Final Technical Report

Low Regeneration Temperature Sorbents for Direct Air Capture of CO_2

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Disclaimer

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List of Acronyms/Abbreviations

BEC	Bare Erected Cost
CAPEX	Capital Expense
CF	Plant Capacity Factor
CO_2	Carbon dioxide
DAC	Direct Air Capture
DOE	Department of Energy
EH&S	Environmental, Health and Safety
EPCC	Engineering, Procurement, and Construction Cost
FCR	Fixed Charge Rate
FECM	Office of Fossil Energy and Carbon Management
FT-IR	Fourier-Transform Infrared Spectroscopy
NDIR	Non-Dispersive Infrared
NETL	National Energy Technology Laboratory
NMR	Nuclear Magnetic Resonance
OCFIX	The sum of all first year of operation fixed annual operating costs
OPEX	Operating Expense
PEI	Polyethyleneimine
RH	Relative Humidity
SoCalGas	Southern California Gas Company
SOTA	State-of-the-art
TASC	Total As-Spent Cost
TEA	Techno-Economic Analysis
TGA	Thermogravimetric analysis
TOC	Total Overnight Cost
TPC	Total Plant Cost
TRL	Technology Readiness Level
TSA	Temperature Swing Adsorption
UWy	University of Wyoming
VSA	Vacuum Swing Adsorption
XRD	X-Ray Diffraction

Executive Summary

Susteon Inc., in partnership with University of Wyoming and SoCalGas, successfully met all major technical objectives to (1) scale up the ionic liquid catalyst for amine-based sorbents for improved desorption and absorption kinetics, (2) evaluate the catalyzed amine-based sorbents for direct CO_2 capture process to determine CO_2 adsorption and desorption rates and energy requirements, and (3) based on the experimental results, develop a conceptual process design to perform a preliminary economic assessment to evaluate the potential for DAC process cost reduction using the catalyzed sorbents.

Amine doped solid sorbents are effective for DAC applications and can be regenerated by heat or by a combination of heat, steam, and vacuum. The best sorbent composition identified was polyethyleneimine (PEI) on fumed silica with 200 ppm ionic liquid catalyst. This sorbent formulation was shown (see **Figure E-1**) to have a CO_2 breakthrough capacity twice that of the non-catalyzed sorbent, in laboratory tests with air at 75% relative humidity (RH). The CO_2 adsorption rate was also 40% higher than that of the non-catalyzed sorbent. This type of sorbents has the attributes required for lowering the overall cost of DAC with high CO_2 capacity and high rate of adsorption.

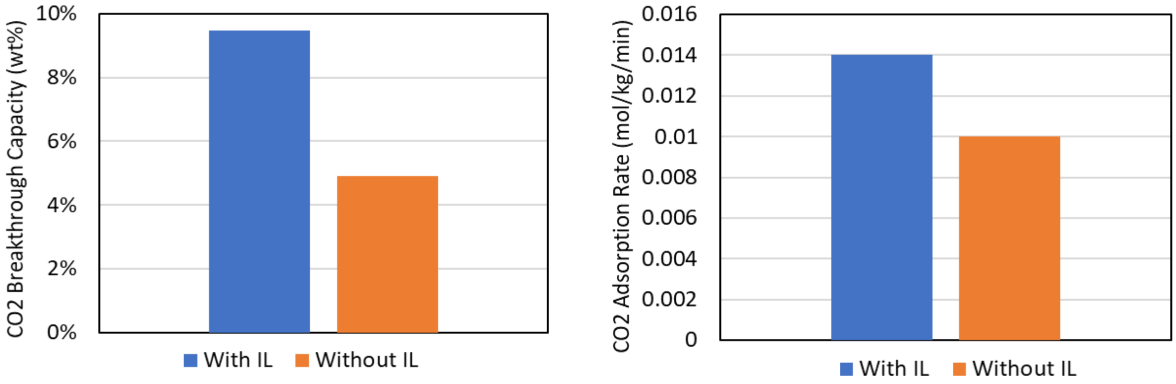

Figure E-1: Performance comparison of FS-PEI sorbents with and without ionic liquid catalyst

The combination of an industrially utilized amine-based sorbent with a highly active catalyst to form a new class of materials for DAC provides a technically viable pathway for reducing the cost of DAC to <\$100/tonne of CO_2. Laboratory measurements show that the silica/PEI (polyethyleneimine) sorbents with 100 ppm of ionic liquid catalyst have almost 100% higher CO_2 cyclic capacity and 40% higher adsorption rate. Generally, CO_2 desorption occurred at higher temperatures with a rate of desorption 10 times faster than adsorption (which occurred at ambient conditions). Therefore, adsorption rate is a much more important factor in the cost of DAC because it is directly linked to the CAPEX of the total system and the cycle time (i.e., sorbent productivity in ton/day of CO_2 captured per unit volume of the air contactor).

An initial process design, coupled with techno-economic analysis, based on optimal experimental results and preliminary resulting from structured sorbent testing, showed a path to lower the DAC cost from the current cost of over \$200/tonne CO_2 to less than \$100/tonne with a scale-up, mature state of the technology, with projected material and process improvements. These results demonstrate the effectiveness of the catalyst in silica/PEI sorbents in enhancing sorbents' CO_2 working capacity, in (a) increasing the rate of adsorption and desorption, and (b) in lowering the CAPEX and OPEX of the DAC system employing the ionic liquid catalyzed sorbents.

Introduction and Objectives

Introduction

This project was aimed at using an ionic liquid as catalyst in amine-based direct air capture (DAC) sorbents with enhanced adsorption and desorption kinetics to lower the cost of DAC. Current estimates for DAC suggest that it can cost between \$300 - \$1,500/tonne of CO_2 captured. The key to lower the cost of DAC rests on three factors: 1) the energy required for releasing CO_2 from the capture agents, 2) reduce the size of the air contactor thus lowering the capital costs, and 3) the pressure required to move the air through the capture device.

To achieve this objective, we developed amine-doped solid sorbents catalyzed by our ionic liquids that have the potential to increase the CO_2 adsorption and desorption rates. In laboratory tests, the proprietary ionic liquid catalyst was shown to increase the CO_2 desorption rate by up to 60 times at 85°C when added in ppm levels as a catalyst to an MEA solvent. Similar results for a different solvent have been peer-reviewed and published in the Nature Communications journal in mid-2018. This transformational discovery enables solvent regeneration at much lower temperatures to dramatically reduce energy consumption, amine degradation, and solvent emissions.

In this project, we aimed to (1) scale up the ionic liquid catalyst for amine-based sorbents for improved desorption and absorption kinetics, (2) evaluate the catalyzed amine-based sorbents for direct CO_2 capture process to determine CO_2 adsorption and desorption rates and energy requirements, and (3) based on the experimental results, develop a conceptual process design to perform a preliminary economic assessment to evaluate the potential for DAC process cost reduction using the catalyzed sorbents.

The anticipated benefits of this novel sorbent process are 1) greater than 20% reduction in the energy required for sorbent regeneration, 2) the potential to use waste heat for sorbent regeneration, 3) drastic increase in sorbent lifetime/stability due to lower regeneration temperature, and 4) lower sorbent replacement cost due to the increased stability.

Technical Approach

To achieve the project objectives**, (1) we scaled up the ionic liquid catalyst, incorporated the catalyst into polyethyleneimine (PEI) on silica DAC sorbents, (2) tested catalyzed sorbents in DAC process in the lab, and (3) performed a preliminary cost analysis of the DAC process**. The technical approach utilized by the project team is detailed below.

Approach to Achieve Project Objectives

The primary objective of this project was to develop sorbent materials for DAC applications with the required properties and improvement to ultimately lower the cost of DAC. By the selection of amine-based sorbents as starting materials incorporating the novel catalysts into these adsorbents, the following were the expected properties of the sorbent materials:

[1] To be highly selective for CO_2 from other components in air
[2] To have improved CO_2 adsorption and desorption kinetics due to incorporation of the novel catalysts
[3] To be made into structured adsorbent beds with low pressure drop
[4] To be stable in high humidity air, and most importantly
[5] To be regenerated at temperatures lower than the SOTA sorbents for reduced energy consumption

Project Team

The project team consisted of Susteon Inc., SoCalGas, and UWy with extensive experience in CO_2 capture material development and testing, with existing laboratory equipment to achieve the objectives of this project. Furthermore, this project was built on our catalysts that have been shown in the laboratory to be effective in increasing the desorption kinetics by several orders of magnitude and, builds on SOTA amine-based sorbents that are proven to be effective for DAC applications. This combination of these two proven concepts held the potential to yield next generation sorbents to significantly lower the cost of DAC.

