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

• 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

**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

**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

**Budget:**
- FY21-NCTE from FY20 to 9/30/2021
- Total DOE funds received to date: $2.0 MM
Highly efficient MOLS design focuses on three key subsystems of MCLs

- 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 $\Delta B$ during AMR cycle
- Integrate all subsystems to execute AMR cycle illustrated in T-S diagram
- Efficiency measured by figure of merit (FOM)
  - $\text{FOM} = \text{Ratio of ideal to real work rates (see eqn.)}$
  - Regenerators designed to minimize all sources of $\Delta S_{\text{IRR}}$ and auxiliary work rate input
  - $\text{FOM} \sim 0.60$ achievable

$W_{\text{real,layer}} = (\dot{Q}_{\text{CHEX}} + \dot{Q}_{\text{LC}} + \dot{Q}_{\text{Para}}) \left(\frac{T_H}{T_C} - 1\right) + \frac{T_H \int_{T_C}^{T_H} \Delta S_{\text{IRR}} dT}{\int_{T_C}^{T_H} dT}$
MCL prototype test apparatus was upgraded with new superconducting magnet subsystem

- 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

- 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; $B = \mu_0(H + M(T, B))$ for agreement between 2-D/3-D
- Experiments at end of March showed great improvement
- $\Phi = \int B^* A$ = 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

- 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 \( \Delta T_{He}^{ave} \) 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 \( m_{He} \) flow

- He temperature out of demagnetized layers changes during flow steps of cycle are time dependent. For hot-to-cold flow:
  \[ T_{He}(t) = T_{COLD} - \left( \int_0^{t_{flow}} \Delta T_{Cold}(t) dt \right) \]
  Too little \( m_{He} \) underblows \( \dot{Q}_{C}(T_{COLD}) \) – reduces cooling power of magnetic refrigerant
  Too much \( m_{He} \) 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

- 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.

Progress update

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Proudly Operated by Battelle Since 1945
A 200 psia liquid propane heat transfer fluid subsystem is being built and tested

- Pressure drop of 400 psia He HTF is largest source of $\Delta S_{IRR}$ 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

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

- Continuous distillation of O\textsubscript{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

- 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

❖ 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

• 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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  ▪ David Lyons

• DOE- Fuel Cell Technology Office
  ▪ Neha Rustagi

• The Team
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