DOE Award Number: DE-SC0017221

High-Efficiency Post Combustion Carbon Capture System

National Carbon Capture Center Contractor Report

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Summary:

The current report details work completed at the National Carbon Capture Center (NCCC) in Wilsonville Alabama as part of SBIR Phase 2 project DE-SC0017221 High-Efficiency Post Combustion Carbon Capture System by contractor Precision Combustion Inc. The Phase 2 project duration was May 27, 2019 – May 27, 2023.

PCI's innovation is a compact, modular Post Combustion Carbon Capture System (PCCCS) utilizing high internal volume nanosorbents for carbon capture, supported on a tailorable substrate. Our system enables low pressure-drop, high volumetric utilization and high mass transfer, and is suitable for the rapid heat transfer and low temperature swing regeneration operating modes needed for cost-effective carbon capture. Capital and operating costs are reduced based on lowered energy demand.

During this period, PCI conducted three independent test campaigns at the National Carbon Capture Center – in 2020, 2022 and 2023 on the bench scale test bay (under 50 lb/hr). The first test campaign was conducted with coal-fired flue gas, while the second and third campaigns were conducted with flue gas from the natural gas boiler.

Two different PCCCs units were tested during Phase 2, with a first-generation sorbent and test bed assembly tested in 2020 and a second-generation optimized sorbent and test bed in 2022 and 2023. The test bed consisted of PCI sorbent – a modified metal organic framework (MOF) coated on a thin mesh substrate trademarked as Microlith.

Tests held at NCCC demonstrated sorbent performance and durability in the presence of water and other typical contaminants of the flue gas stream (e.g. SO_2). The sorbent was rapidly thermally regenerated at low temperature (70-90 ^oC) and was stable under all test conditions.

Data acquired at NCCC was utilized to refine PCI's CFD models for CO_2 sorption-desorption kinetics and refinement of our techno-economic analysis for a full-scale PCCCS unit.

Our models indicate that our technology can meet the DOE cost targets for carbon capture by 2035. The technology is flexible to multiple applications including post-combustion carbon capture from industrial sources or direct air capture.

Introduction:

For the high space velocity sorbent structure, PCI has developed and patented a short contact time mesh-based substrate, trademarked Microlith® [i,ii], coated with the densified nanostructured sorbent. The combination enables higher surface areas per unit volume, decreased bed volumes with equivalent effectiveness to other types of monolithic or loose packing, without pressure drop penalty. Additionally, up to twenty times higher mass and heat transfer coefficients are obtainable as compared to other sorbent systems such as monoliths and pellets, due primarily to boundary layer minimization and break-up, boosting CO_2 removal rates with greater sorbent bed utilization and less bypass inherent to packed beds or monoliths. Individual Microlith elements have very low thermal mass and low axial conductivity, and consequently quickly transfer heat from the gas phase to the sorbent (Figures 1,2). Our sorbent manufacturing technology allows for adherent and durable sorbent coatings (as well as other high surface area sorbents) on the Microlith substrate.

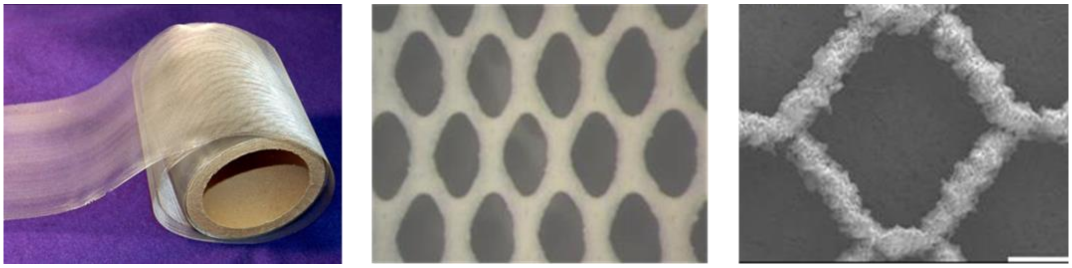


Figure 1: Microlith mesh coated with sorbent – cell opening 600 µm.

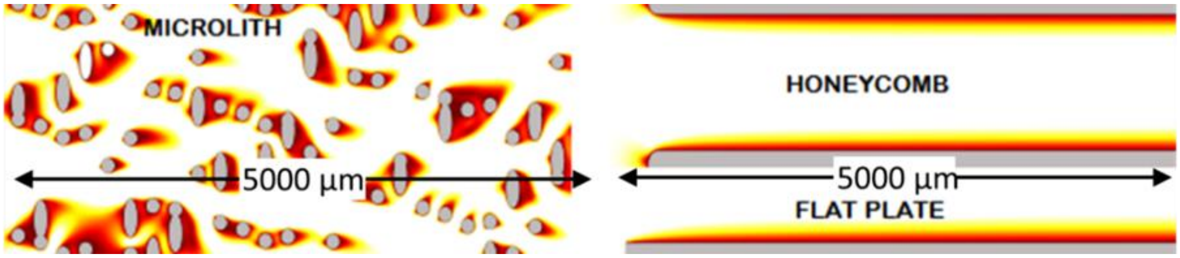


Figure 2: CFD analysis of flow in 2D cross-sections through a stack of 21 mesh elements (left), a single channel in a honeycomb monolith (right, top) and across a flat plate. Solids are shaded grey, velocities of flow within the fluid boundary layer, defined as velocities less than 99% of bulk, are shaded from yellow to red (slowest), bulk flow regimes are not shaded. Individual mesh elements interfere with flow streamlines causing boundary layer disruption mixing in a laminar flow regime and form-drag vortex formation; under the same flow conditions boundary layer and fluid streamlines are well developed in conventional configurations. This disruption reduces resistance to CO_2 transport to and from the sorbent surface and substantially increases volumetric effectiveness.