Experimental Setup

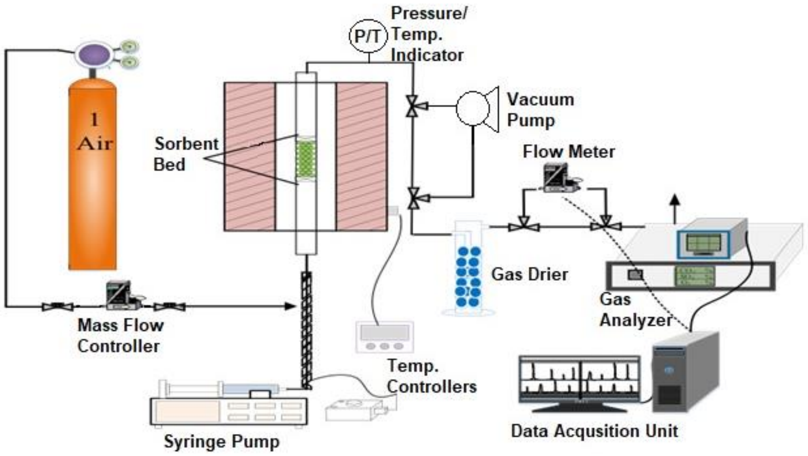

Figure 1: Schematic diagram of the CO_2 capture test setup

The laboratory set-up with flow, temperature, and pressure control capabilities used for DAC sorbent testing is illustrated in **Figure 1**. Sorption tests were performed in a quartz tube reactor with 1 to 10 g of sorbent. The sorbent was placed in the quartz tube using quartz wool as the bed holders and contained within a temperature-controlled furnace (Thermo Corporation, TF55030A-1). Air supplied by an air cylinder at a flow rate of 500-3,500 mL/min was used for all CO_2 sorption tests. Water was introduced into the inlet gas stream by a syringe pump to adjust the relative humidity of air. The effluent gas from the DAC reactor passed through a moisture removal unit to remove moisture and the CO_2 concentration of the gas is then analyzed by a NDIR gas analyzer (California Analytical Instruments Model 703 NDIR/CO_2 and O_2 Gas Analyzer). The measured CO_2 concentration was recorded every second with a data acquisition unit. The CO_2 adsorption amounts were calculated by integrating the recorded CO_2/time profiles. The sorption experiment was stopped when the sorbent is saturated with CO_2. The CO_2 desorption test was performed using the same experimental setup. When the CO_2 sorption test was completed, the reactor temperature was increased to the desorption temperature in the range of 85-110ºC. The desorbed CO_2 from DAC reactor was mixed with N_2 carrier gas after DAC reactor. The desorbed gas was dried by the moisture removal unit and the CO_2 concentration of the gas is then analyzed by the gas analyzer. The variables and approximate range of conditions for testing are shown in the **Table 1**.

Table 1: Variables and their approximate ranges for laboratory testing of DAC sorbents

Variables	Unit	Ranges
Adsorption Temp.	°C	30 to 50°C
Adsorption Pressure	bar-a	1.0 to 1.5
Adsorption Weight Hourly Space Velocity	hr^{-1}	10 to 300
Desorption Temp.	°C	80 to 110
Desorption	bar-a	0.2 to 1.0

The major concerns about the ionic liquid catalyst are its cost and availability. Fortunately, the major starting materials can be obtained from several sources such as BASF Corporation and the cost of the ionic liquid catalyst is estimated at ~$50/kg. A dosage of 2,000 to 3,000 ppm was shown to significantly enhance CO_2 absorption and desorption rates for aqueous 20% MEA solution. In solid DAC sorbents, the amount of catalyst should be substantially less due to absence of water and catalyst amine proximity. Therefore, the added cost of the DAC sorbent with catalyst will be less than 10¢ for 1 kg of sorbent, which is negligible in comparison with the cost of the state-of-the-art CO_2 solid amine-based sorbent, which is ~$15/kg.

While the DAC contactor design is outside the scope of this project, the ionic liquid catalyzed sorbents can be applied to existing low-pressure-drop contactor designs such as those used by Climeworks, Svante, and Global Thermostat. A high-level process was designed based on certain form of contactors with required fans, heat exchangers, steam generator, and heat recovery unit such that the process can be modeled in

AspenPlus™ to obtain process heat and mass balances. This information was useful in sizing the equipment, estimating the utility consumption rate, and the cost of DAC process using the new materials.

Technical Achievements
The major tasks for achieving project objectives are:

- Ionic Liquid Catalyst Synthesis and Characterization
- DAC Sorbent Synthesis and Characterization
- Sorbent Testing
- Data Analysis
- Process Design, TEA, and EH&S Analysis

In execution of these tasks, we have accomplished the following and met all the objectives of the project.

[1] We have successfully scaled up the catalyst synthesis process from gram quantities to kilogram quantities. The large batch catalyst was characterized and compared with small laboratory synthesized samples and no difference was found in the chemical and physical properties between the two samples.
[2] Susteon's patented ionic liquid catalyst enhanced the cyclic CO_2 adsorption capacity of silica-PEI DAC sorbents by 2 to 4 times with much longer breakthrough times.
[3] Ionic liquid catalyzed DAC sorbent showed stable performance in multi-cycle testing.
[4] Ionic liquid catalyzed sorbents demonstrated a 40% to 60% higher rate of CO_2 adsorption which is the most important factor in lowering the CAPEX of the DAC system.
[5] The project team performed a DAC process design and techno-economic analysis using lab data to determine the cost of DAC.

Technical Progress

Task 1.0: Project Management

Subtask 1.1 – Project Management

The project was managed in accordance with a Project Management Plan and met all technical, schedule and budget objectives and requirements. The team, consisting of Susteon, University of Wyoming and SoCalGas, met regularly to coordinate activities and discuss issues to effectively accomplish the project objectives. The results, work plan and products of the project were well documented in quarterly and topic report as well as in briefings to the DOE project management team.

Subtask 1.2 – Technology Maturation Plan

At the start of the project, the project team developed an initial Technology Maturation Plan (TMP) that described the current technology readiness level (TRL) of the ionic liquid catalyzed DAC sorbent technology, related the planned project work to maturation of this technology, described the expected TRL at the end of the project, and described any known post-project research and development necessary to further mature the technology. The initial TMP was submitted as a topic report on time to DOE. In the initial TMP, we had concluded that **the technology readiness level (TRL) for the ionic liquid catalyzed DAC sorbent was TRL 2 at the start of the project**. The basic principles of the ionic liquid catalysis to enhance CO_2 reaction with amines were observed in the preliminary tests which were carried out to measure CO_2 absorption and desorption performance with 20 wt% MEA solution in the laboratory. The data from these measurements clearly showed the catalytic effect of ionic liquid on CO_2 absorption and desorption rates. The utilization of ionic liquid catalyst on solid sorbents for DAC application is a natural extension of this experimental observation.

A final TMP (Appendix A) was also submitted on time and within 90 days of completion of the project. At the completion of the 2-year long research and laboratory testing work, critical technology elements have reached a **TRL level of 3** (**TRL 3**). We scaled up the ionic liquid synthesis process from gram level in the laboratory to kilogram level in pilot scale. The ionic liquid catalyzed PEI/fumed silica sorbent was tested in the laboratory using simulated air with about 400 ppm CO_2 for CO_2 breakthrough capacity and cyclic absorption/desorption behavior.

Task 2.0: Catalysts Preparation and Characterization

The procedure for catalyst synthesis in the lab-scale was developed. Based on this procedure, UWy assembled a setup for catalyst synthesis. The synthesis process was done in a batch mode. UWy procured the feedstocks required for catalyst synthesis. UWy also developed the catalyst characterization protocols and quality control procedures.

