match MCE-cooling power/layer (no flow, no cooling)
  - $\dot{Q}_{He} = \dot{m}_{He} C_{pHe} \Delta T_{He}^{ave}$ where ΔT_{He}^{ave} is ~equal to $\frac{\Delta T_{CD}(T_{COLD})}{2}$
  - $\dot{Q}_{He}$ is ~equal to $\dot{Q}_{C}(T_{COLD})_{AMR-REN}$ which determines main $\dot{m}_{He}$ flow
- He temperature out of demagnetized layers changes during flow steps of cycle are time dependent. For hot-to-cold flow:
  - $\left[T_{He}(t) = T_{COLD} - \left(\int_0^{t_{flow}} \Delta T_{Cold}(t)dt\right)\right]$
  - Too little $\dot{m}_{He}$ underblows $\dot{Q}_{C}(T_{COLD})$ – reduces cooling power of magnetic refrigerant
  - Too much $\dot{m}_{He}$ overblows $\dot{Q}_{C}(T_{COLD})$ – reduces cooling power and reduces efficiency
- MCL designs can have three different important HTF flows:
  - **Main flow** – energy balance - changes as refrigerant mass/layer changes
  - **Diversion flow** – adjusts main flows between adjacent layers
  - **Bypass flow** – adjust flow after coldest layer to balance flow for differences in thermal mass of refrigerant – unique feature that can increase efficiency a lot

Diversion flow matches HTF in layers
e.g., 50% of hot-to-cold flow diverted at ~160 K

