Overview of CLC Research at the National Energy Technology Laboratory

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NATIONAL ENERGY TECHNOLOGY LABORATORY

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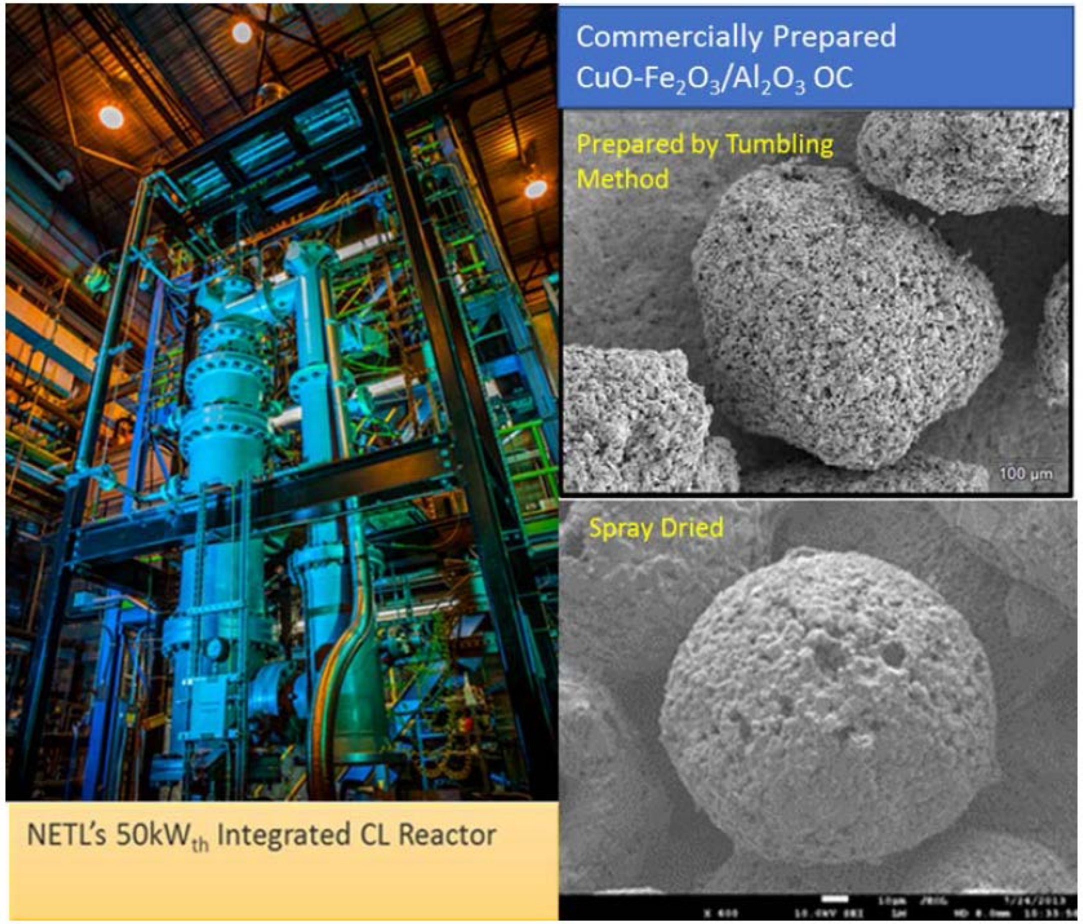
Solutions for Today | Options for Tomorrow

U.S. DEPARTMENT OF ENERGY

Overview of CLC Research at the NETL

Outline

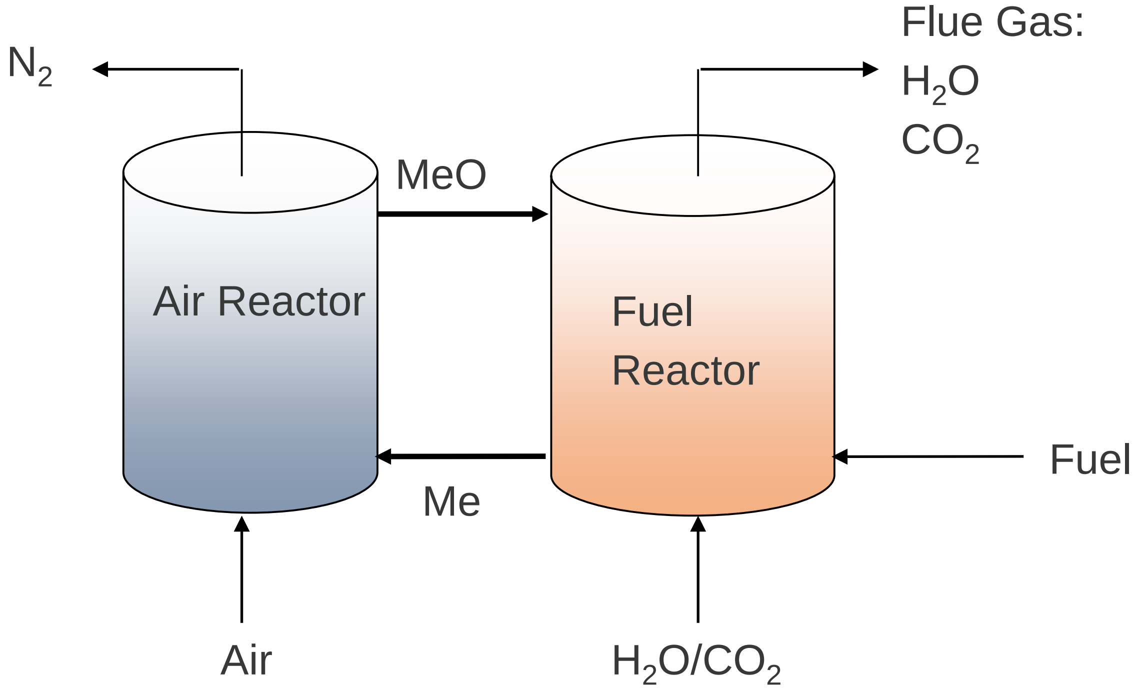
- **Motivation**
  - Purpose
  - Current status
  - Path forward
- **Review of technology issues**
- **Summary of NETL/RIC efforts**
- **Future work**



Ref: Siriwardane, R., Riley, J., Bayham, S., Straub, D., Tian, H., Weber, J., and Richards, G., 2018, "50-kWth methane/air chemical looping combustion tests with commercially prepared CuO-Fe2O3 -alumina oxygen carrier with two different techniques," Applied Energy, V213, pp92-99

What Is Chemical Looping Combustion (CLC)