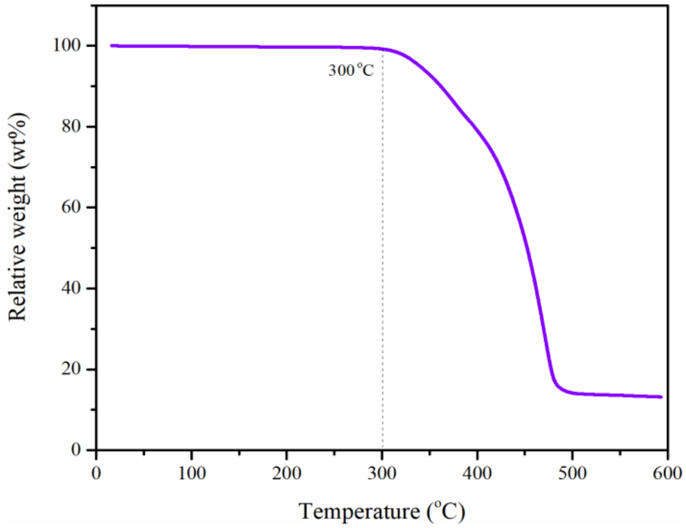

Figure 2: TGA confirming catalyst thermally stability at 300 °C

The produced batch of ionic liquid catalyst was characterized with TGA, ^{1}H-NMR, ^{13}C-NMR, FT-IR, and ESI-MS. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker spectrometer (400 MHz) in the solvent of DMSO. ESI-MS spectrum was obtained by ultra-high-resolution electrospray time-of-flight mass spectrometry (Bruker microTOF II, Germany). FT-IR spectra were collected using a Thermo Nicolet Magna-IR 760 spectrometer with a resolution of 4 cm^{-1} by scanning 32 times from 4000 to 400 cm^{-1}. Thermogravimetric analysis (TGA) of IL was obtained using a TA Instruments SDT Q600 apparatus with a heating ramp of 10 °C min^{-1} at the temperature of 20-600 °C with a nitrogen flow rate of 100 mL min^{-1}. The characterization results showed that the ionic liquid batch met all chemical specifications. The TGA results, shown in **Figure 2**, indicate that the ionic liquid catalyst is stable up to 300°C.

Ionic liquid catalyst synthesis was scaled up by Susteon to kilogram scale. Susteon procured raw materials and produced about 20 kg of ionic liquid catalyst for testing. The large batch of catalyst was also characterized using the same techniques as the lab synthesized IL catalyst. The produced batch of IL catalyst was characterized with TGA, ^{1}H-NMR, ^{13}C-NMR, FT-IR, and ESI-MS. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker spectrometer (400 MHz) in the solvent of DMSO. ESI-MS spectrum was obtained by ultra-high-resolution electrospray time-of-flight mass spectrometry (Bruker microTOF II, Germany). FT-IR spectra were collected using a Thermo Nicolet Magna-IR 760 spectrometer with a resolution of 4 cm^{-1} by scanning 32 times from 4000 to 400 cm^{-1}. Thermogravimetric analysis (TGA) of IL was obtained using a TA Instruments SDT Q600 apparatus with a heating ramp of 10 °C min^{-1} at the temperature of 20-600 °C with a nitrogen flow rate of 100 mL min^{-1}. The characterization results showed that the IL batch met all chemical specifications. The results of these characterization showed that the large batch of IL is identical to the lab batch.

Task 3.0: Sorbent Synthesis and Characterization

Selected sorbent substrate as SBA-15, MCM41, alumina, and fumed silica were used as support to produce the DAC sorbent. Polyethyleneimine (PEI) was used as an active amine. Several sorbents, listed in **Table 2**, were synthesized, and dry baseline tests were done. A wet baseline test was also performed for a selected sorbent. UWy developed a promising method to control feed gas moisture. It provided an opportunity to test sorbent performance at different air moisture levels and determine energy consumption and sorbent lifetime. Sorbents of varying amount of PEI, varying amount of ionic liquid catalyst, and different synthesis methods were prepared in this task and tested under relevant DAC conditions. Test results are summarized in Tasks 4 and 5 sections.

Table 2: CO_2 capture capacity for different sorbent – baseline data (dry condition)

Sorbent Composition	CO_2 Capacity (µmol/g)	CO_2 Capacity (wt%)
20% PEI/$FSiO_2$	1.05	4.62
20% PEI/$FSiO_2$ (Humid conditions)	1.36	5.98
20 % PEI/SBA15	0.86	5.98
20% PEI/MCM-41	0.18	3.78
20% PEI/γ-Al_2O_3-nano	0.61	0.79
20% PEI/ γ-Al_2O_3-ACP	0.58	2.68
20% PEI/ZSM-5	0.26	2.55

Tasks 4.0 & 5.0: Sorbent Testing and Data Analysis

UWy assembled a CO_2 capture setup for sorbent testing. Photo of the experimental setup for CO_2 sorption, desorption, and breakthrough tests is shown in **Figure 3**. Sorption tests were performed in a quartz tube reactor, and the prepared sorbent was loaded at the middle of the tube with quartz wool as the holder. The tube was then placed in a furnace, and the temperature was controlled and monitored by the temperature controller and a thermocouple.

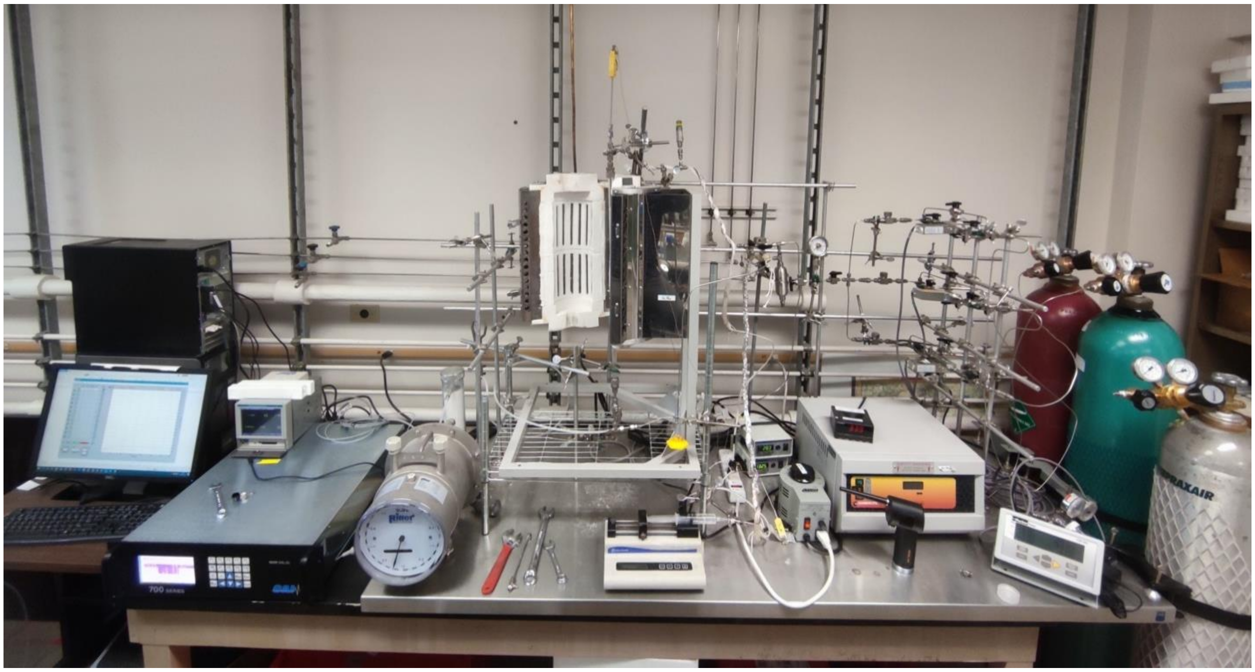

Figure 3: Picture of sorbent CO_2 capture test setup

A CO_2 and N_2 mixture gas stream were introduced into the tube reactor from the gas cylinders for all CO_2 sorption tests. The CO_2 concentration and flow rate were adjusted and monitored by mass flow controllers and the control panel. Water was introduced into the feed gas stream and into the tube reactor whenever needed by a syringe pump through a heating tape covered gas line to change the inlet gas humidity. The effluent gas leaving the reactor entered a water trap before being analyzed by the gas analyzer (700 Series, NDIR CO_2/O_2 analyzer, CAI). The data acquisition system recorded the measured CO_2 concentration and the effluent gas flow rate. The CO_2 adsorption amounts were calculated by integrating the registered CO_2 concentration vs. time profile. The sorption experiment was stopped when the sorbent is saturated with CO_2 indicated by the CO_2 concentration at the effluent gas.

The CO_2 desorption test was performed using the same experimental setup. When the sorption cycle was finished, the reactor temperature was increased to the desorption temperature, and then a purge gas was introduced into the reactor for desorption. The flow rate and CO_2 concentration were recorded during the desorption and conditioning cycles.