The architecture is modular and allows us to use absorbent more efficiently, intensively, and controllably, with shorter and more efficient regeneration cycles that use significantly less total energy and enable longer lifecycles before replacement. Compact size enables retrofit to existing volumes. The use of these engineered substrates provides a high degree of uniformity and control. Elements within the bed can be graded in size and tailored to different adsorbents as well as being configured in planar or radial design. Internal bed structure reduces the potential for settling and bypassing. The geometry promotes greater and more uniform adsorbent surface-air contact with reduced pressure drop, higher surface mass and heat transfer rates enable faster and more complete regeneration with less local overheating and reduced local peak temperature leading to longer adsorbent life. As a result, this technology will provide energy savings, high adsorbent utilization with less sorbent mass required while providing performance equivalent to a pellet or powder bed, extended service life, lower maintenance, and reduced waste compared to disposable systems and alternative regenerable systems that lack this architecture [iii].

For this application, PCI developed metal-organic frameworks (MOF) based sorbents. Metal Organic Frameworks (MOFs) are novel materials that combine organic ligands and metal or ion clusters in a robust 3D structure with very high (thousands of m^2/g) internal surface area. Through judicious design, these structures can be tuned towards the preferential adsorption of certain molecules such as CO_2. Our sorbents were developed to be selective for CO_2 with good sorption capacity, resistant to humidity and other impurities and have lower regeneration energy compared to other types of sorbents to reduce overall cost of carbon capture.

PCI has successfully developed methods for coating metal organic-frameworks (MOF)-based sorbents on Microlith mesh (Figure 1) with excellent adhesion and cohesion. The coatings maintained their integrity over hundreds of thermal cycles and the sorbent performance was unaffected by the coating process.

Small-scale Post Combustion Carbon Capture (PCCCS) modules were assembled at PCI based on mesh supports coated with sorbent in different configuration and tested at NCCC through multiple test campaigns through the duration of the Phase 2 project.

The focus of the Phase 2 work was to evaluate the operability and regenerability of the PCCCS unit and to obtain test results in a real flue-gas environment. Tests were performed at the lab-scale in the under 50 lb/hr test bay (Figure 3).

The specific objectives of the field trial testing were to demonstrate:

- Operability and regenerability of the PCCS unit
- Low pressure-drop and low power consumption for operation and for regeneration.
- Acquire data for full scale-modelling of PCCS unit and balance of plant components.
- Develop techno-economic analysis to estimate the increase in COE.

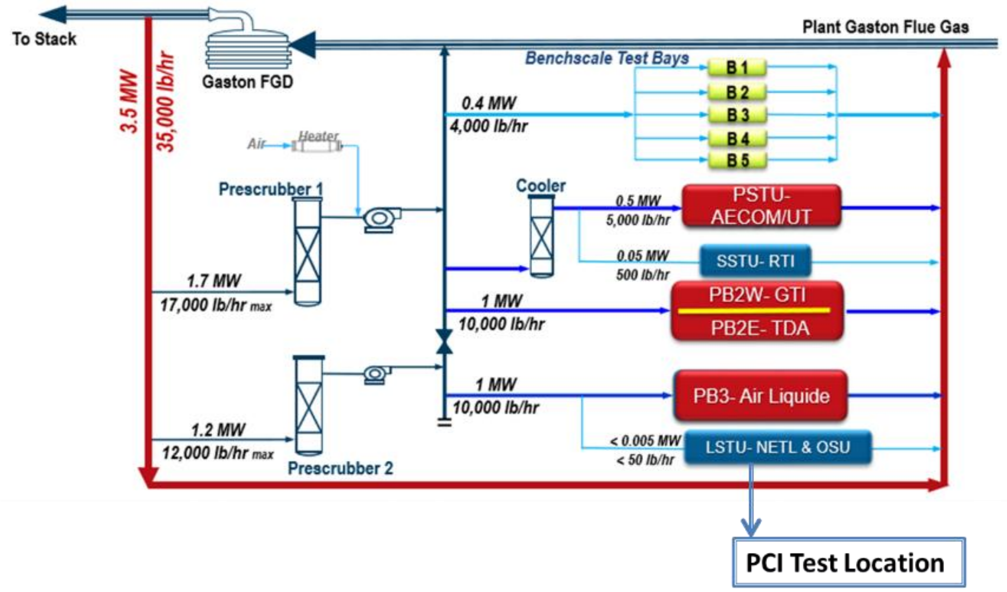


Figure 3: PCI test location in NCCC facility.

The timeline of the NCCC test campaigns is detailed below:

- 2020 – First Skid Assembly.
- 2020 – First Testing Campaign – coal derived flue gas – 1 week.
- 2021 – Skid Returned to PCI post pandemic shut-down.
- 2021-2022 – Testing on site at PCI with simulated mixtures (coal derived).
- 2022 – Second skid assembly.
- 2022 – Second skid tested at NCCC with natural gas derived flue gas - 2 weeks.
- 2022 – Skid returned to PCI to resolve by-pass issues.
- 2023 – Third test campaign with natural gas derived flue gas – 1 week.

Results:

2020 Skid Design and Installation

The adsorber module tested in 2020 consisted of a "jelly-rolled" coil of Microlith mesh coated with sorbent, with the fluid flow being delivered through the center of the coil towards the outlet (Figure 4).