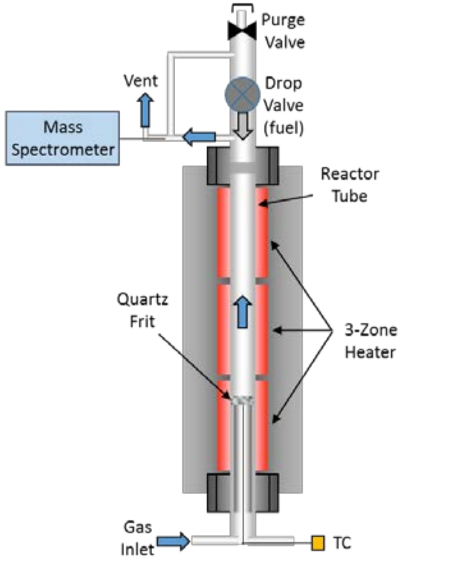
- **Fundamentally different approach to combustion**
  - Fuel and air do not mix
  - Oxygen transport is provided by solid O_2 carrier
- **CO_2 separation is as simple as condensing water vapor from flue gas (in theory)**
- **Typical temperature range (800-1000C)**
  - Too low for thermal NOx production
- **Capital equipment and process design is similar to CFB combustors**

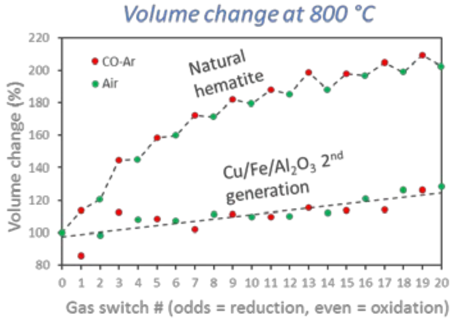


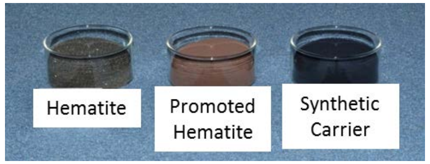
NETL On-Site CLC Research Purpose

What is our end goal?

- **By 2020, determine if CLC is a technically feasible option for coal-power generation**
    - →Data and information for strategic decision making

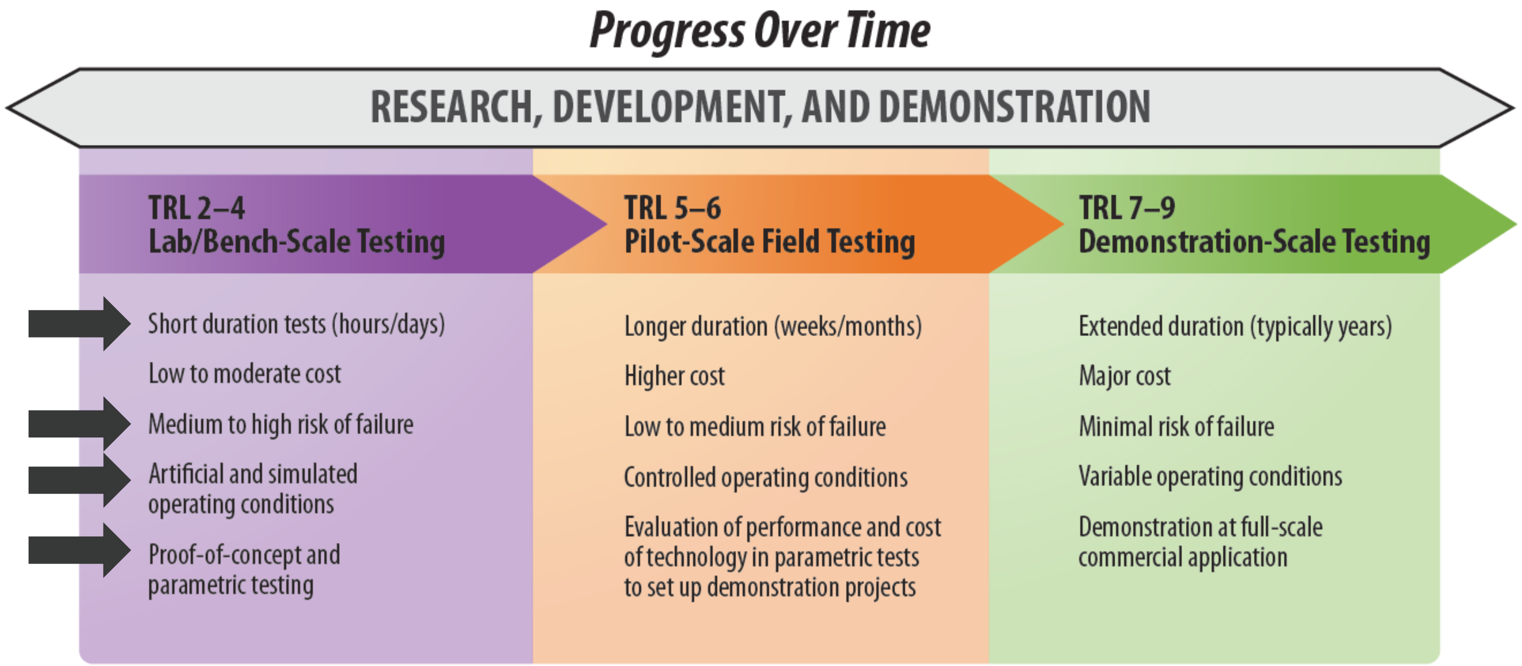






Current Status of CLC Technology

Where Are We Now?



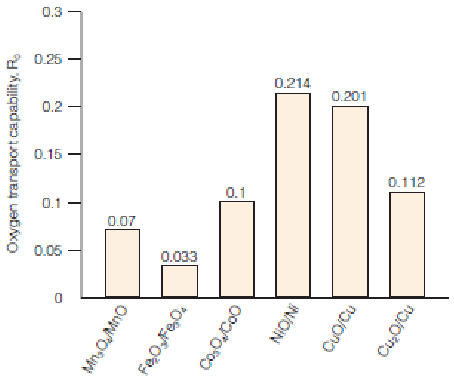
- **Techno-economic analyses (TEA's) show encouraging potential for GHG mitigation**
  - Operating costs are a concern (oxygen carrier makeup costs)
- **CLC test facilities exist**
  - Operating experiences are limited to less than ~100 hrs
  - Data quality and reliability need improved

Technical Challenges and General Approach

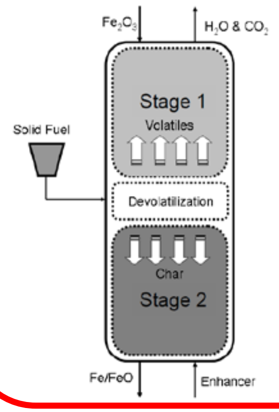
How Do We Get There?