The baseline sorbents synthesized in Task 3 were tested using the experimental system illustrated in **Figure 3**. The impact of moisture on the CO_2 capture capacity of the sorbent was also investigated. **Figure 4** illustrates the breakthrough curve of each sorbent synthesized in Task 3 and tested in Task 4. 300 mg sorbent was used for each test. The CO_2 concentration of the feed gas was 400 ppm CO_2 (balance N_2) with a total flow rate of 1,030 ml/min, and all adsorption tests were performed at 25 ºC.

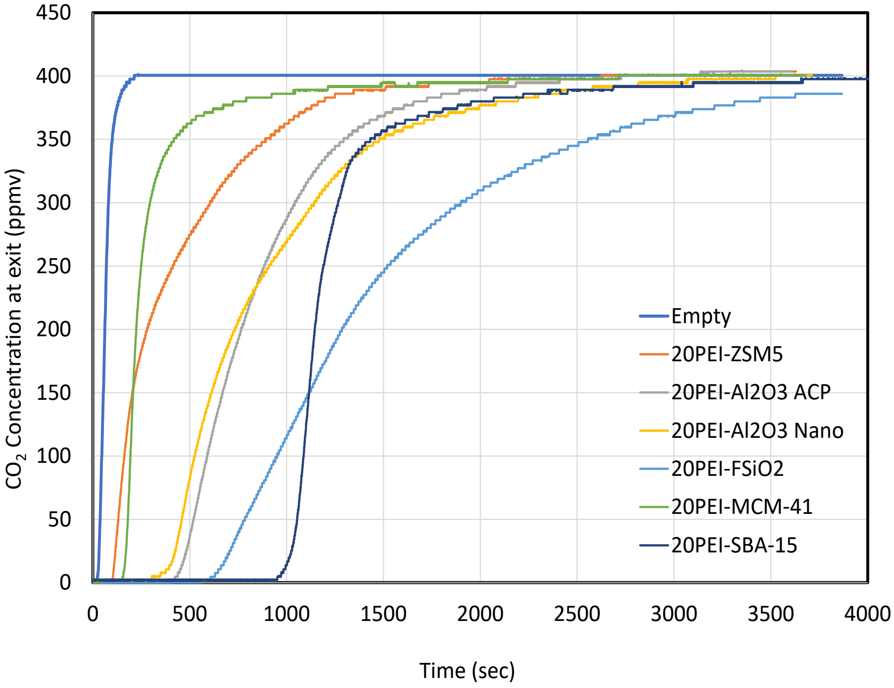

Figure 4: The breakthrough curves of different sorbents – Baseline

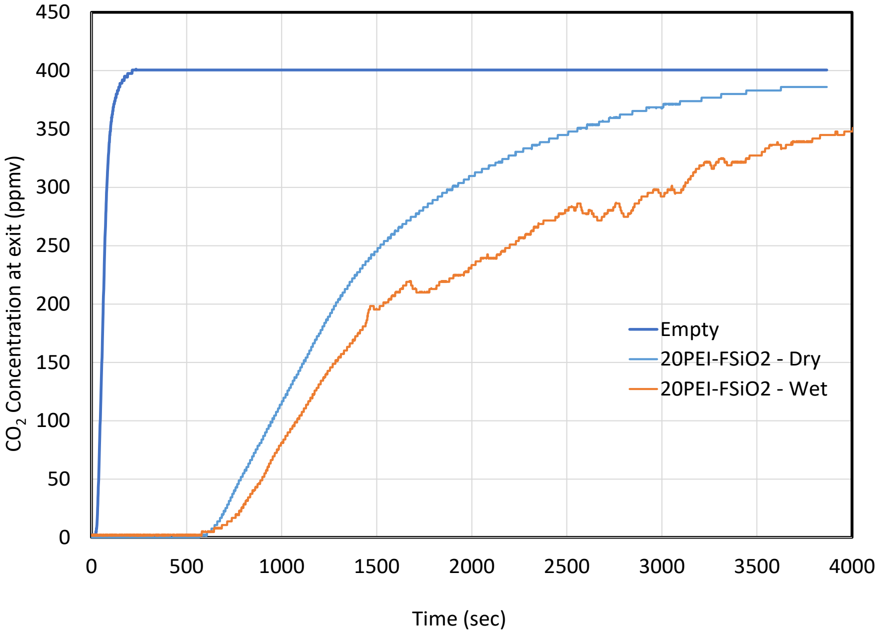

Figure 5: The breakthrough curves of 20% PEI/FSiO$_2$ in dry and wet conditions.

Based on breakthrough curves, the CO_2 capture capacity of each sorbent was determined and summarized in **Table 2**. The impact of humidity also was investigated using the moisture control method. The breakthrough curves of 20% PEI/$FSiO_2$ (fumed silica) under dry and wet (~100% humidity) conditions are illustrated in **Figure 5**. This figure shows that the CO_2 breakthrough and equilibrium capacity is higher under humid conditions for the same sorbent. This is helpful because air is never dry and contains much more water than CO_2. As shown in **Table 2** earlier, under humid conditions, the CO_2 capacity increased from 4.62 to 5.98 wt%, which is a 29% increase.

While the fumed silica ($FSiO_2$) and SBA-15 supported sorbents showed the highest capacity and breakthrough time, the optimal PEI loading in the sorbent needed to be determined with its impact on the CO_2 capture performance. Ionic liquid catalyst was added to Susteon's PEI doped silica sorbent at a concentration of 100 ppm via incipient wetness impregnation and the results are shown in **Figure 6**. The catalyst increased the breakthrough time by more than 100% for CO_2 adsorption without changing the sorbent's equilibrium CO_2 adsorption capacity as expected. The ionic liquid catalyst can only improve the reaction kinetics, not the reaction equilibrium, thus this result is consistent with our hypothesis.

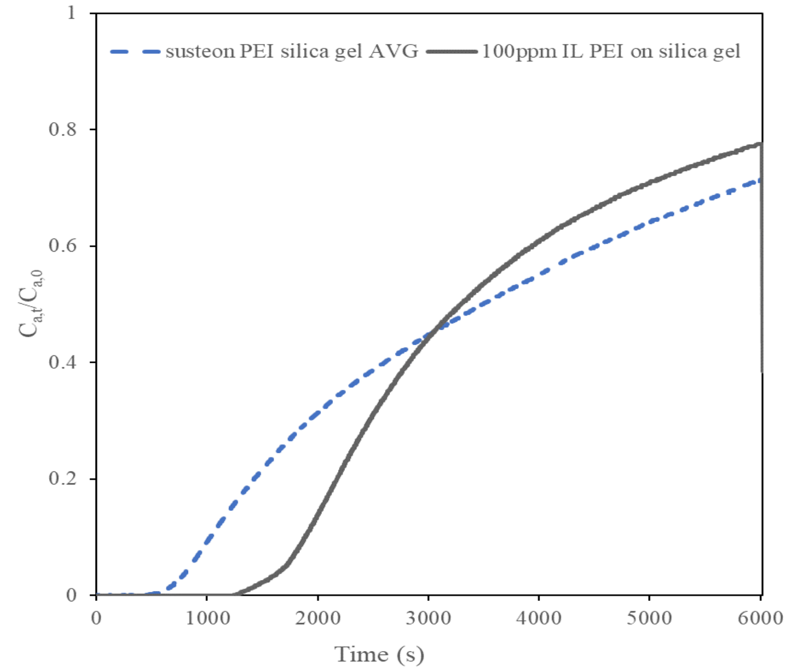

Figure 6: Comparison between Susteon PEI on silica gel (no catalyst) and 100ppm IL catalyst.
[Adsorptions: 0.4g sorbent; 400ppm CO_2*; 500mL/min; 25°C; where* $C_{a,0}$ *is* CO_2 *concentration at t=0 during adsorption]*

The adsorption breakthrough curves for 30, 40, and 50 wt% PEI on fumed silica and the effect of the IL catalyst on these sorbents are shown in **Figure 7**. Breakthrough adsorption time for all three (no catalyst) are within a similar range between 2500 and 3000 seconds. Addition of the 100ppm IL catalyst significantly increased the adsorption performance of 30, 40, and 50 wt% PEI on fumed silica. For 50 wt% PEI, the catalyst increased the breakthrough adsorption time from 2,900 seconds to 4,000 seconds. With the addition of catalyst, the 100% adsorption time rank of the samples are 50 wt% PEI > 40 wt% PEI > 30 wt% PEI. Test results showed that fumed silica is a good support for PEI and that ionic liquid catalyst is effective in enhancing DAC sorbent performance.