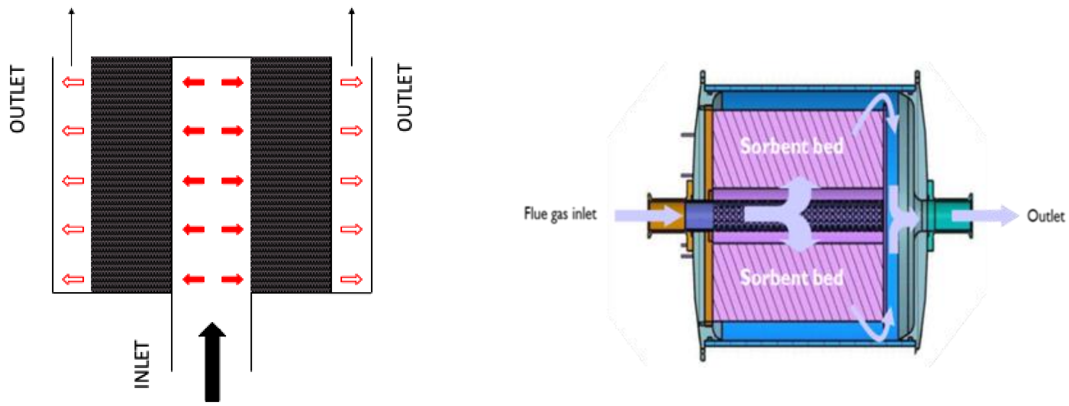
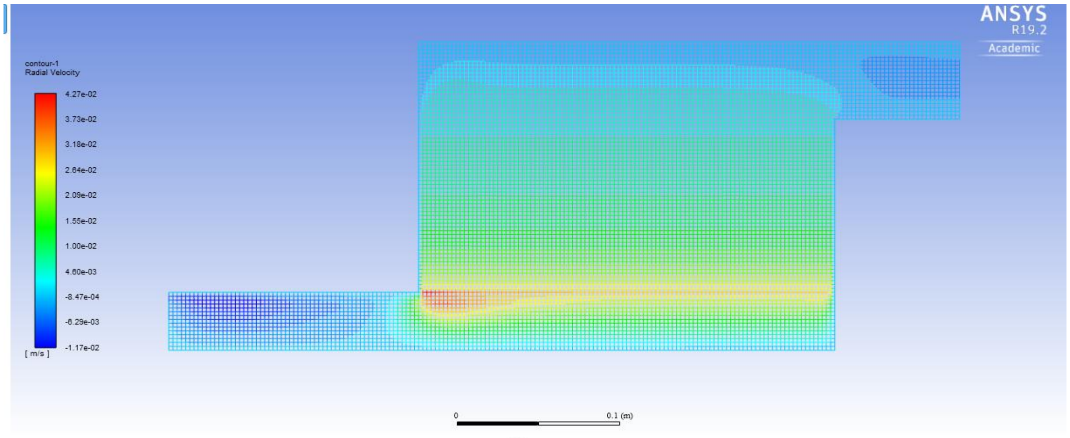


Figure 4: Schematic of adsorber module flow pattern and coated mesh assembly.

Work was done during design and shake-down testing to ensure that the fluid flow velocity in the unit is constant through the jelly rolled Microlith coil. It is important to have uniform flow so that the porous reactor is used efficiently, without areas of low flow or areas of recirculation.

A CFD model (using Ansys Fluent) of the flow through a jelly rolled reactor showed uniform distribution of the gas flow in the reactor at the viscous resistance typical of flue gas mixtures – Figures 5, 6. For the modelling effort, we assumed temperatures, pressures and viscosities typical of coal-derived flue gas mixtures.



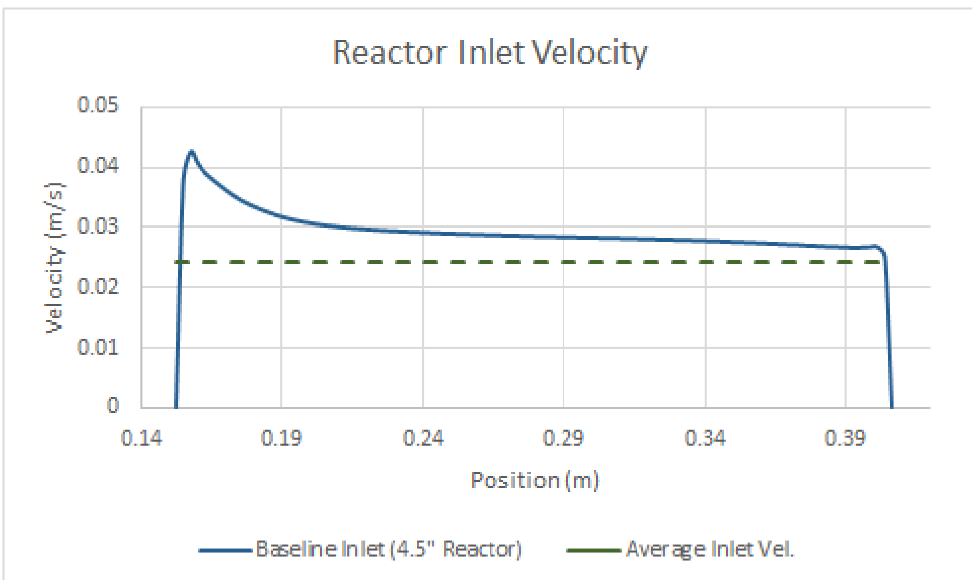


Figure 5: CFD model of Radial Velocity across jelly roll reactor.

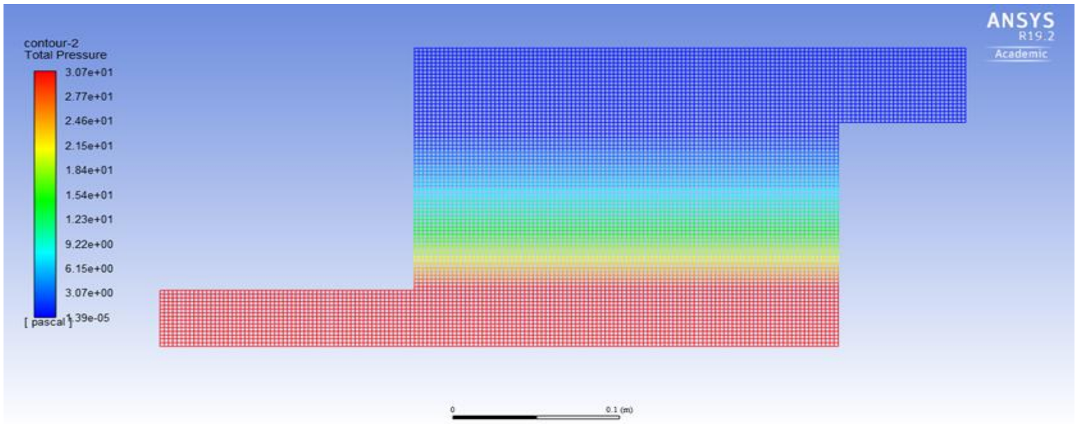


Figure 6: CFD model of pressure drop across jelly roll reactor.