- **NETL/RIC general research approach**
  - Focus on major issues common to most CLC concepts
  - Provide realistic data to support NETL process modeling initiatives
  - Integrate smaller research components toward common goals and overall purpose
- **Technical challenges for CLC**

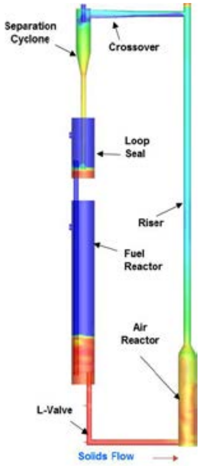
1. Oxygen Carrier Performance & Durability



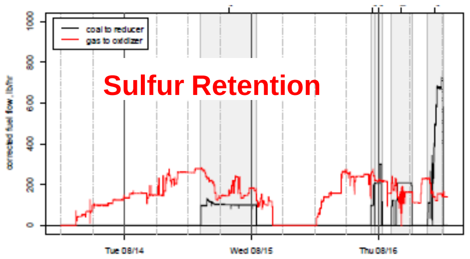
2. Fuel Conversion



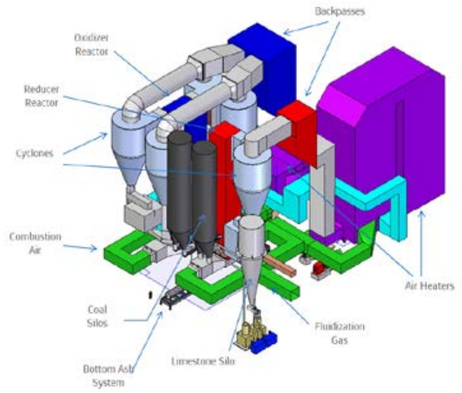
3. Solids Transport & Control



4. Coal type and trace species

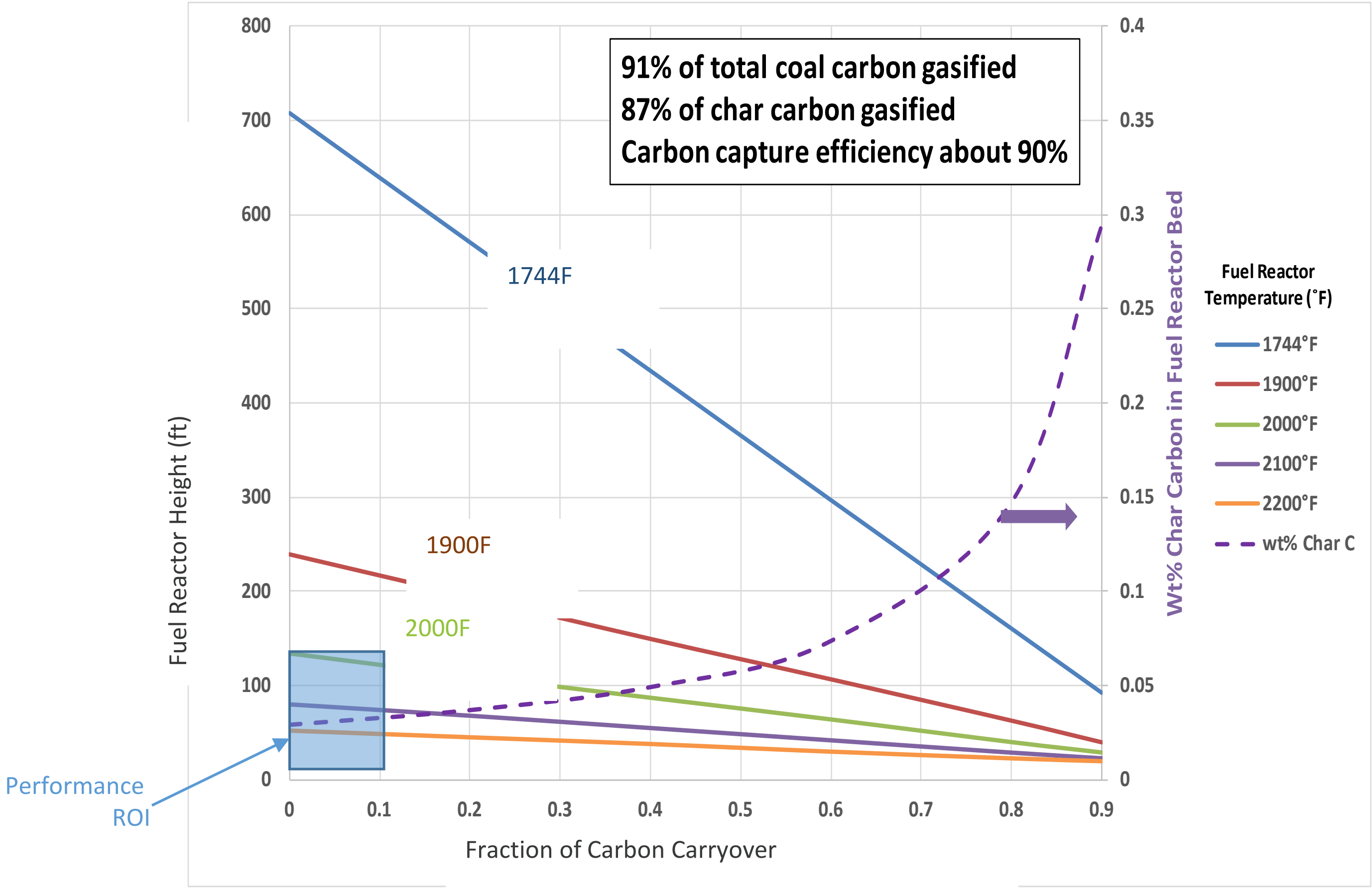


5. System Engineering Design



Fuel Conversion Issue – Background

- **Char gasification is rate limiting step**
- **Model assumptions**
  - Circulating CFB configuration
  - FR height constraint (~130 feet)
  - 90% CO_2 capture constraint
- **Options**
  - Char/carbon-stripping
  - Higher temperatures
  - Higher oxygen capacity OC's
  - CLOU oxygen carriers