To further determine the impact of ionic liquid catalyst on DAC performance, ionic liquid catalyst was added to PEI doped silica sorbent at a concentration of 100 ppm via incipient wetness impregnation. The catalyzed sorbents were tested for CO_2 breakthrough under DAC conditions and the results are shown in

Figure 8. The ionic liquid catalyst significantly increased the CO_2 breakthrough time and thus increased the breakthrough CO_2 capacity by more than 500% for CO_2 adsorption. It should be noted that ionic liquid catalyst does not increase overall sorbent equilibrium CO_2 capacity.

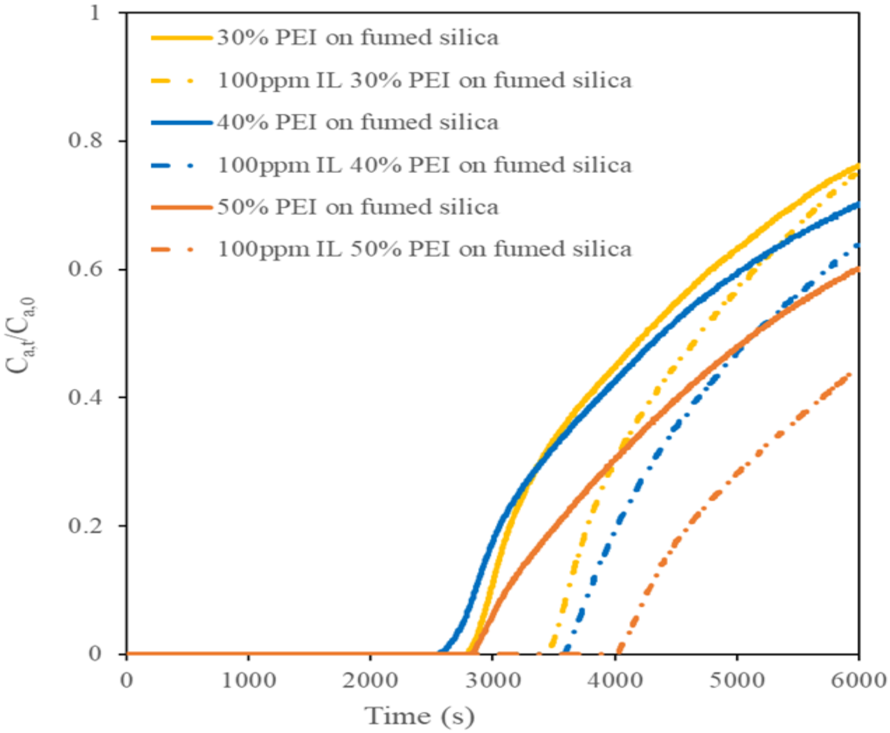

Figure 7: Different PEI loading amounts on fumed silica.

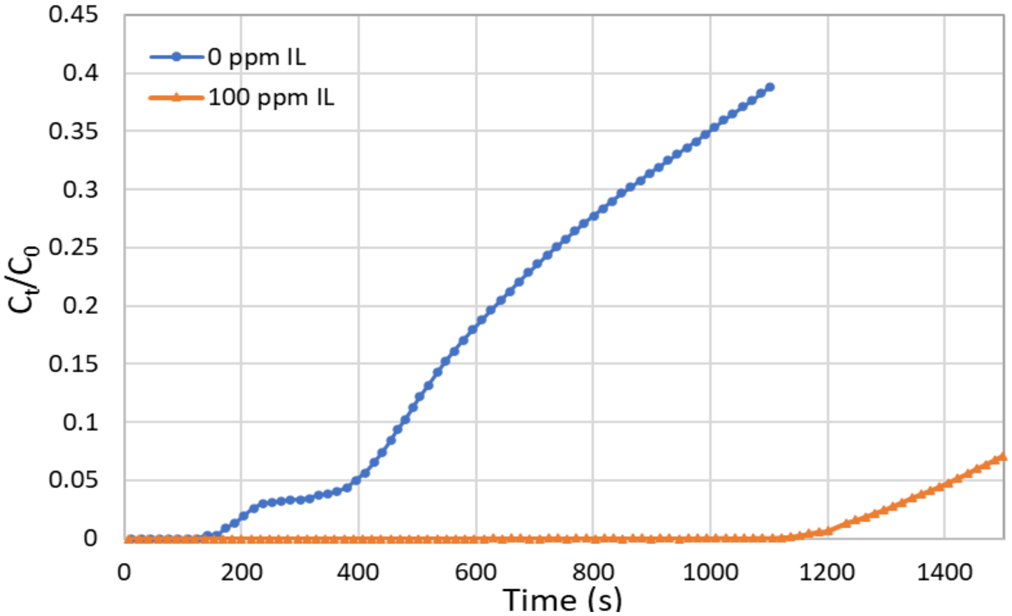

Figure 8: Breakthrough behavior of PEI on silica gel with and without ionic liquid catalyst.
[Adsorptions: 0.4g sorbent; 400ppm CO_2; Flow rate: 500mL/min; 25°C]

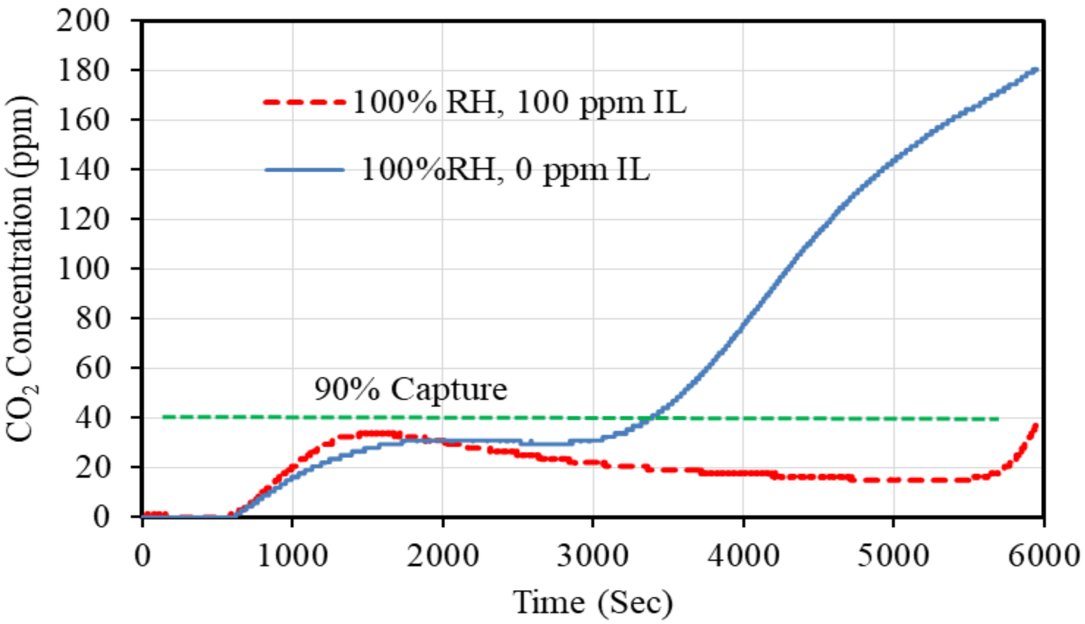

Figure 9: Breakthrough testing results of fumed silica with 50 wt% PEI with and without IL catalyst.

As shown in **Figure 9**, at 90% CO_2 capture (~40 ppm exit gas CO_2) with air at 100% relative humidity, the breakthrough times for 50 wt.% PEI on fumed silica with and without ionic liquid catalyst are 3,300 seconds and 6,000 seconds, respectively, reflecting a near 100% increase in breakthrough CO_2 capacity.

To study the impact of ionic liquid catalyst concentration, we prepared 50 wt% PEI on fumed silica with 0, 100, 200 and 300 ppm by weight of ionic liquid catalyst and measured their CO_2 breakthrough behavior. The data from these measurements are shown in **Figure 10.** As seen in this figure, FS-PEI-50 with 100 ppm catalyst showed the best adsorption performance with the longest breakthrough time. CO_2 adsorption performance declined slightly when the catalyst loading was increased from 100 ppm to 200 ppm. As the catalyst loading increased to 300 ppm, CO_2 adsorption performance showed no improvement as compared with that of the sorbent without catalyst. Therefore, catalyst loading needs to stay within 100 to 200 ppm range. Through extensive optimization and testing, we selected 200 ppm as the final catalyst loading and 35 wt% PEI on fumed silica as the final sorbent composition. The selection of 35 wt% PEI resulted from testing of sorbents with 20, 35 and 50 wt% PEI and from consideration of PEI loading on PEI accessibility.