A sub-scale reactor was assembled and tested at various fluid flow velocities with simulated flue gas to determine pressure drop which validated the CFD model (Figure 7). The pressure drop through the Microlith reactor is under 0.05 psi at up to a gas hourly space velocity (GHSV) of 160000 h^{-1} – conditions typical of a full-scale PCCCS unit.

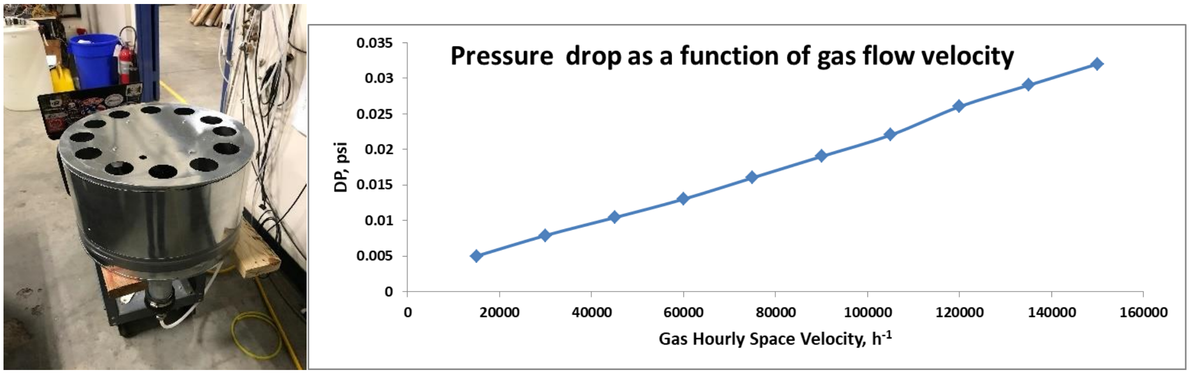


Figure 7: Subscale Microlith assembly and measured pressure drop as a function of fluid flow velocity.

The Microlith mesh was rolled into a jelly-rolled adsorber where the flue gas will be delivered through a central shaft and passed through the bed towards the outlet.

The unit was fitted with all BOP components including the dehumidifier, sorbent housing, flow meters, heater controls, bypass, thermocouples, valves etc. Figure 8 shows the process flow diagram for the system and Figure 9 shows a photograph of the test unit as assembled at the NCCC test location. A condenser was installed prior to the sorbent bed to evaluate the effect of water on the sorption process. The sorbent can be by-passed as needed to allow flow of moisture into the bed.

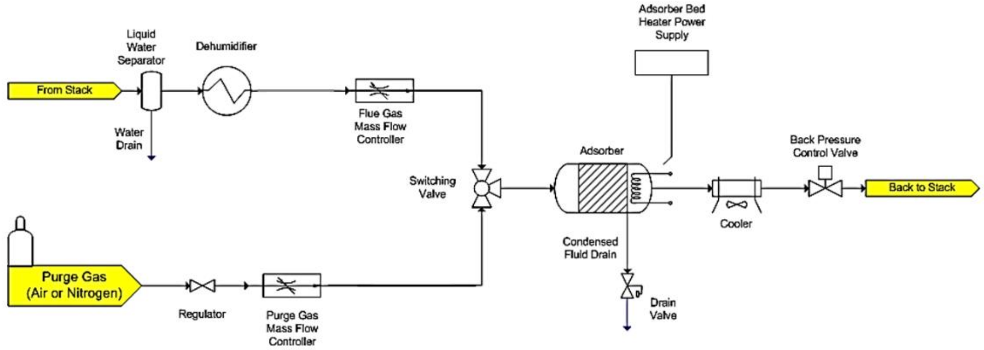


Figure 8: Process Flow Diagram of NCCC CO_2 Capture Test Setup – 2020 test campaign.

Process Control functions:

Flue Gas flow to targeted flow rate during adsorption stage

Purge gas flow to targeted flow during purge stage

Bed temperature warmup to targeted desorption temperature during purge stage

Adsorbent bed pressure control for pressure maintenance

Switching between flue gas and purge gas during stage changes

CO_2 monitor of exhaust gas

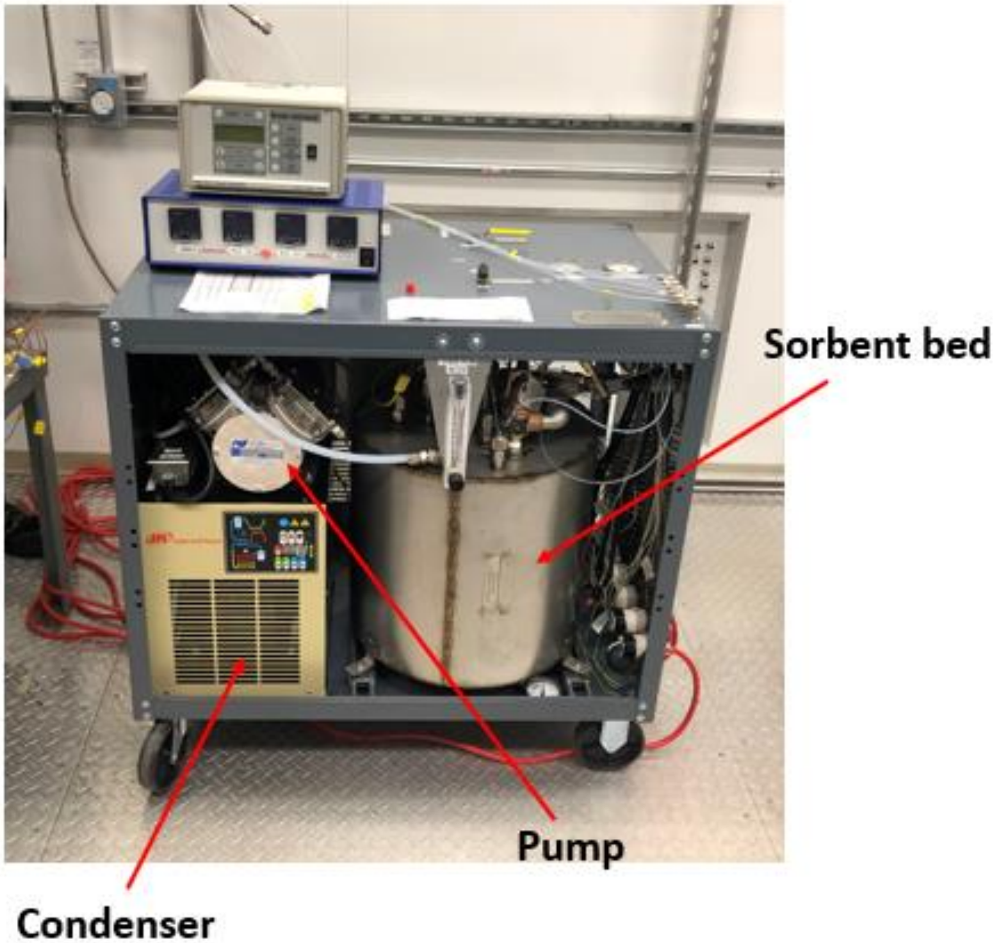


Figure 9: PCI Test Skid On-site at NCCC.

In the March of 2020 testing campaign, the PCI team travelled to NCCC to install the test rig on the PC-4 flue gas stream. The rig has been installed, leak tested, pressure checked and all balance of plant components were tested (including heaters, mass flow controllers, pressure valves etc.), in accordance with the HAZOP parametric study performed by PCI and NCCC prior to skid arrival. Before testing, the sorbent bed was preconditioned by heating the bed and flowing N_2 through in order to purge CO_2 and moisture from the bed picked up during shipment to NCCC. Following the COVID-19 shutdown the skid returned to PCI for in-house testing.

2022-2023 Skid Design and Installation

The skid tested in spring of 2022 was a second iteration of the PCCCs unit. Due to 2020-2021 travel restrictions, the first iteration was mostly tested in-house with simulated flue gas mixtures.

In Phase 2 A of the project, both the sorbent and the bed design have been optimized. Figures 10 and 11 show the second iteration of the PCCCS unit that was tested in 2022 at NCCC.

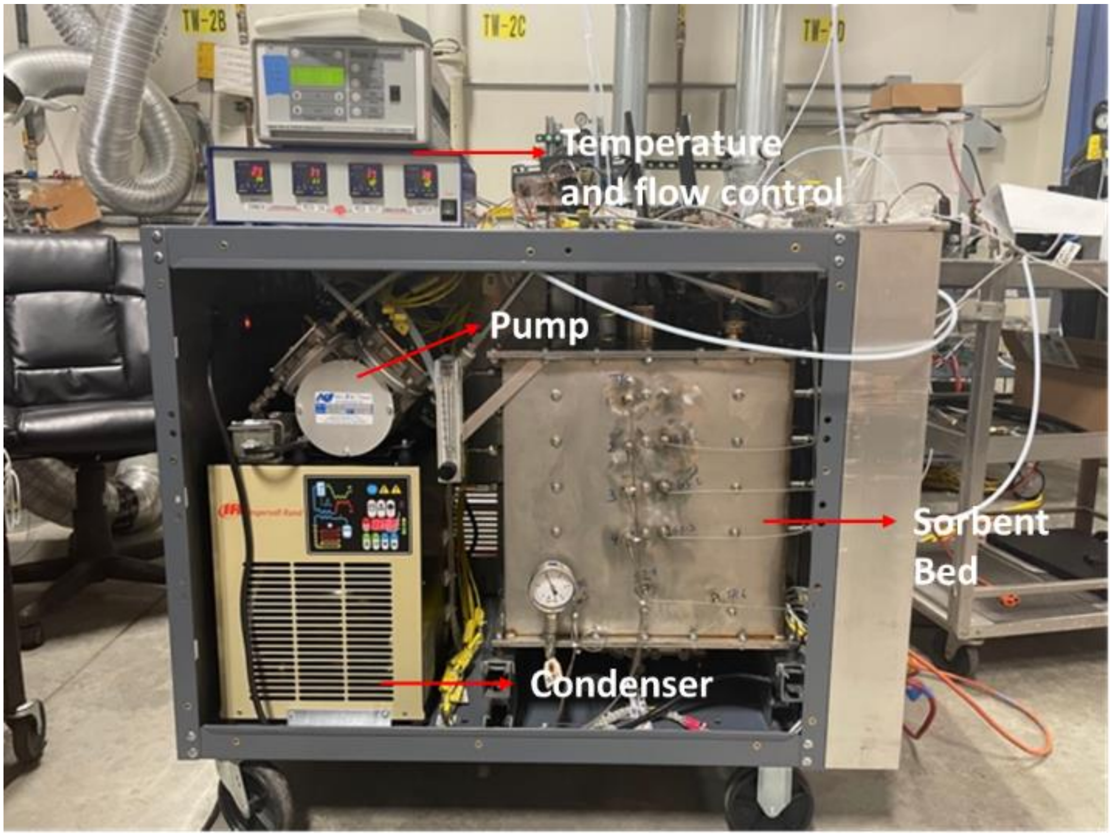


Figure 10: PCCCS Unit tested at NCCC.

For the second iteration we assembled mesh coated with sorbent in a stacked sheet configuration with the flue gas delivered perpendicular to the coated mesh to ensure better uniformity. Heat exchange tubes were integrated with the coated mesh to ensure uniform heat distribution through the bed. The pressure drop and velocity distribution were similar to the jelly-roll configuration and the process flow diagram and operations follow the same order as described in Figure 8.

Test Data

For a typical test we monitored:

- Inlet compositions and properties – CO_2 content off the stack; water, oxygen and nitrogen content, inlet temperature and contaminants (in particular SOx)
- Adsorption-desorption CO_2 curves – CO_2 evolution was measured both at the outlet of the test bed and with sample ports every 5 inches withing the sorbent bed. CO2 capture rate calculations were based on this data.
- Temperature – inlet, outlet and test bed temperatures were monitored with multiple thermocouples through the test bed both during adsorption and desorption. Energy balance calculations were based on these measurements.
- Monitoring of humidity – different levels of humidity up to saturation were passed through the bed to assess the effect of water on both stability and capacity of the sorbent.
- Adsorption and desorption temperatures were monitored and varied to determine optimal operating conditions of the unit.
- Pressure drop was monitored for all tests to design pumps, blowers and other balance of plant components of the full-scale system.