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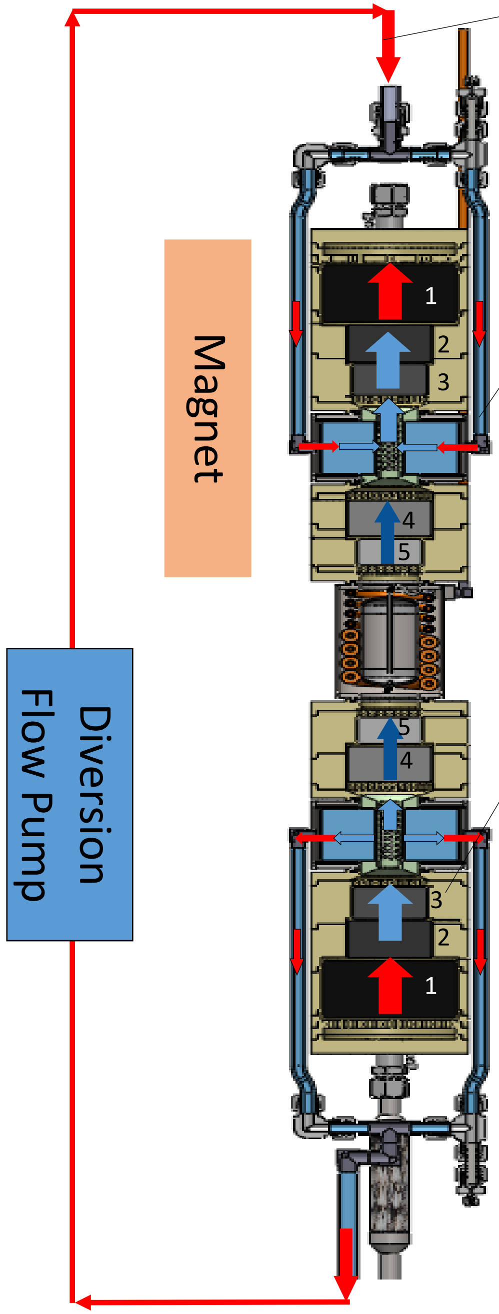
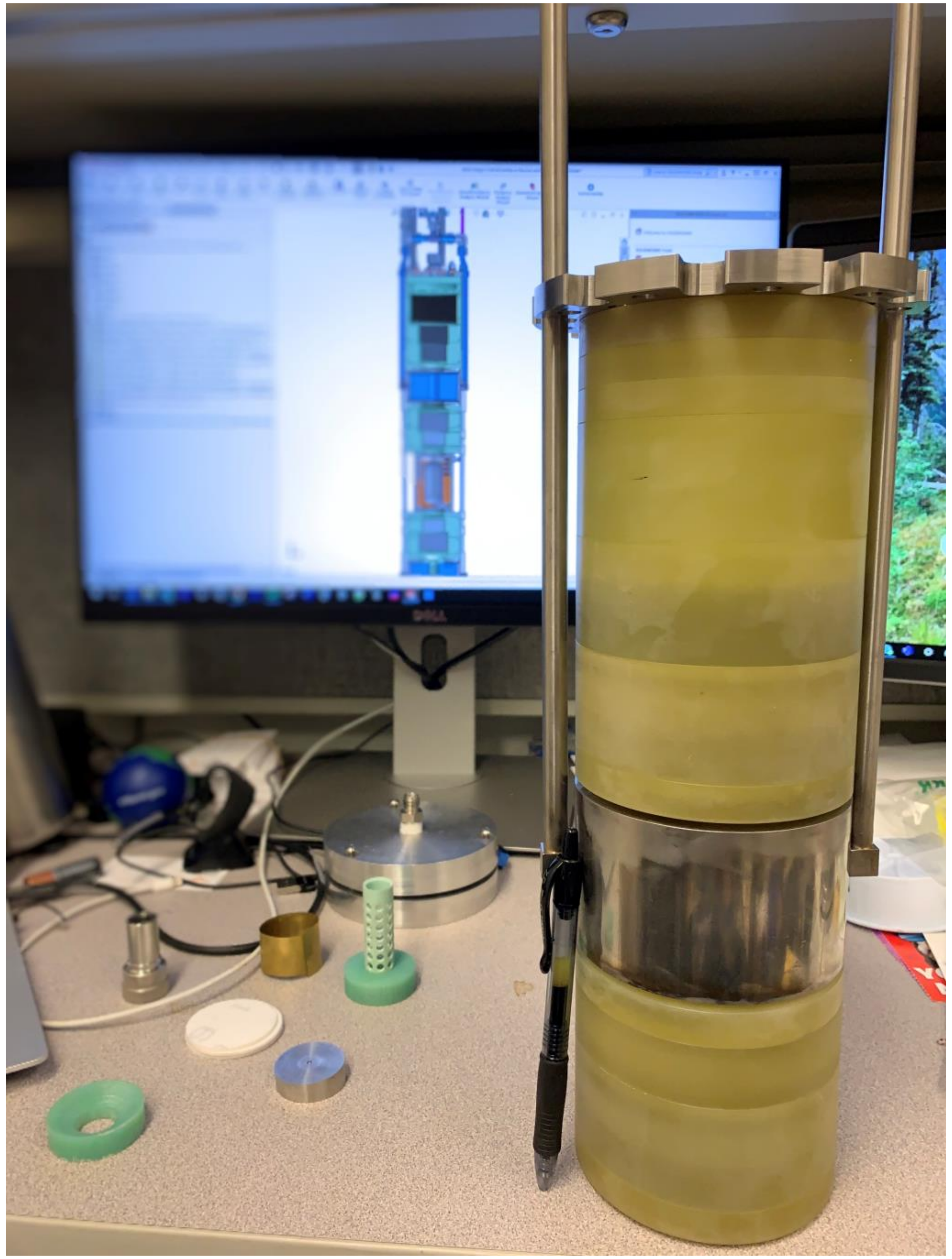