To study the effect of air relative humidity (RH) on sorbent performance, we tested catalyzed sorbents at 0, 75, and 100% RH. **Figure 11** shows the effect of relative humidity on 35 wt% PEI on fumed silica with 200 ppm IL catalyst. The breakthrough time (= 90% CO_2 recovery) of 0.5 g of sorbent at 500 mL/min air flow rate and 0% humidity was about 5,350 seconds (breakthrough CO_2 capacity of 7.0 wt%). With 100% humidity, the breakthrough time increased to 8,300 seconds (~55% increase from dry air, and a breakthrough capacity of 10.9 wt%). At 75% relative humidity, the breakthrough time increased to 8,450 seconds (~58% increase from dry air, and a breakthrough capacity of 11.1 wt%). It should be noted that the breakthrough curve under dry as well as under humid conditions was quite sharp. Sharp breakthrough curves make the process design, for maximum sorbent utilization and lower energy consumption, much easier.

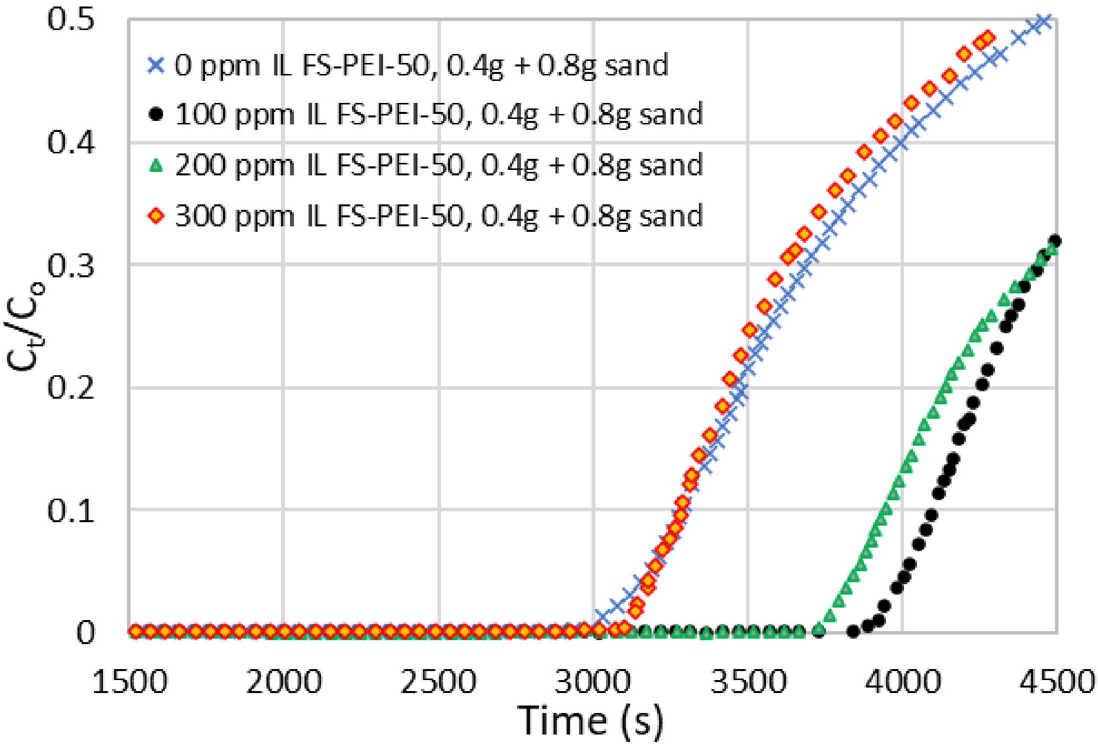

Figure 10: Effect of catalyst concentrations

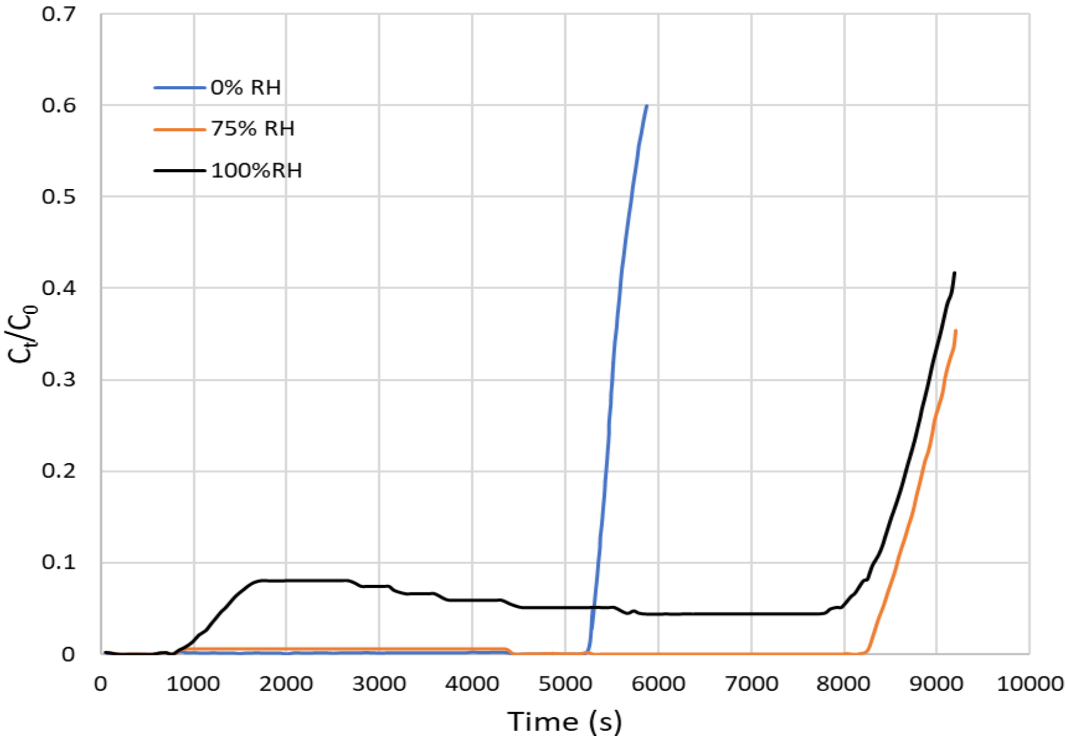

Figure 11: Effect of relative humidity on CO_2 adsorption.

The cyclic stability of sorbent with both dry and humid air was investigated. These cyclic studies consisted of an adsorption step in 400 ppm CO_2 with 0% or 100% RH followed by a heating step to 110°C in flowing N_2 to facilitate endothermic desorption of CO_2. The cycle results for 100 ppm ionic liquid catalyzed FS-PEI with 50 wt% PEI loading are shown in **Figure 12.** CO_2 breakthrough capacity remained stable with minor variations under both dry and humid conditions. The breakthrough times and hence breakthrough CO_2 capacity were significantly greater in the presence of humidity. Fewer cycles were run under humid conditions due to the long breakthrough time. When the sorbents were coated on monolith with high dispersion of active capture agent, relatively high space velocities were used to obtain a breakthrough time in the 45-75 min range. The data shown in **Figure 12** confirms that CO_2 can be adsorbed and desorbed from the ionic liquid catalyzed sorbents with good cyclic stability and high cycle capacity.