Recent Accomplishments

Fuel Conversion Challenges

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Technical Challenge	NETL/RIC Research Progress	Consequence	Impact
Fuel Conversion	• Lab-scale carbon stripping concept achieves greater than 90% fines removal from a dilute (< 1 wt. %) polydisperse mixture with a separation flux that is ~20X higher than a bubbling fluidized bed approach	• Unreacted char can be separated from the carrier stream and re-circulated to the fuel reactor	• Effective char carry-over mitigations can include novel solid-solid separation devices
	• TEA screening studies have been completed for several general types of oxygen carrier in a CFB/CLC configuration	• CLOU materials can eliminate fuel conversion issues • High capacity oxygen carriers can eliminate fuel conversion issues • Oxygen carrier materials that operate at fuel reactor temperatures in excess of 1100C can eliminate fuel conversion issues	• Eliminate fuel conversion issues for coal-CLC • Eliminate requirement for char/carbon stripping

Oxygen Carrier Durability and Cost

- **Oxygen carrier material is key reactant in CLC process**
  - NETL/RIC has focused on synthetic Cu/Fe/alumina OC
  - Gen 1 – spray dried, ~100 micron, good reactivity, expense was a concern
  - Gen 2.0 – mechanical mixing, 200-600 micron, good reactivity, cost reductions are possible
- **Predictive models for particle degradation, particle collision/force pdf's, and attrition models**

→ Demonstrate progress on NETL's Gen 2.0 OC material

➢ Goal: 50X improvement in oxygen carrier makeup cost

→ Transfer lessons learned, approach, models, and other tools to external CLC developers

Recent Accomplishments

Oxygen Carrier Performance and Durability Challenges

Technical Challenge	NETL/RIC Research Progress	Consequence	Impact
Oxygen carrier development, performance, and durability	• NETL/RIC OC materials have an oxygen transport capacity that is 5-10 times higher than most	• Decreases stoichiometric carrier-to-fuel ratio • Decreases carrier circulation rate	• Can reduce carrier losses
	• NETL Gen 2.0 material has been successfully tested under realistic "auto-thermal" CLC conditions for more than 10 hours in two separate test campaigns	• Validate industrial process to manufacture a synthetic OC • Validate auto-thermal operating mode for NETL's 50kW test unit	• Successfully test Gen 2.0 material under realistic CLC conditions (Increases technology readiness level to TRL 3-4)

	• NETL/RIC is developing predictive models and unique diagnostic tools to improve understanding and predictions for all CLC oxygen carriers.	• Fundamental understanding of degradation mechanisms
		• Tools to predict environmental forces on particles
		• Models to predict attrition rates
			• Develop toolsets that can be applied to all OC mat'ls
			  ◦ More robust OC material formulations
			  ◦ Improve process design considerations
			• Reduce risks for scale-up

Oxygen Carrier MakeUp Costs

What is NETL/RIC's target?

- Coal
  - 1 MW_{th} @ \$40/ton → ~\$7/MW_{th}-hr
  - 1 MW_{th} @ \$60/ton → ~\$10/MW_{th}-hr
- Oxygen carrier makeup cost target
  - **Less than \$5/MW_{th}-hr**

Is this goal measurable, achievable, realistic, etc.?

Exhibit ES-3 Cost of electricity breakdown comparison

Cost	Fe_2O_3 (\$/MWh)	$CaSO_4$ (\$/MWh)	Conventional PC BBR Case 12
Capital	49.6	53.4	73.1
Fixed	11.3	12.2	15.7
Variable	25.7	8.4	13.2
Maintenance materials	3.2	3.5	4.7
Water	0.4	0.4	0.9
Oxygen carrier makeup *	18.7	1.1	N/A
Other chemicals & catalyst	1.9	1.7	6.4
Waste disposal	1.4	1.7	1.3
Fuel	28.4	30.8	35.3
Total	115.1	104.7	137.3

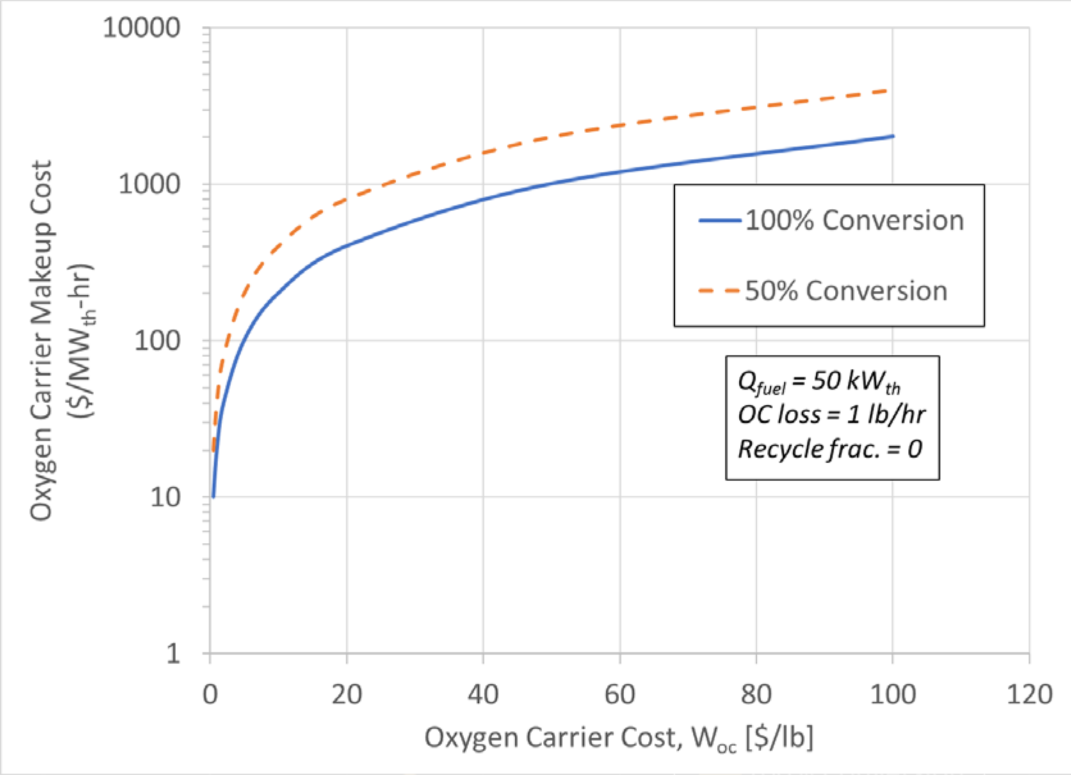
*Fe_2O_3 oxygen carrier makeup: 132 tons/day @ \$2,000 per ton; Limestone carrier makeup: 439 tons/day @ \$33.5 per ton