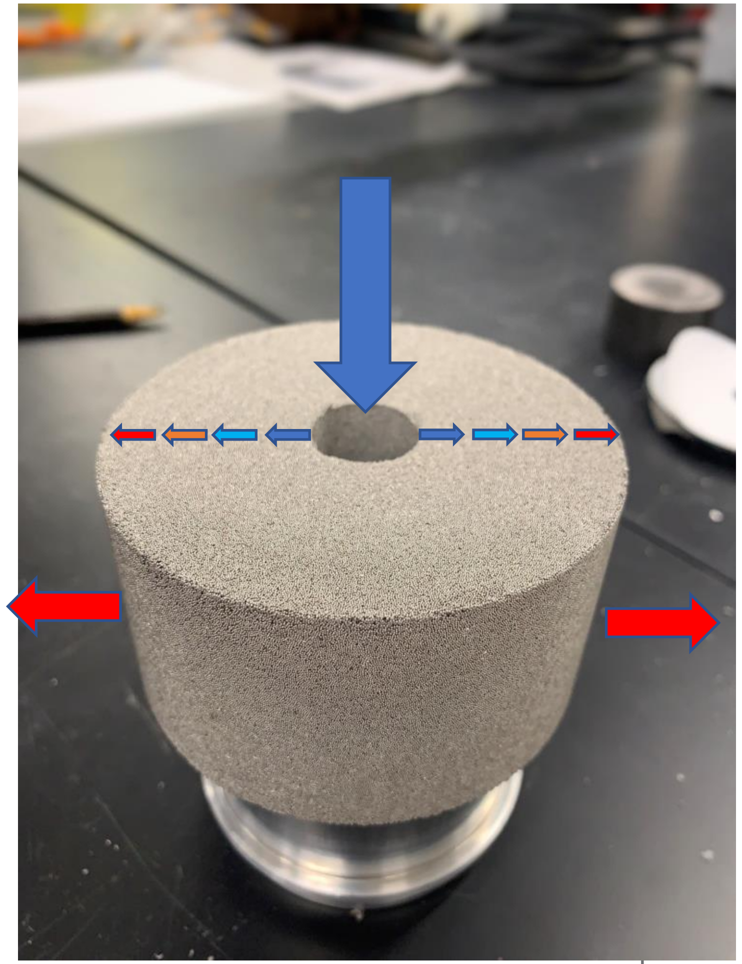
Diversion flow path

50% 160 K HTF added to C-to-H flow from layers 4-5 to reject heat

Hot-to-Cold HTF flow for layers 1-3 50% greater than for layers 4-5.



OD (in)	ID (in)	Height (in)	Mass (kg)
3.50	0.75	1.88	1.43



A 200 psia liquid propane heat transfer fluid subsystem is being built and tested

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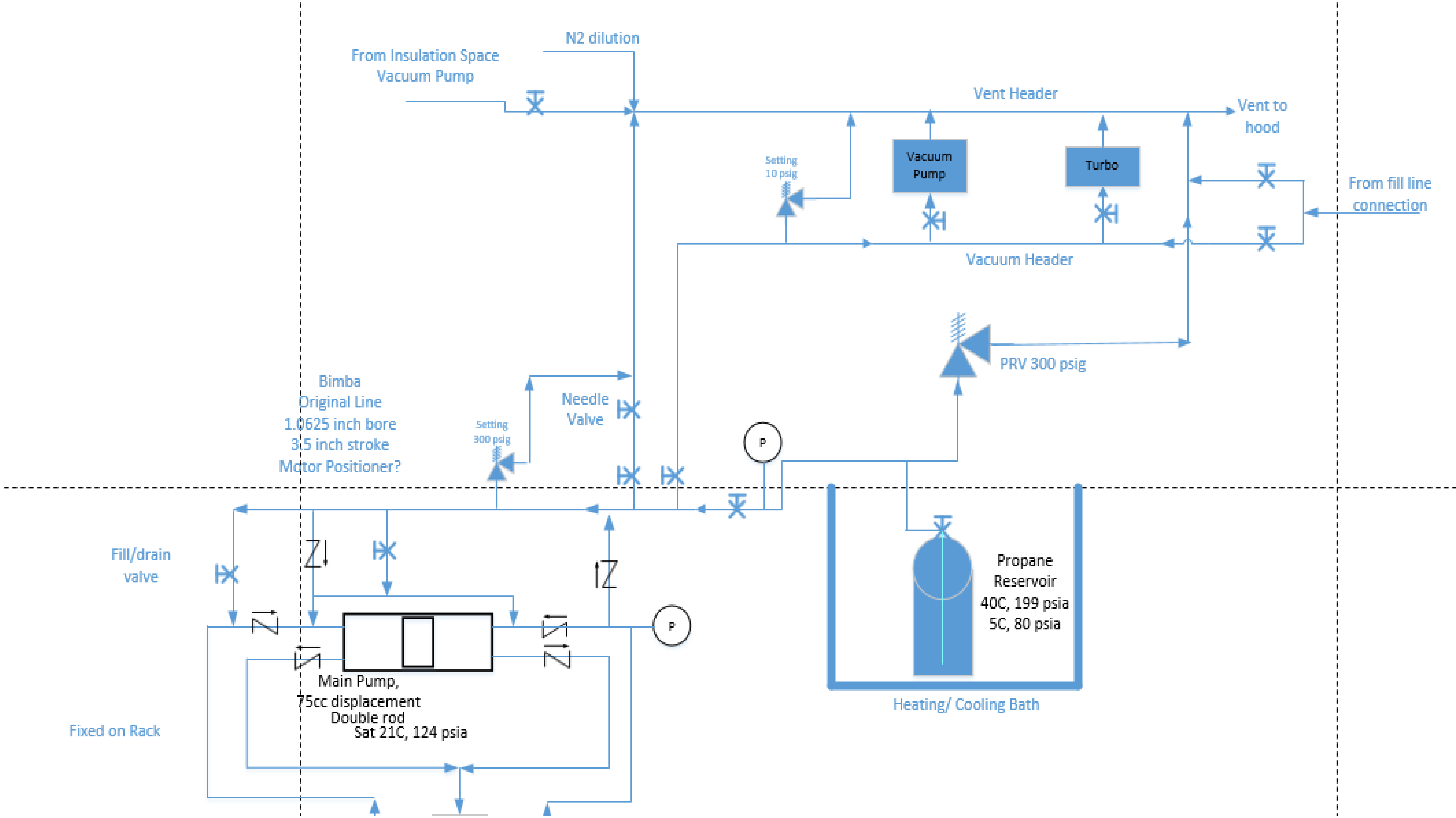
- Pressure drop of 400 psia He HTF is largest source of ΔS_{IRR} in 290 K to 100 K AMRs
  - Want HTF with higher density with low viscosity
  - Liquid propane at ~200 psia is excellent choice
  - Remains saturated liquid over entire T span
  - Freezes at ~90 K
- Designed propane pump per schematic to test