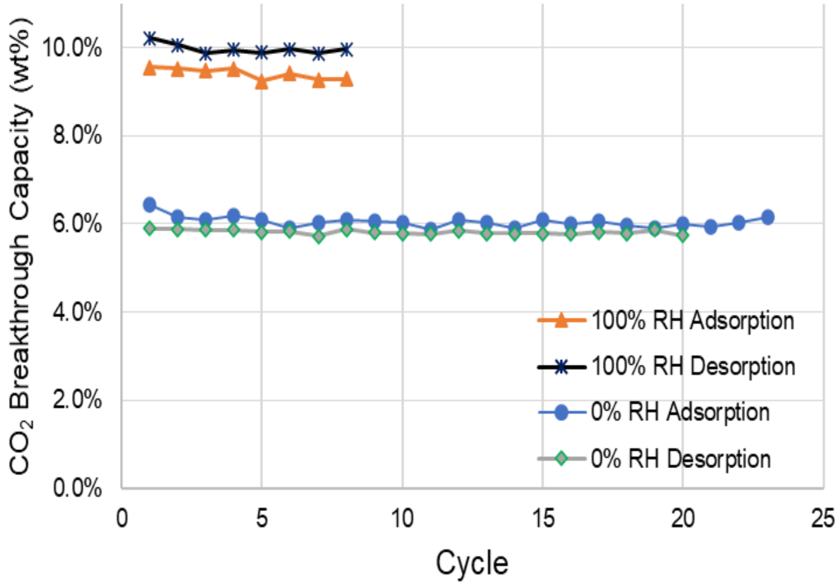

Figure 12: Cyclic stability of 100 ppm IL catalyzed sorbent.

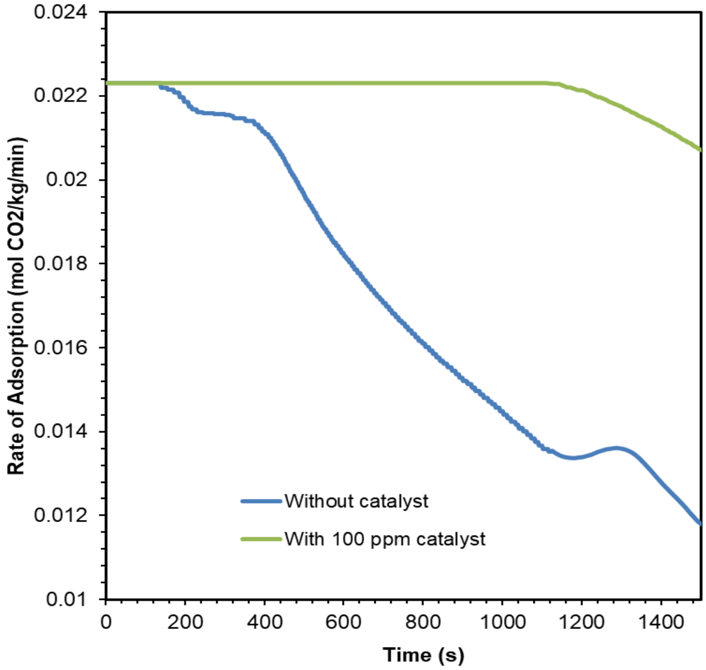

Figure 13*:* CO_2 adsorption rate as a function of time on stream. *Absorption: 0.4 g sorbent; 400 ppm CO_2; 500 mL/min gas flow; 25°C; RH: 60% at 20°C; GHSV 38,000 h^{-1}*

The adsorption rates with and without ionic liquid addition to the sorbent were determined as a function of time on stream as shown in **Figure 13**. These results show that the catalyzed PEI sorbent stays at the higher adsorption rate for much longer time (~8 time longer), and that the adsorption rate of the catalyzed sorbent decreases much more slowly. For example, the adsorption rate of the catalyzed sorbent deceased from 0.0223 to 0.0207 (mol/kg/min) in 1,500 seconds with a rate of decrease of 1.07 x 10^{-6}, and that of the un-catalyzed sorbent decreased from 0.0223 to 0.0118 with a rate of decrease 7 times greater at 7.0 x 10^{-6} than that of the catalyzed sorbent.

As shown in **Figure 14**, higher inlet air space velocity led to higher rates of CO_2 adsorption (as high as 0.1 mol/kg/min), indicating that the adsorption is mass transfer limited and not limited by reaction kinetics. Comparison of adsorption and desorption rates for sorbents with and without ionic liquid is shown in **Table 3** for one of the PEI sorbents tested. The desorption rate of sorbent without ionic liquid catalyst was not measured. However, the desorption rate was observed to be about 10 times faster than that of the corresponding adsorption rate. It should be noted that the inlet air space velocity was constrained by the pressure drop across the air contactor. Therefore, CO_2 adsorption rate is also constrained by the same.

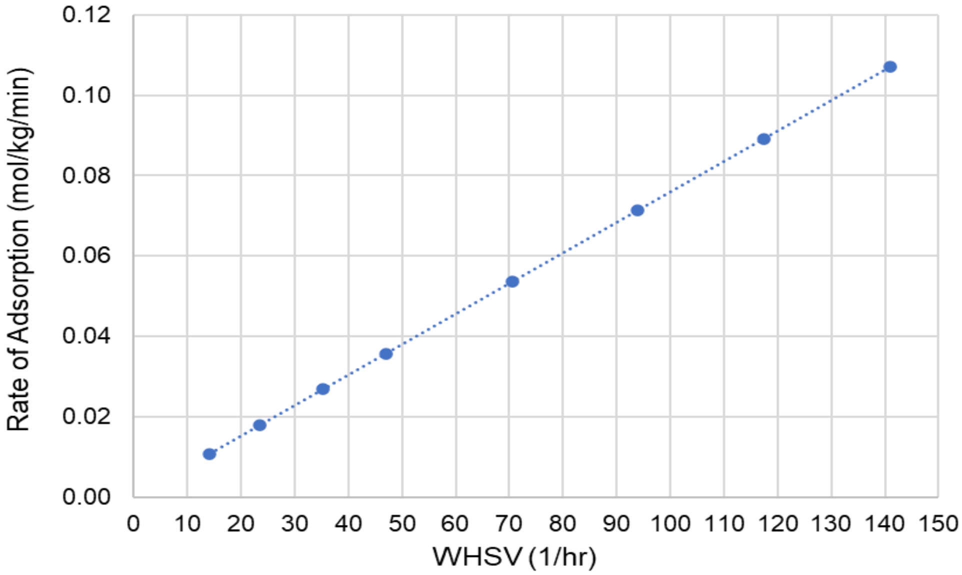

Figure 14: CO_2 adsorption rate as a function of gas weight hourly space velocity (WHSV)

Table 3: Comparison of adsorption and desorption rates

Parameters	Unit	With IL	Without IL
IL Catalyst	*ppmw*	100	0
CO_2 capacity at breakthrough	*wt.%*	9.49%	4.91%
Rate of CO_2 adsorption	*mol/kg/min*	0.014	0.010
Rate of CO_2 adsorption	*g CO_2/g sorbent/s*	1.23E-05	8.80E-06
Rate of CO_2 Desorption	*g CO_2/g sorbent/s*	1.14E-04	-

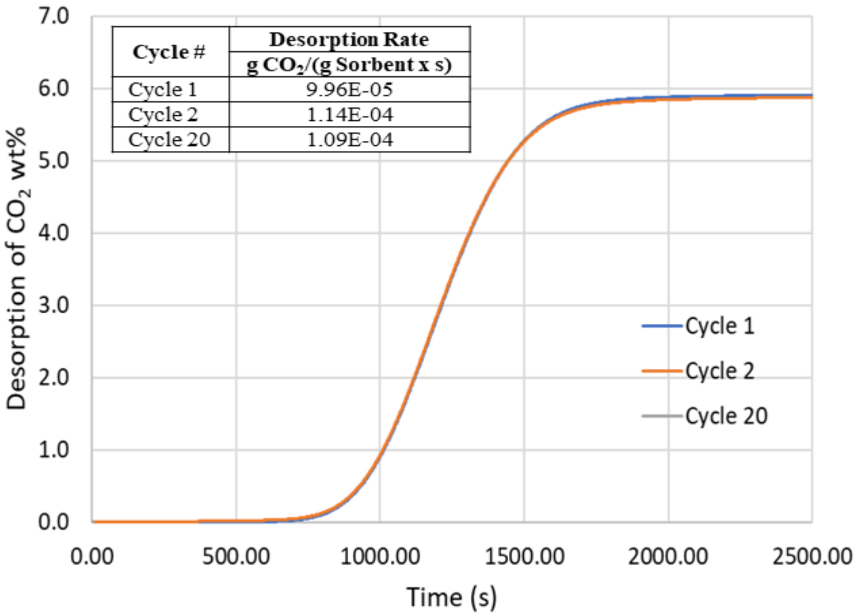
Cycle #	Desorption Rate g CO_2/(g Sorbent x s)
Cycle 1	9.96E-05
Cycle 2	1.14E-04
Cycle 20	1.09E-04

Figure 15: CO_2 desorption curves for three cycles

CO_2 desorption rates were also measured in cyclic tests for 20 cycles as shown in **Figure 15**. These data confirm that the rate of desorption is stable in the 20 cycles and about 10 times faster than the rate of adsorption, which is very encouraging for scalability of this process. Laboratory data have clearly demonstrated the proposed sorbent composition for DAC application and provided clear proof that these sorbent compositions have been successfully developed on commercially available fumed silica support. Furthermore, the ionic liquid catalyst was scaled up to a kilogram scale. We produced about 20 kg of this catalyst at a commercial manufacturer for several testing campaigns. The best sorbent composition identified was **polyethyleneimine (PEI) on fumed silica with 200 ppm ionic liquid catalyst**. This sorbent formulation was shown to have a CO_2 breakthrough capacity of 11 wt.% [in laboratory tests with air at 75% relative humidity (RH)].