Some examples of test data are shown in Figures 11 and 12.

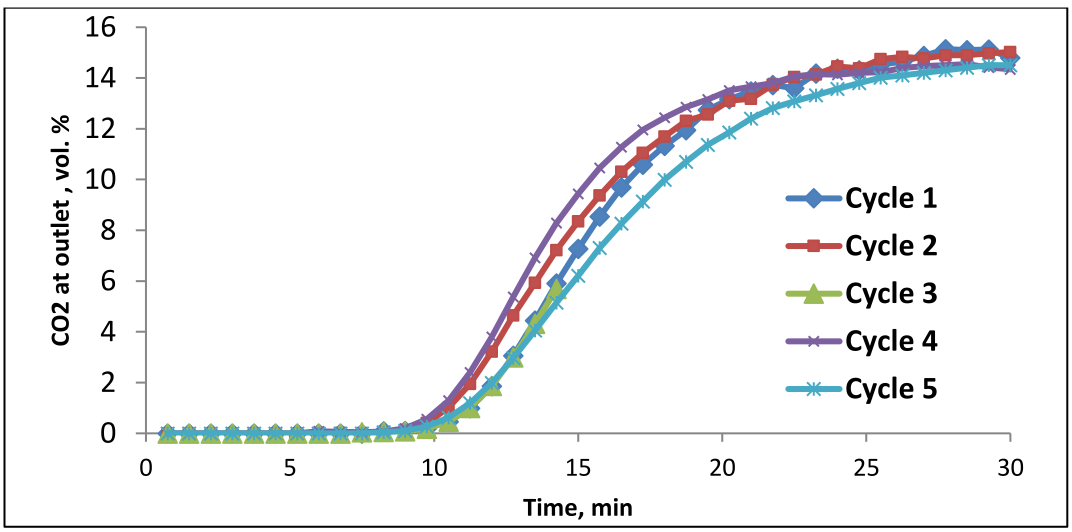


Figure 11: Five consecutive sorption cycle with coal-fired flue gas on Microlith unit installed in 2020 at the NCCC location. GHSV- 6000 h^{-1}; inlet temperature 30 ^oC, CO_2 15 % vol.

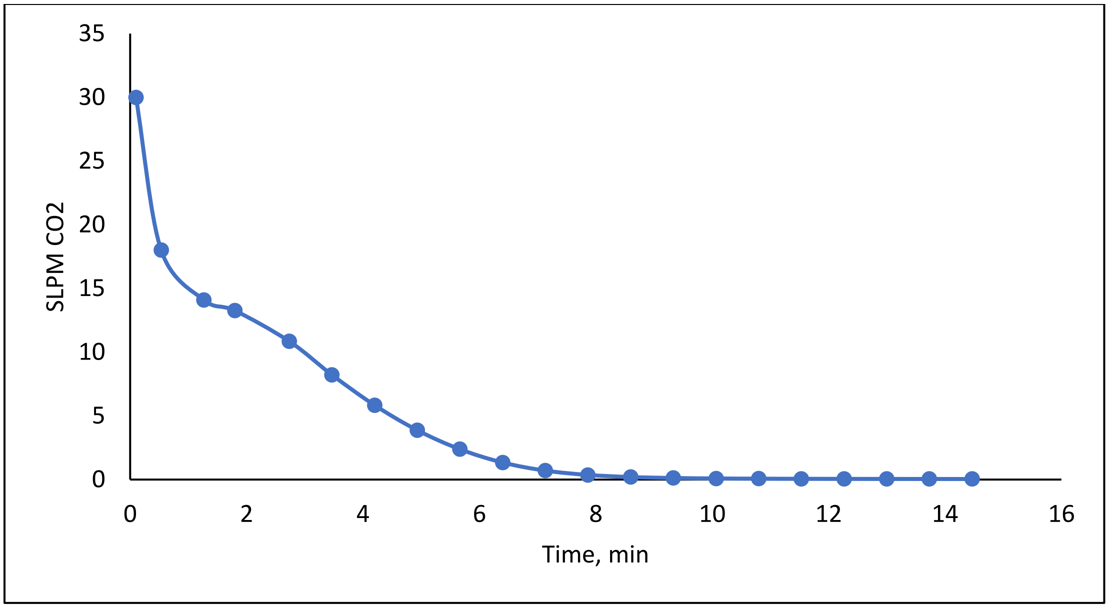


Figure 12: Typical thermal desorption curve post sorption of CO_2 from coal-fired flue gas.

A summary of the test conditions and test results from the 2020 test campaign are shown in Table 1. Figure 13 shows sorbent capacity as a function of the cycle number.

For this test campaign the first 20 cycles were tested in house (shakedown) followed by cycles 20-40 at NCCC. Shakedown was performed with a mixture containing 12% CO_2 vs 15-16% for cycles 20-180 and as such the capacity was lower. Following the 2020-2021 COVID-19 shutdown, the test skid was returned to PCI where testing continued in-house with simulated flue-gas.

The gas composition tested at PCI was blended to reproduce as faithfully as possible the stack composition from NCCC including humidity, CO2, nitrogen and oxygen content.

To assess the durability of the sorbent against flue gas contaminants we introduced additional SO_2 in the flue gas blend increasing the SO_2 concentration from under 10 ppm available at NCCC to 75 ppm in house.

Table 1: Summary of test conditions and results from the 2020 test campaign.