Measurable Expression for OC Makeup

Bayham, Straub, and Weber, "Operation of the NETL Chemical Looping Reactor with Natural Gas and a Novel Copper-Iron Material". United States. doi:10.2172/1350960. https://www.osti.gov/servlets/purl/1350960

$$\text{Oxygen Carrier Makeup Cost} = \frac{W_{\mathrm{OC}} \cdot \dot{m}_{loss}(1-\alpha)}{\dot{N}_{fuel} X_{fuel\rightarrow \mathrm{CO_2}} \cdot HHV_{fuel}}$$

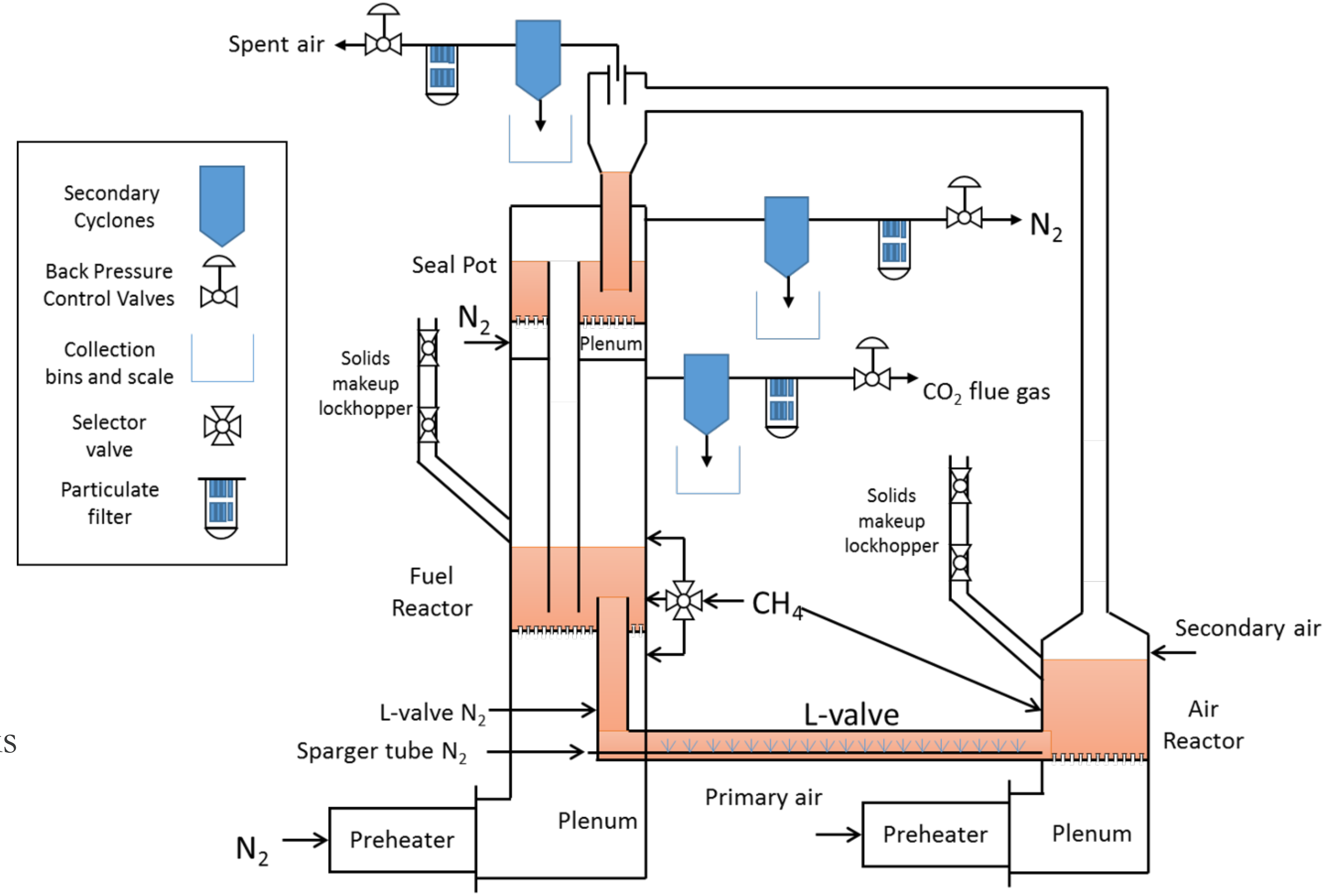
- W_{OC} = **Oxygen carrier cost [$/lb]**
- $\dot{m}_{\mathrm{loss}}(1-\alpha)$ = **Carrier loss rate due to attrition, degradation, etc. [lb/hr]**
  - α = fraction of material lost that can be recycled and re-used
- $\dot{N}_{\mathrm{CH_4,in}} X_{\mathrm{CH_4 \rightarrow CO_2}}$ = **Carrier reactivity/Fuel converted to CO_2 [mol/sec]**
- HHV_{fuel} = **Heating value of fuel [MJ_{th}/mol]**



NETL 50 kW$_{th}$ Circulating CLC Testing

Test Setup

- **Carbon steel shell/refractory lined**
- **Fuel Reactor**
  - Bubbling bed (8" dia)
  - Natural gas (1 of 3 locations)
- **Air Reactor**
  - Turbulent fluidized bed (6" dia)
  - Natural gas for startup
- **Gas Seal/Seal Pot**
  - Bubbling bed (8" dia)
- **Vent lines (3 individually controlled)**
  - Cyclones remove hot solids prior to filter banks
  - Back-pressure control valves