Developed a lab-size micro-channel distillation (MCD) column to produce LOX from liquid air

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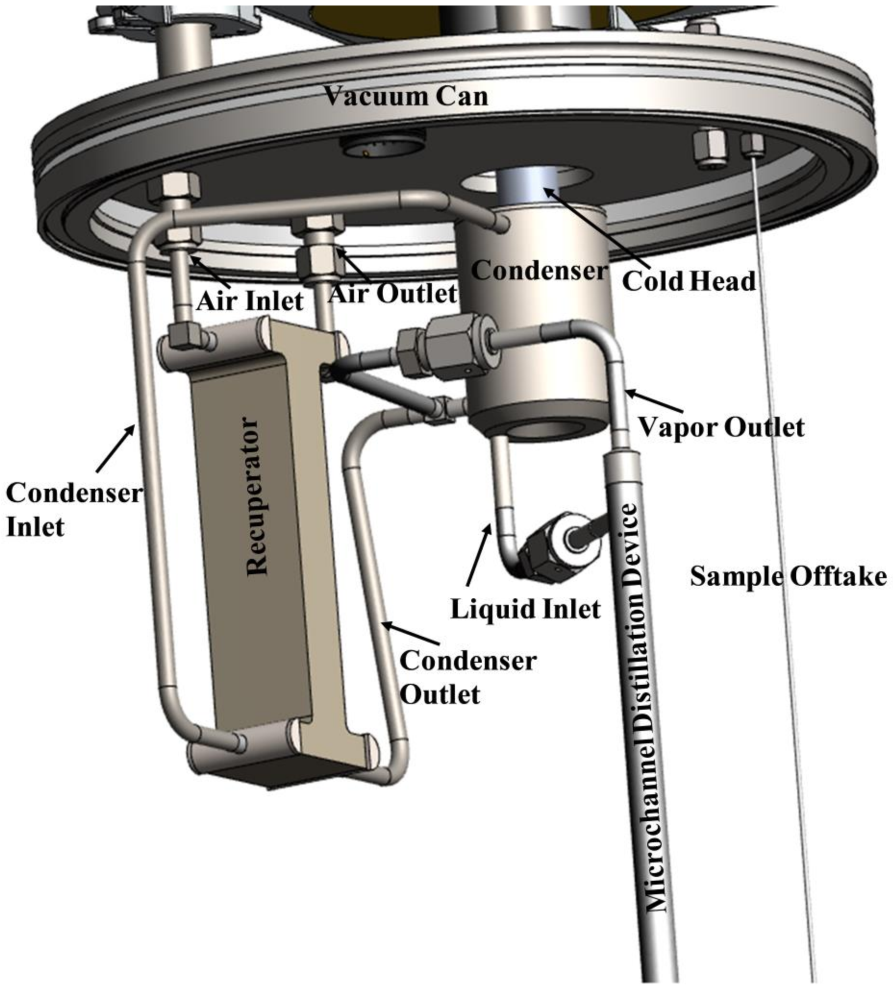
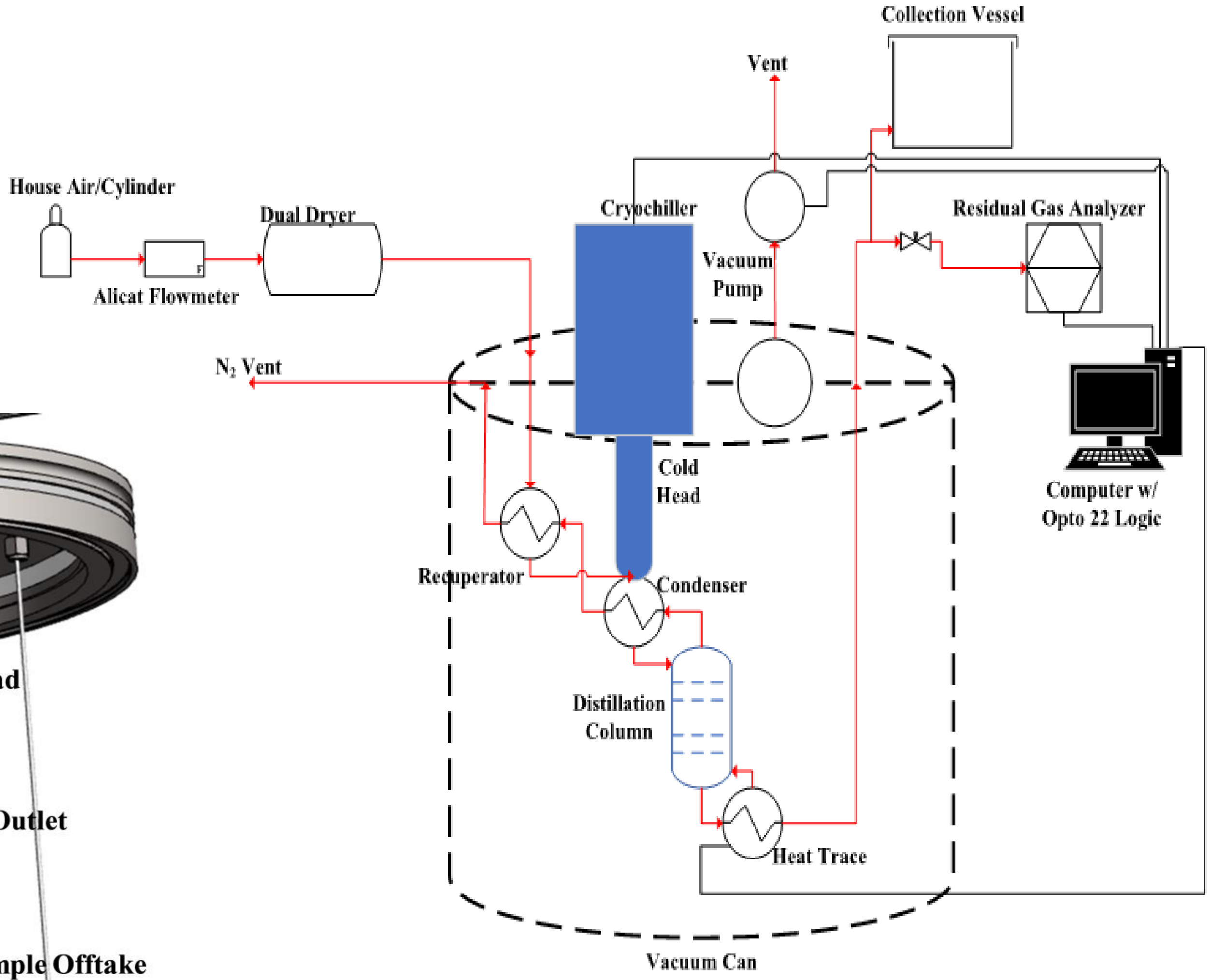
- Continuous distillation of O_2 from cold dry air modeled by using CHEMCAD simulations
- Process intensification of MCD reduces column length from ~20 cm to ~4 cm to perform this separation.
- Results indicate that oxygen is enriched to > 98 mol% in a compact distillation column
- Demonstrated separation of LOX from liquid air