Metric	**Value**
Inlet flowrate, SLPM	50
Inlet CO2 concentration, vol. %	10-15
Humidity	Saturation
SO2, ppm	75
Inlet temperature, °C	20-40
Average sorption time, min	15
Average regeneration Temperature, °C	80
Average regeneration time, min	7
Number of cycles	180
Average CO_2 working capacity, wt. %	5
Average CO_2 working capacity, mmol/g	1.14
Average Capture Rate, %	**73-80**
Average kWh/Kg CO2 (includes water)	**0.7 -0.9**

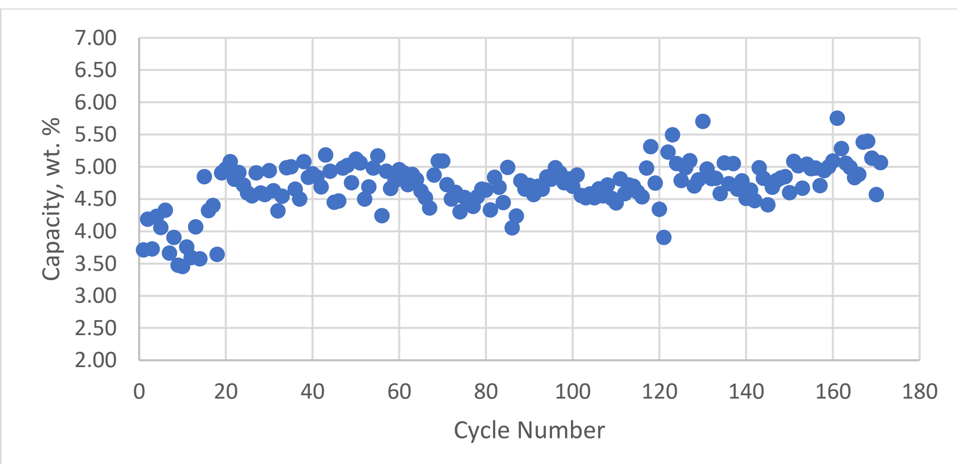


Figure 13: Average cyclical CO_2 capacity of sorbent bed. The first 20 cycles shakedown were completed with 10-12 vol. % CO_2. Cycles 20-40 were performed at NCCC. The reminder were performed in house at PCI's location.

A summary of the test conditions and test results from the 2022-2023 test campaign are shown in Table 2. Figure 14 shows sorbent capacity as a function of the cycle number.

For this test the inlet contained approximately 4 vol. % CO_2. A second-generation sorbent was tested in this campaign with flue gas from the NCCC natural gas boiler. During the first test campaign we varied several parameters including flow rate (contact time), inlet temperature, regeneration temperature etc.

Following the first test campaign in 2022 we identified several bypass issues in the contactor. The skid was returned to the PCI facilities where it was tested and reconfigured to avoid the bypass. The second test campaign was completed in 2023 with the reconfigured unit. The bypass issues were resolved as evidenced by the higher sorbent capacity recovered under identical test conditions to 2022.

Table 2: Summary of test conditions and results from the 2022-2023 test campaign.

Metric	**Value**
Inlet flowrate, SLPM	20-50
Inlet CO_2 concentration, vol. %	4
Humidity	Saturation
SO_2, ppm	1-2
Inlet temperature, ^{o}C	20-30
Average sorption time, min	15-60
Average regeneration Temperature, ^{o}C	80-100
Average regeneration time, min	10-15
Number of cycles	40
Average CO_2 working capacity, wt. %	5.5
Average CO_2 working capacity, mmol/g	1.25
Average Capture Rate, %	**75-85**
Average kWh/Kg CO_2 (includes water)	**0.8 -1.1**

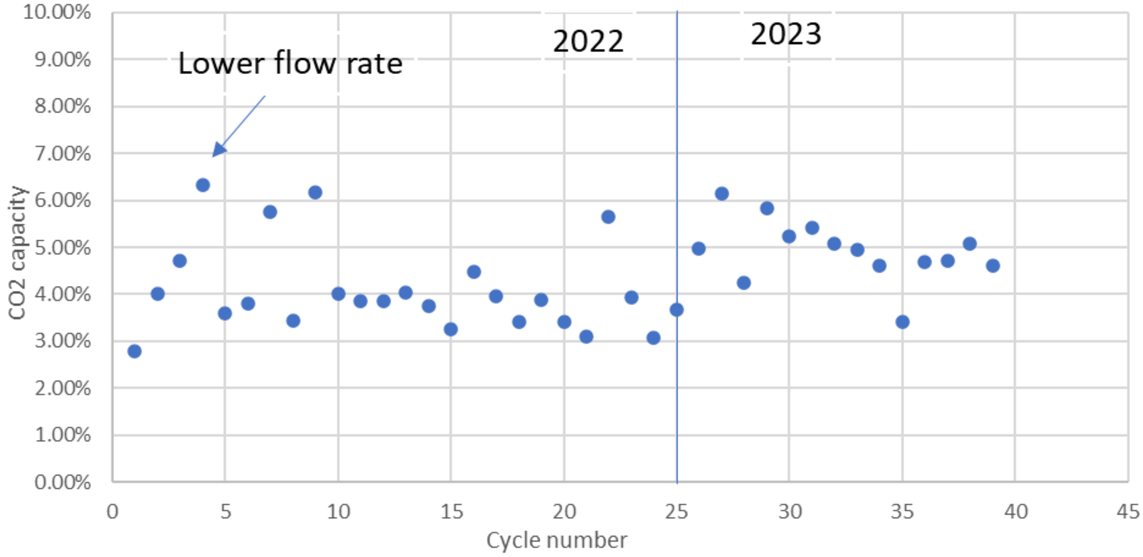


Figure 14: Average cyclical CO_2 capacity of the sorbent bed 2022-2023 test campaign.

The data collected through the NCCC test campaigns was utilized in the design of a full-scale Microlith unit. Adsorption reactions and hydrodynamics were studied individually to determine appropriate models for the various physics with the aid of machine learning.