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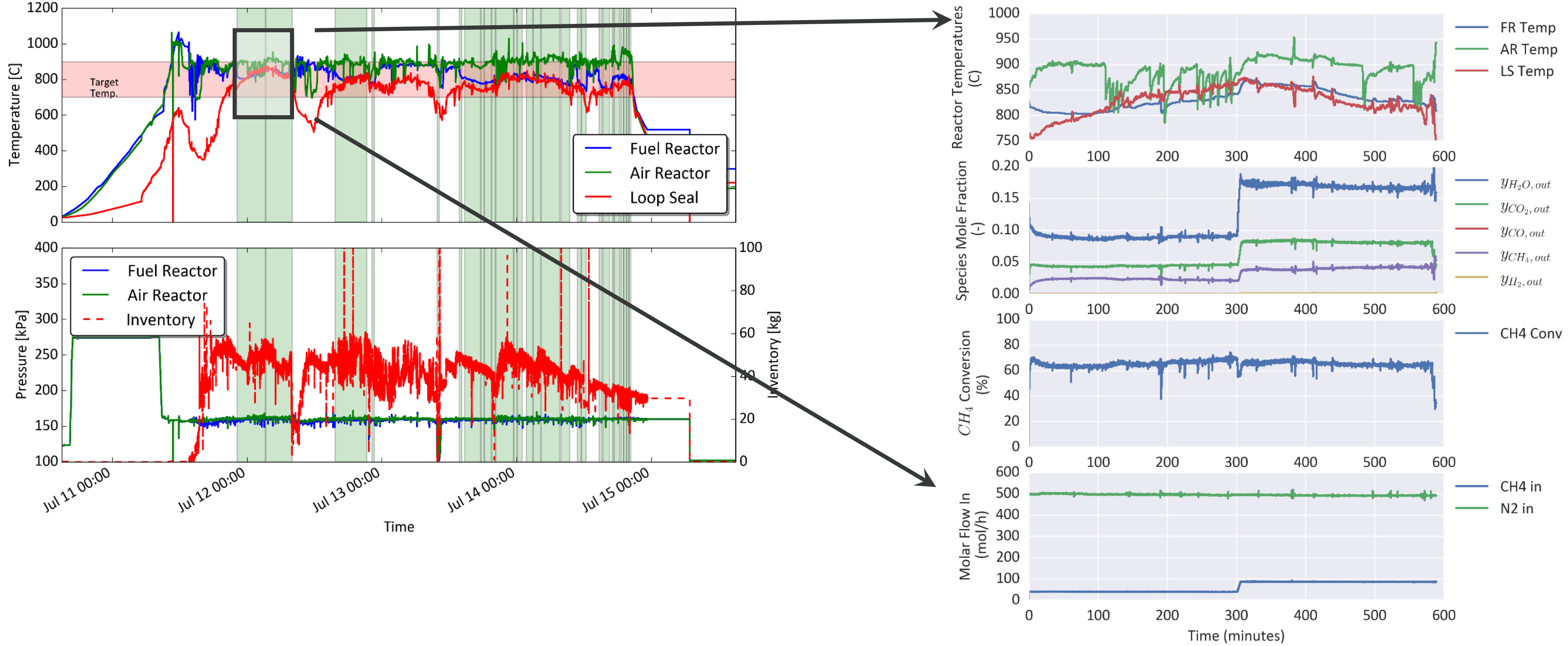
Gen 2.0 – Circulating CLC Test Results



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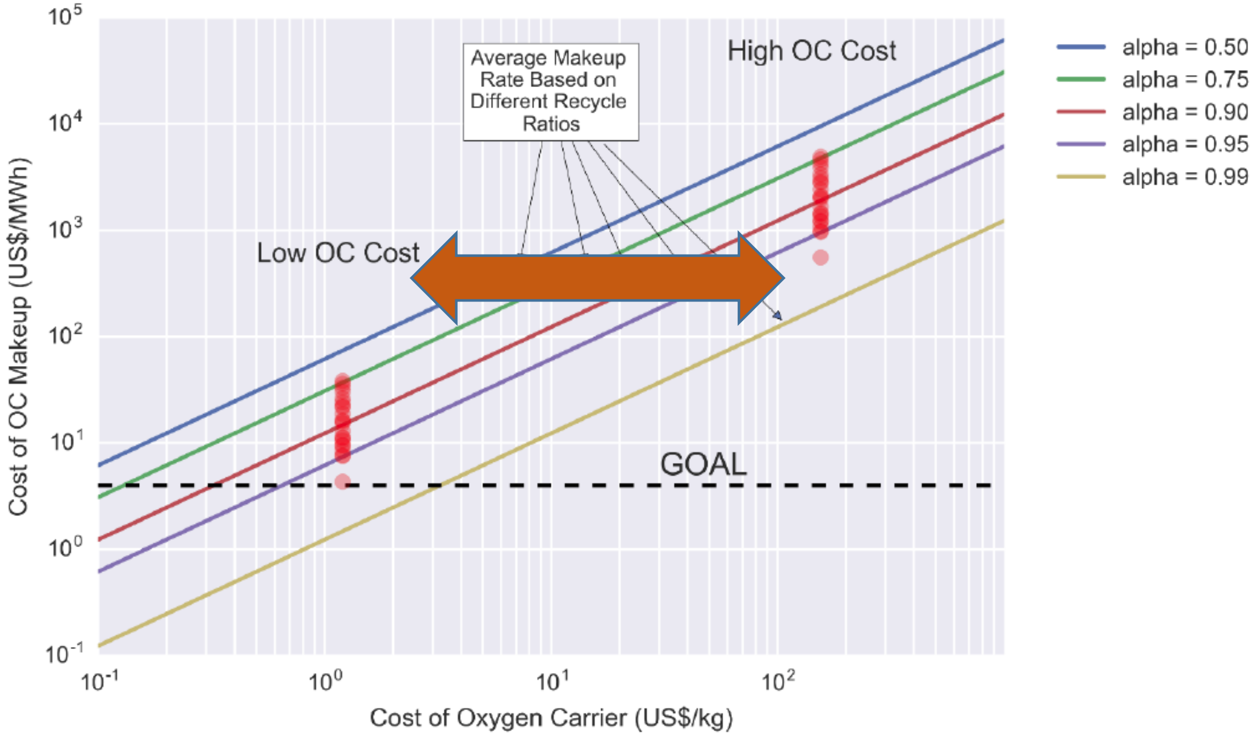
NETL 50 kW_{th} Circulating CLC Testing

Demonstrate oxygen carrier make-up costs \$5/MW_{th}-hr in a circulating CLC test facility

- **O_2 carrier make-up costs**
  - Baseline for 50kW_{th} test unit estimated
  - Key issue for CLC technology maturation
- **Gaps to address . . .**
  - Lower-cost O_2 carriers
  - Fundamental effects of redox cycling on attrition
  - Need longer duration tests under redox and circulating conditions
  - Detailed study of OC manufacturing process, including OC cost estimates