Results for distillation of air to make LOX with MCD after calibration of cryocooler

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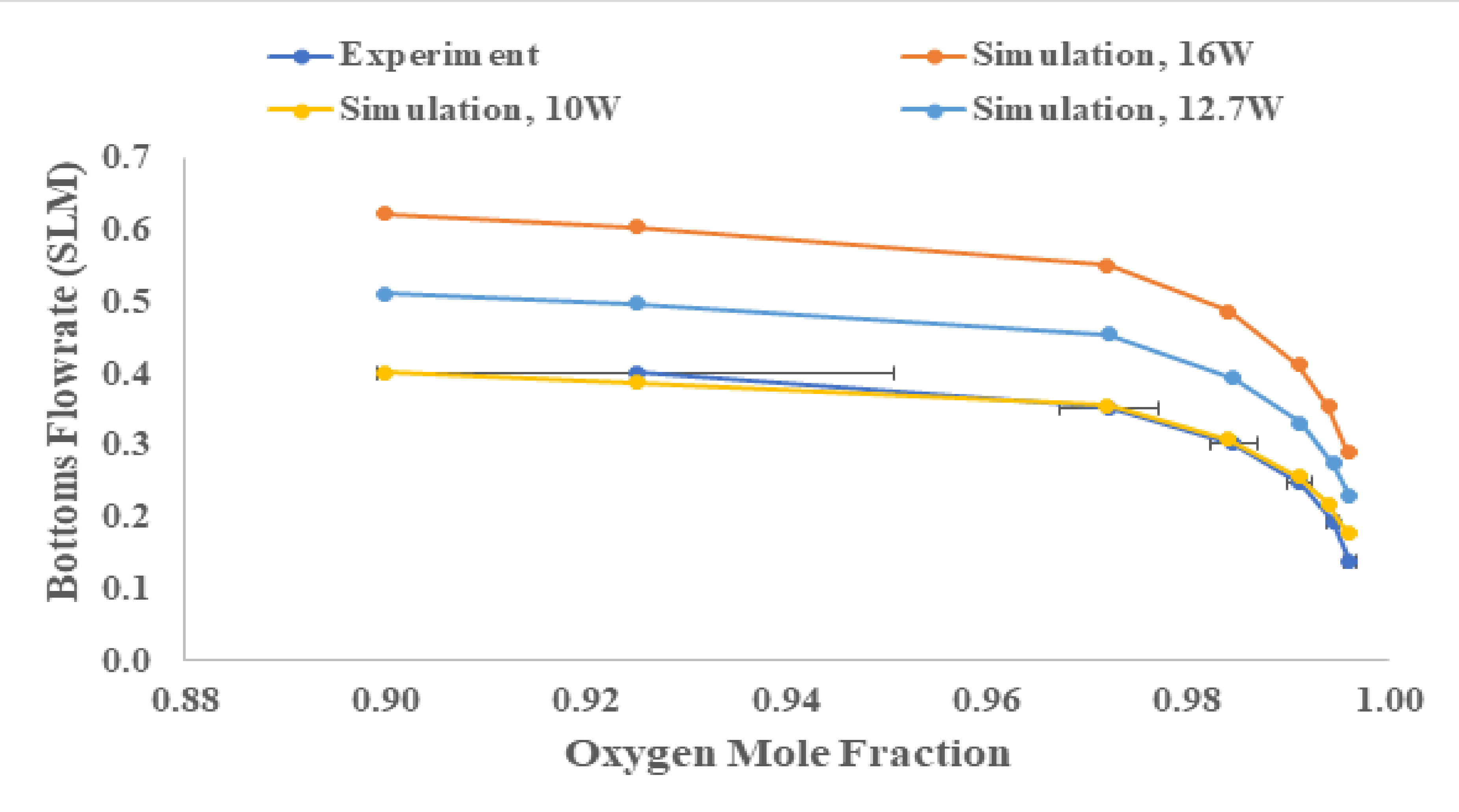
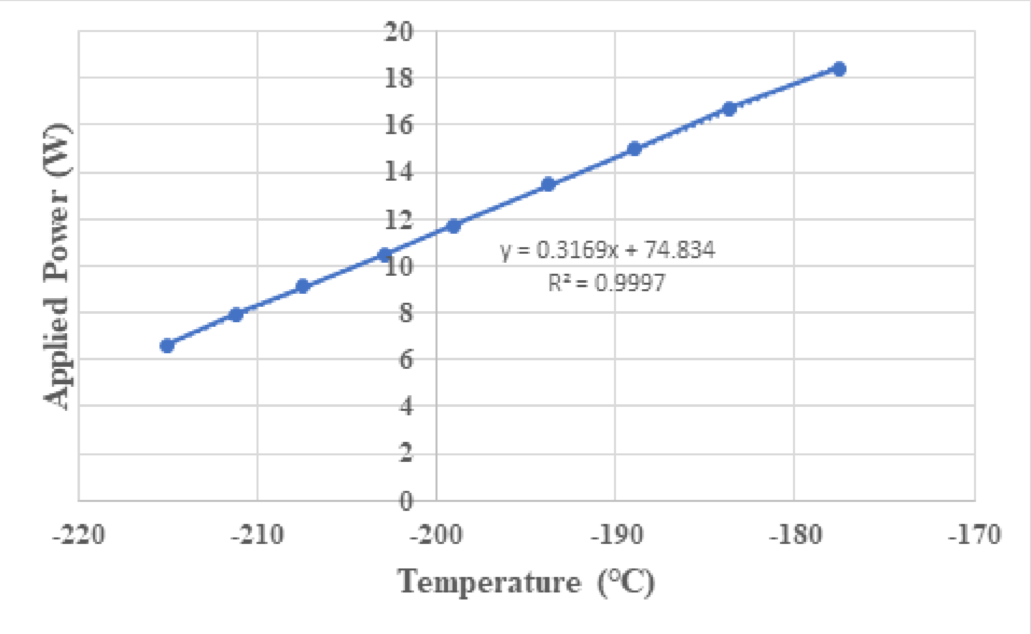
- Test results are excellent!
- ChemCAD model shows LOX flow rate vs. purity with 12.7 W of cooling power at inlet T from cryocooler
- Independent calculations of parasitic heat leak gave ~2.5 W
- Test runs confirmed it is possible to reduce size of MCD





