### Magnetocaloric Cryogenic System for High Efficiency Air Separation

### (aka "Magnetocaloric Oxygen Liquefaction System"- MOLS)

FWP-73143 – Annual Review May 2, 2022 John.Barclay@pnnl.gov



### Distributed low-cost liquid oxygen enables more effective gasification of coal and biomass



- Industrial air separation units (ASU) produce O<sub>2</sub>, N<sub>2</sub>, Ar, He, and other noble gases
  - Cryogenic or non-cryogenic ASU provide different capacity and purity
  - Cryogenic ASU used for high-capacity rates and high product purity
  - Existing cryogenic ASU produce 1,000-2,000 tonne/day of LOX and 4,500-9,000
- Immediate need for more efficient and less expensive smaller-scale ASU for LOX
  - Project target is to develop ASU to produce 10-90 metric ton/day of low-cost LOX
- Objective of PNNL project is to develop efficient, small-scale, cost-effective cryogenic ASU by leveraging integration of two innovative PNNL technologies
  - Replace turbo-Brayton cycle air liquefiers with magnetocaloric liquefiers (MCL)
    - $\checkmark$  MCL designs can increase ASU energy efficiency by ~50% and capex by ~25%
  - Replace conventional dual distillation columns with microchannel distillers (MCD)
    - ✓ MCD designs can reduce ASU footprint by ~5 times
- This presentation summarizes progress made during the past year

## The entire MCL test apparatus was upgraded around improved 6.5 T magnet system



- Reciprocating dual multi-layer magnetic regenerators system
- Helium heat transfer system -
- Superconducting magnet in high-vacuum cold box and 4 K cryocooler
- Reciprocating drive mount assembly
- ~48 different T, P, flow, force, position, field sensors and linear drive interfaces
- LabVIEW DAQ and Control system to run entire apparatus
- First run with 5-layer MCL was in mid January.
- Most subsystems worked well, but several surprises encountered too.



#### **Key deliverable in SOW - Demonstrate an** efficient MCL prototype for LAIR for an ASU/LOX **Pacific Northwest**

- Unexpectedly had magnet quenches from excessive heating when dual regenerators moved axially in/out of high-B region!
  - Magnetic moment changes in dB/dz cause magnetic force imbalance
    - ✓ Magnetic flux conservation in s/c magnet causes flux-jump AC losses from  $\Delta$  H
- COMSOL modeling showed axial movement caused timedependent radial **B** changes in magnet
  - Time-dependent dB\*A induces emf in electrically conducting magnet form and coils
    - ✓ Emf causes micro currents which cause Joule heating in magnet

83.5

Force measurement experiments on different magnetic rod length validated hypothesis



0.25 Hz

2 000

- No Gap with Ends - No Gap No Ends - 9.5in Gap with Ends 9.5in Gap No Ends - 19in Gap with Ends - 19in Gap No Ends

2 500

3 000

0.25 Hz



Drive shaft (not shown) is non-magnetic stainless steel (ss)

Passive regenerator for diversion flow is non-magnetic ss spheres

CHEX and axial support spacers are non-magnetic ss or G-10

Passive regenerator for diversion flow is non-magnetic ss spheres

### Key milestone – testing MOLS prototype exoskeleton to eliminate magnet eddy current +AC loss heating













## SOW deliverable - A new 200-psia liquid propane heat transfer fluid subsystem has been successfully tested

- 400 psia He HTF pressure drop limits efficiency in 290 K to 100 K MCL designs; Need high density, low viscosity HTF
  - Subcooled, saturated, liquid propane HTF @ ~200 psia freezes at 90 K
  - Test results

- Leak rate of 1 x 10<sup>-8</sup> torr\*liter/sec; vent system meets safety protocols
- Bulk pressure of 173 psia
- Both primary and diversion flows tested for virtual MOLS MCL volumes
- Pressure/temperature sensors show oscillations agree with pumping speed











### SOW Deliverable – Design dual column MCD to integrate with MCL MCL for ASU





- Three modules in ASU (from left to right)
  - Clean, dry air compression, air liquefaction, and dual distillation columns for LOX and LN2
- 100 kg/day of air intake at 100 psia
- 12 kg/day of 99.0% pure LOX at 100 psia output
- 35 kg/day of 99.0 % pure LN2 at 15 psia output
  - 53 kg/day recycle stream 79% N2, 20% O2, 1% Ar
- Used MCD technology for mechanical design
  - Rapid prototype full-size mockup of one column





## SOW deliverable – Design an efficient MCL module to integrate with MCD for ASU

- Two rotary wheel stages cool 100 psia air from 295 K and liquefy it at ~100 K
- Six proven thermomagnetic refrigerants
  - Refrigerant mass/layer sequentially decreases from hot-to-cold stages
- Liquid propane HTF flows vary by layer
- All stages have ~6.5 T uniform flux density over ~120° arc of magnetic wheel
- Variable frequency with ~1 Hz design choice
- Expected performance for MCL is FOM = 0.60; improvement over FOM = 0.25 of conventional
- Estimated cost of ASU will be analyzed after MCL/MCD design integration is completed.
- Determine how this type of ASU scales
  - Expect ISO container size will be a few tonne/day
  - Scaling to larger sizes will be accomplished by putting containers in parallel





# Technoeconomic analysis for integrated ASU making 10-90 tonne/day of LOX is a deliverable

1825

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• Cornelissen and Hirs analysis of large-scale ASU shows largest efficiency impact are air compression and liquefaction modules.

CORNELISSEN and HIRS: EXERGY ANALYSIS OF CRYOGENIC AIR SEPARATION

Unit	Exergy losses (in kW)	Rational efficiency
Air compressor and cleaning	2751	0.48
Main heat exchanger	333	0.86
Distillation unit	788	0.46
Liquefaction unit	4853	0.25
Argon purifying unit	85	0.02
Total	8810	0.28

- Previous TEA for 6-stage MCL for LAIR operating at 0.5 Hz for 10 tonne/day of LOX with no GN2 return indicated the percentages of total capex for different components.
  - s/c magnets and magnetic refrigerants are largest expenses
  - Use layered regenerator to enable only 2 magnets
  - Use higher frequency to reduce mass of refrigerants
- Finish TEA analysis for ASU 10-90 tonne/day of LOX
- Industrial collaborators want proof of performance before investing in new technology



## Progress Summary of project-73143 since May 2021 and completion plans for FY22



- Deliverables completed 4/15/2022
  - New liquid propane pump assembled and tested
  - MOLS improved to eliminate magnetic force and eddy current magnet heating
  - MCD section of ASU analyzed and designed for small engineering scale (100 kg/day)
  - Detailed design of MCL module for integrated ASU on track
- Deliverables are on track to be completed before 9/30/2022
  - Demonstrate MOLS prototype cools to ~100 K
  - Demonstrate liquefaction of 100 psia air at ~1 kg/day
  - Design ASU with rotary wheel MCL using propane HTF and dual column MCD
  - Complete Techno-Economic Analysis of integrated ASU design as function of scale
  - Test propane as HTF in MOLS
  - Write 2-3 papers describing results
  - Complete several IDRs; 3 in progress and 2 more identified
  - Contact potential collaborative partners interested in cost sharing and licensing ASU IP
    - ✓ Plan to cross the "Valley-of-death" to convince for larger companies the performance is real.





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Thank you!