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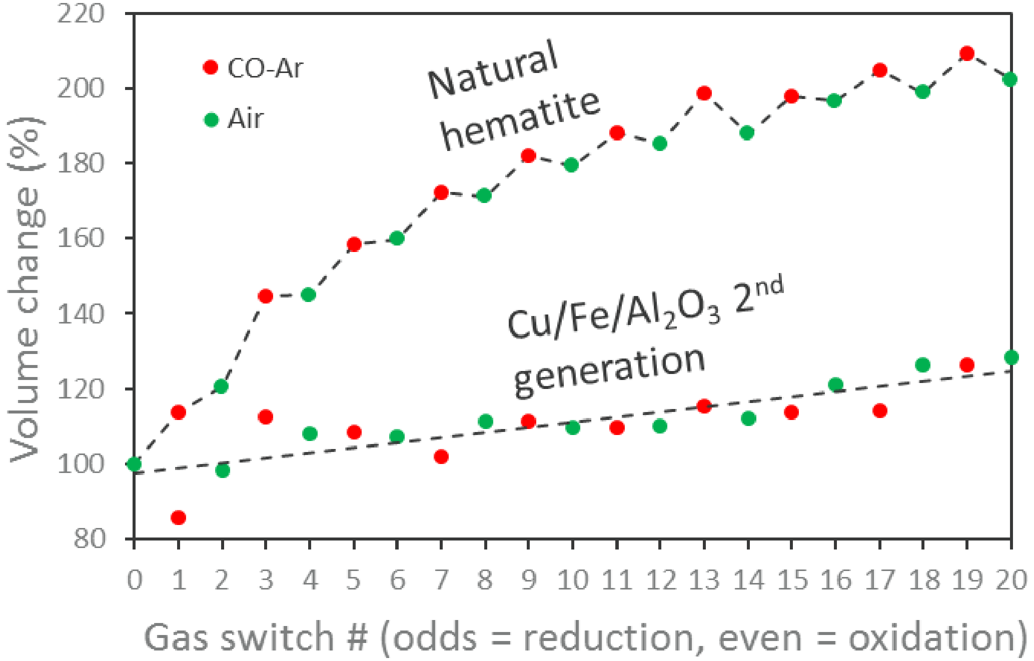
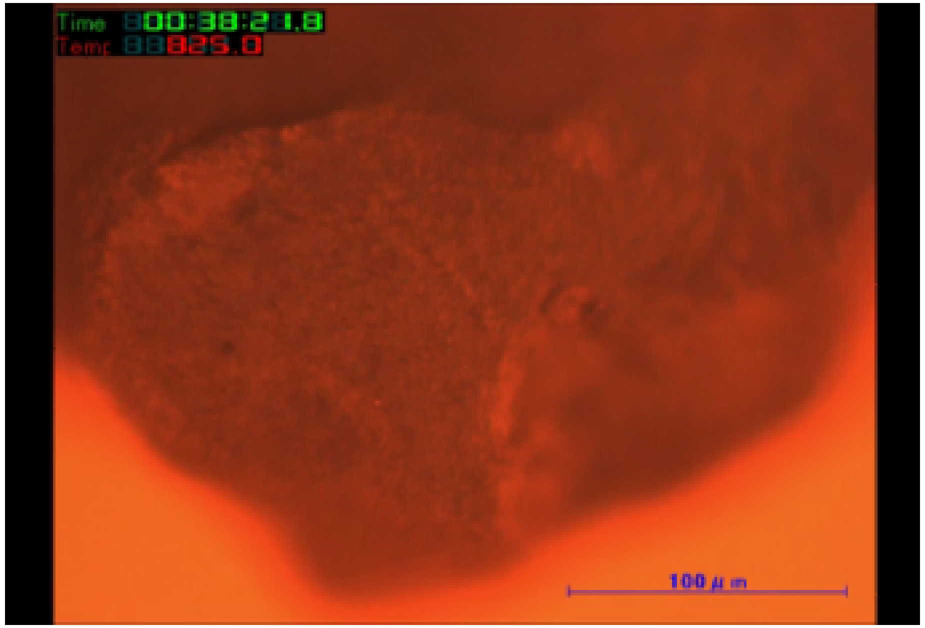
Tools and Models For OC Degradation

Volume change at 800 °C



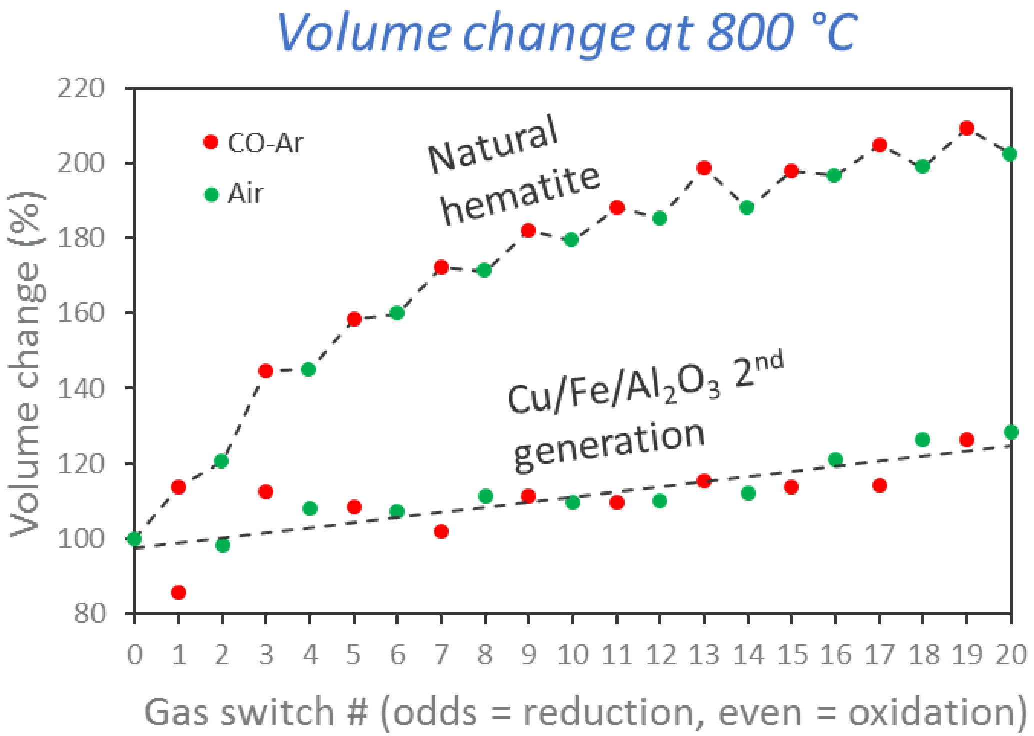
Developed tool to characterize particle changes as a function of time and/or redox cycles
→ Use to improve OC make-up costs

Hematite particle reacting with CO (3rd Cycle)



(playback time is ~5x actual speed)