Status of FY20 Tasks includes NCTE to 9/30/21 due to COVID

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- ❖ **Task 1:** Testing 5-layer dual regenerator liquefier to demonstrate cooling to ~100 K and liquefaction of ~1 kg/day of liquid air.
- ❖ **Task 2:** Analyze test results, compare with performance models to validate our simulation models, and identify required developments.
- ✓ **Task 3:** Model and implement magnetic force balance for 5-layer dual regenerators in steady state; studying effects of magnet heating from unbalance during start up
- ✓ **Task 4:** Design, build, and test HTF system to circulate pressurized ~200 psia liquid propane as liquid heat transfer medium
- ✓ **Task 5:** Model, build and test diversion flow for 2- and 3 -layer regenerators
- ✓ **Task 6:** Model, assemble and demonstrate a lab-scale microchannel distillation device for LOX production from liquid air
- ❖ **Task 7:** Project management to achieve milestones and deliverables, write progress reports, present reviews, and communicate challenges and surprises as they occur.

Concluding remarks and potential future work

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- FY20 Deliverables on target to be completed
  - Demonstrate magnetocaloric liquefier that cools to ~100 K and liquefies air at a rate of 1 kg/day
  - Analyze 5-layer dual regenerator performance under different He mass flow rates, different frequencies, and different magnetic fields
  - Validate performance models
  - Finish testing liquid propane pump at near room temperature
- Next steps FWP-FY21 pending
  - Address External Peer Review feedback
  - Design simpler, multi-stage MCL integrated with a MCD into engineering-scale ASU
  - Perform Techno-Economic Analysis of modular ASU as function of scale
  - Test propane as HTF in the MOLS prototype
  - Develop collaborative partners who are interested in cost sharing and licensing ASU IP.
  - Write paper describing results

Thank you to the project team

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- DOE- Fossil Energy/Carbon Capture
  - Venkat Venkataraman
  - David Lyons
- DOE- Fuel Cell Technology Office
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- The Team
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  - Mike Powell
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  - John Barclay
  - Jamie Holladay