Figure 15 shows a comparison between NCCC data (2020 test campaign) and simulated data using the CFD model developed by PCI in collaboration with Boston University. (CFD) and machine learning (ML) methods were used to develop reduced order models (ROMs) that are accurate but computationally inexpensive. With the ROMs, we used ML algorithms to predict optimized Microlith reactor designs. Using the Multiphase Flow with Interphase eXchanges (MFiX) CFD software, we have built models of the PCI jelly roll and stack reactor configurations to represent the two separate iterations of the PCCCS unit. MFiX is computationally expensive to run but is able to accurately predict CO_2 capture in the PCI design. MFiX simulations of the reactor are being used to train kernel ridge regression (KRR) algorithms to develop a ROM of the reactor. The ROM created by KRR can then be used with a Bayesian optimization algorithm to optimize reactor design. The KRR model was built by considering the hydrodynamics, reactions, and heat transfer separately and then combing the sub-models into a complete model of the reactor.

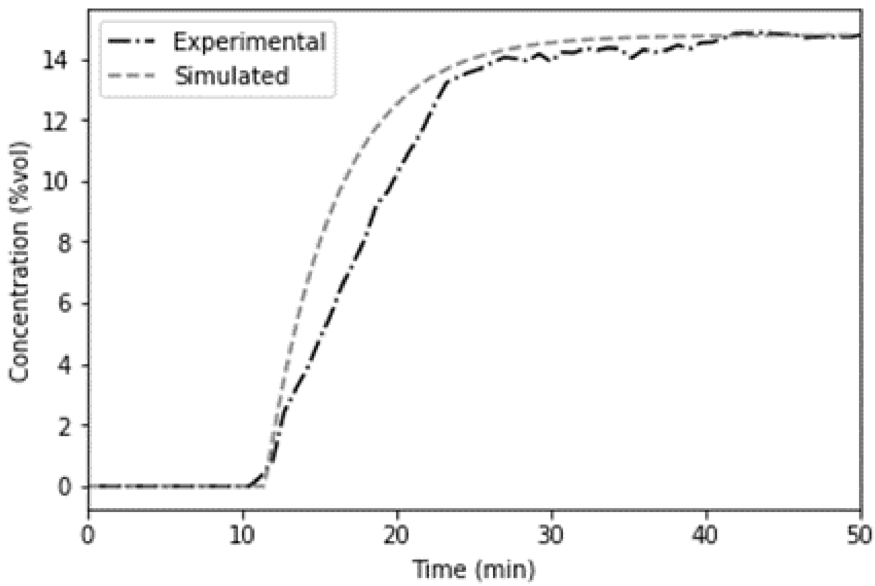


Figure 15: Experimental NCCC versus simulated (CFD model) data.

The NCCC experimental data and the models that were developed based on it were then integrated with NETL's Power Plant Flexible Model (PPFM) to predict costs of large-scale carbon capture using PCI's technology. The PPFM model is a tool part of the carbon capture initiative software that was initially developed for liquid-amine based carbon capture. The model was converted from amine-based to MOF and under pre-determined operating conditions of the sorbent capital, operating, and energy costs were tabulated and impact on electrical power and cost per ton of CO2 captured were calculated. Results of the TEA are shown in Table 3.

Table 3: TEA analysis preliminary results – full scale PCCCS unit.

Plant Type	Base Plant	MEA Plant	Microlith Plant
CO2 Emissions (kg/MWHnet)	789	109	105
CO2 captured (kg/MWHnet)	0	981	945
CO2 Capture Efficiency (%)	0	90	90
Net Plant HHV Efficiency (%)	39.4	28.6	29.7
Existing Coal Producer Cost of Electricity ($/MWh)	39.95	65.24	54.35
Increase in Cost of Electricity (%)	0	63.3	36.0

Conclusions:

Precision Combustion completed three test campaigns at the National Carbon Capture Center throughout the duration of the SBIR Phase 2 project.

Two different small-scale PCCCS units were assembled and tested. The first unit was tested with coal-fired flue gas and the second unit was tested with flue gas derived from the natural gas boiler.

Multiple test conditions were interrogated to measure the efficiency of the contactor and the sorbent performance – these included flow rates, temperature of adsorption and desorption and contaminants.

The sorbent was stable thorough the duration of the test and proved to be resistant to effects of humidity and contaminants. Capture rate was found to scale with capacity, with a maximum of 85%.

The data was utilized to develop full-scale models of PCCCS units operating with flue gas derived from either coal or natural gas plants.

A techno-economic analysis of the full-scale unit was completed using NCCC test data and showed an increase of COE from base case of 36%.

Impact and Future Outlook:

Worldwide, carbon emissions are forecasted to grow dramatically, with fossil fuel combustion and industry accounting for most of the CO_2 emissions. Biofuel combustion is a smaller but fast growing segment. Carbon capture technologies will be an important segment of the overall strategy to reduce carbon emissions and will provide CO_2 for integration with value-added chains of several industries including an emerging industry of converting CO_2 to value added chemicals such as fuels and plastics. Converting CO_2, currently a waste stream, into valuable fuels and chemicals, offers an opportunity to add value to CO_2 producing concerns, such as power plants, cement manufacturers and other fuel-burning industries. Replacing the cost of carbon capture or storage with a positive income stream will improve American industrial efficiency and provide for new business opportunities and markets. Remote locations where it may have been uneconomical to either develop or improve capture technology may now have a viable option. Reduced cost of electricity, replacement of petroleum-derived products with locally, domestically derived drop-in replacements, and enhancement of the US chemical manufacturing sector are all direct benefits of our approach. Additional considerations are given to enhanced oil recovery processes which injects large quantities of CO_2 into geological formations to recover fossil fuels.

PCI will continue to mature the PCCCS technology into Phase 3 and beyond through: Development of integration pathways into practical anticipated applications; Design, guided by test data and analytical modeling results, to make it scalable for Phase III pilot -test platforms and mapping of the cost analysis through further refinement of the TEA and integration into market cost components.

Acknowledgments:

PCI would like to acknowledge financial support from the Department of Energy, grant **DE-SC0017221** as well as the continued support of teams from NETL and the National Carbon Capture Center.

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[i] S. Roychoudhury, D. Walsh, and J.L. Perry, *SAE*, 2005-01-2866 (2005).
[ii] S. Roychoudhury, D. Walsh, and J. Perry, *SAE Int.*, 2004-01-2442 (2004).
[iii] S. Roychoudhury, D. Walsh, and J.L. Perry, *SAE*, 2005-01-2866 (2005).