Tools and Models For OC Degradation



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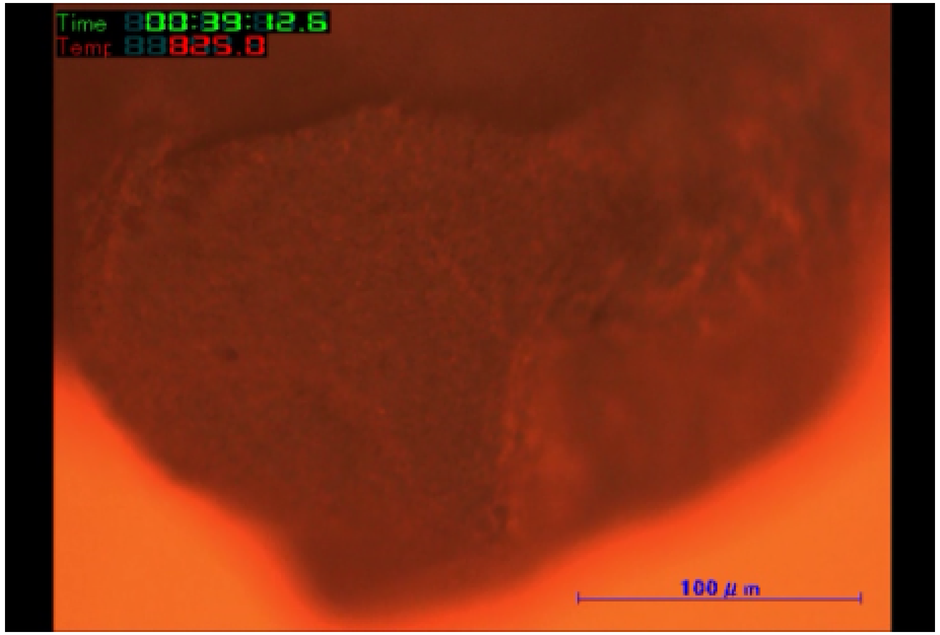


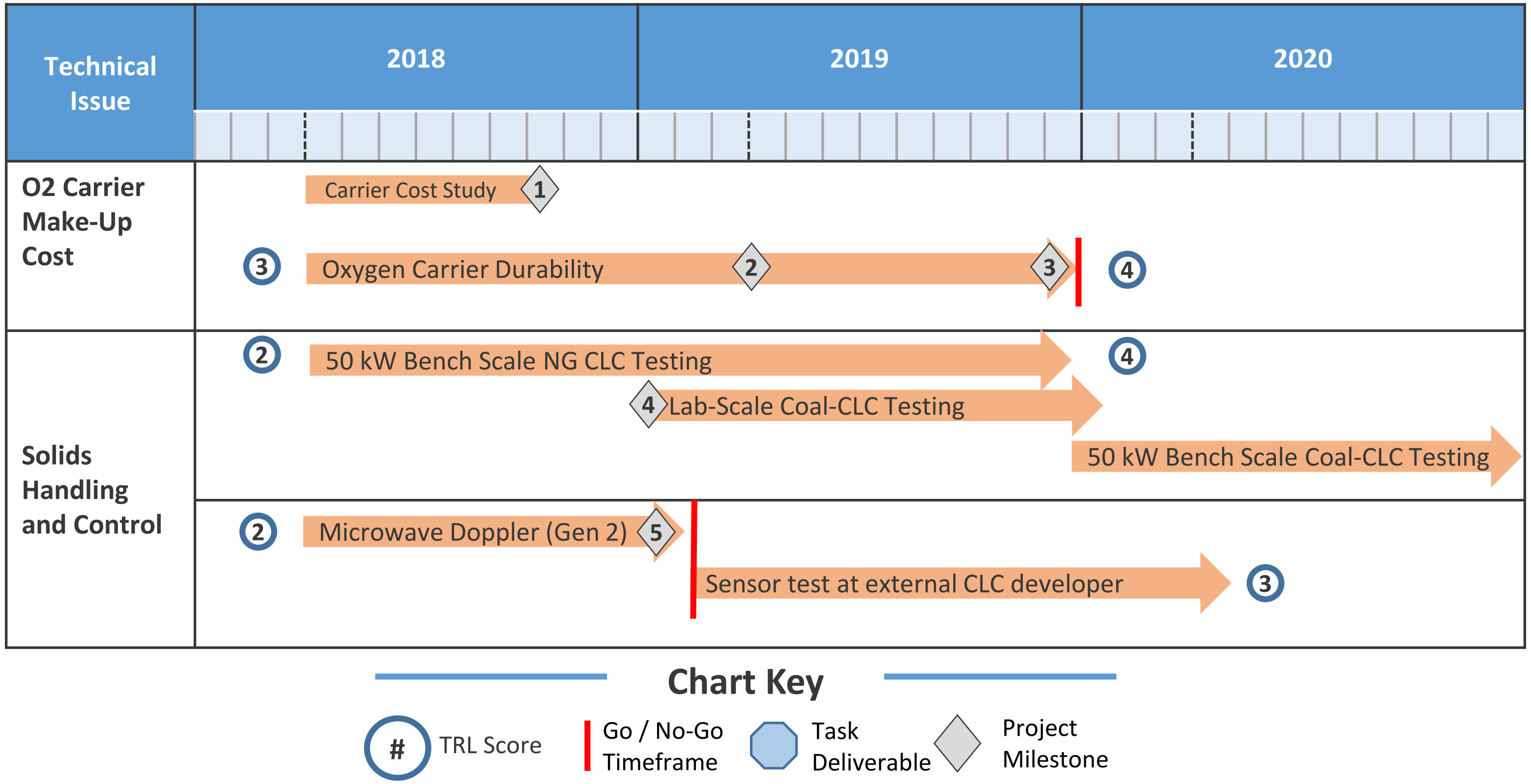
(playback time is ~5x actual speed)

Summary and Conclusions

- **CLC is a "young" (low TRL) technology with several challenges**
  - Several options are available to address fuel conversion issues for coal-CLC
  - Significant progress made on carrier durability and cost issues
- **NETL/RIC is addressing a critical metric and goal for this technology**
  - Oxygen carrier makeup cost < $\$5/MW_{th}$-hr
- **Results from NETL's 50kWth circulating test facility**
  - Established a baseline performance OC makeup cost using NETL's Gen 2.0
  - 40 hours of CLC operation on NETL Gen 2.0 OC
  - More details are available from OSTI report (Bayham et al., 2017)

Future Work

Major Project Milestones



	Major Milestones
1	Complete oxygen carrier production cost study
2	Preliminary technology readiness assessment (Target: >50 hrs and 25X reduction in relative OC make-up cost)
3	Final technology readiness assessment (Target: > 100 hours and 50X reduction in OC make-up cost)
4	Complete lab-scale solid fuel reactor commissioning
5	Complete testing of Gen 2 microwave sensor

Acknowledgements

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This work was performed under FWP #1022401 under the guidance and leadership of John Rockey, Chris Channel, Geo. Richards, and Bhima Sastri. The research staff in this project continue to exceed expectations and rise to meet the next big challenge. These team members deserve all the credit for the successes in this project.

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