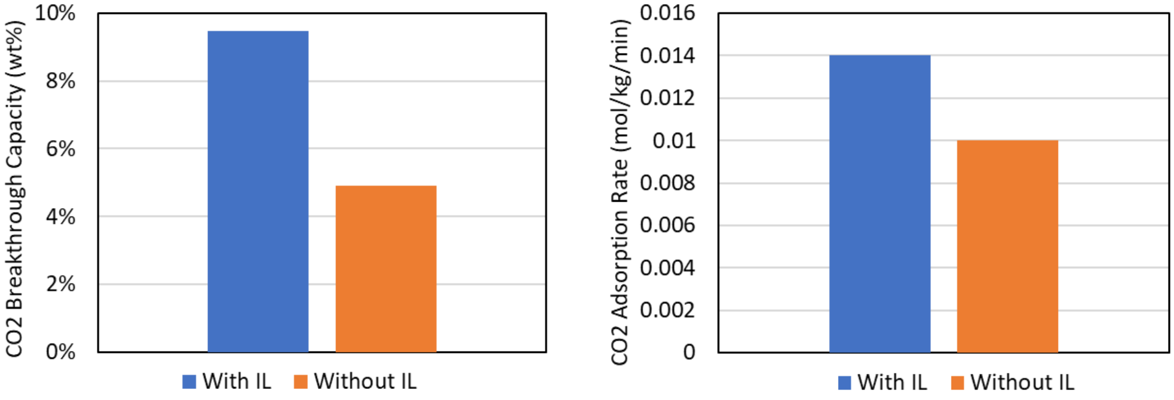

Figure 16: Performance comparison of FS-PEI sorbents with and without ionic liquid catalyst

To summary, the ionic liquid catalyzed silica/PEI sorbents CO_2 working capacity and rate of CO_2 adsorption are shown in **Figure 16**. The sorbents have the attributes required for lowering the overall cost of DAC with high CO_2 capacity and high rate of adsorption.

Task 6.0: Process Design and EH&S Analysis

DAC Plant Size

Given the low TRL and the risks associated with scale-up of the DAC technology, 100 tonnes CO_2 per day (41,515 tonnes/year) was selected as the design for the base case study. In our sensitivity studies, we scaled up the results to the minimum 2018 45Q threshold for tax credit qualification of 100,000 tonnes CO_2/yr and up to 1,000,000 tonnes CO_2/yr. Since this is a net removal target, the gross CO_2 removal rate from the air (and, therefore, the actual CO_2 product flow rate from the DAC plant) will be higher. The excess CO_2 required to be removed from the air to meet the net removal target will be dependent on many factors, including electrical auxiliary load, plant efficiency, system configuration (e.g., electricity generation on-site versus purchased power from the grid), and sorbent performance characteristics.

CO_2 Transport and Storage

The cost of CO_2 transport and storage (T&S) in a deep saline formation was estimated using the Department of Energy (DOE) Office of Fossil Energy and Carbon Management (FECM)/NETL CO_2 Transport Cost Model (CO_2 Transport Cost Model) and the FECM/NETL CO_2 Saline Storage Cost Model (CO_2 Storage Cost Model). The far-right column of Error! Reference source not found. shows the total T&S costs used i n NETL system studies for each plant location rounded to the nearest whole dollar. Only the $10/tonne value is used in this report since all cases are assumed to be in the Midwest.

Table 4: CO_2 transport and storage costs

Location	Basin	Transport (2018 $/tonne)	Storage Cost at 25 Gt (2018 $/tonne)	T&S Value for System Studies**

				(2018 $/tonne)
Midwest	Illinois	2.07	8.32	10
Texas	East Texas		8.66	11
North Dakota	Williston		12.98	15
Montana	Powder River		19.84	22

**The sum of transport and storage costs rounded to the nearest whole dollar

The reduction in the cost of CO_2 transport & storage was considered during scale-up process according to the work of Smith[1] based on global integrated assessment. CO_2 transport costs drop from \$10/tonneCO_2 to \$2/tonneCO_2 when scaled-up to 50 million tonnes of CO_2 per annum as shown in **Figure 17** . CO_2 storage and monitoring cost dropped from \$20/tonneCO_2 to \$8/tonneCO_2 as shown in **Table 5**.

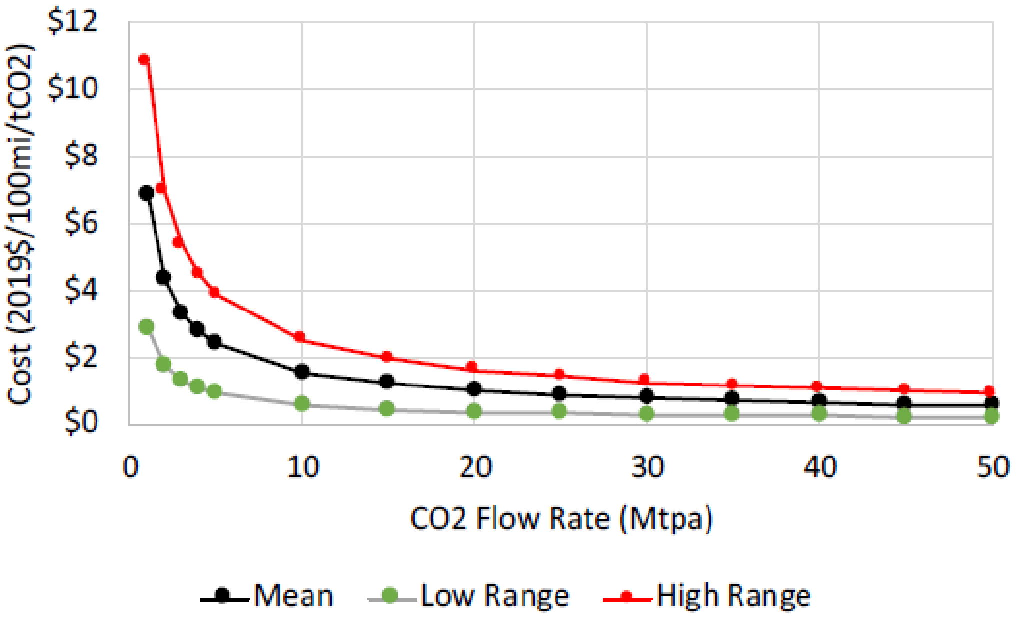

Figure 17: CO_2 transport costs for a 100-mile pipeline in the US in 2019 dollars (Smith, 2015)

Table 5: U.S. storage cost range (2008 \$/$_{\text{tonneCO2}}$)-USDOE (2017) model (Smith, 2015)

Rate Mtpa CO_2	**Low**	**Mean**	**High**	**Mean with extra monitoring**
1	\$6.81	\$11.51	\$16.22	\$19.67
3.2	\$3.67	\$5.59	\$7.51	\$10.59
6	\$3.05	\$4.70	\$6.36	\$8.86
15	\$2.83	\$4.36	\$5.90	\$8.03

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[1] Smith, Erin E. 2015. "The Cost of CO_2 Transport and Storage in Global Integrated Assessment Modeling." University of California, Berkeley.

Cost of CO_2 Capture Estimation Methodology

A framework for the techno-economic analysis, using QGESS[2] for assumptions and estimation methodology, was established to estimate the Levelized Cost of Capture (LCOC). Utilizing this TEA framework, a comparative evaluation between a base case and an improved case was performed. The experimental data from lab work were transferred to the process model set up in AspenPlus™ to finalize the design and integration into the DAC plant flowsheet. The complete process model was used to determine equipment sizing, costs, and utility requirements, which was then used to develop the capital and operating costs inputs for the techno-economic analysis given in the separate sections for individual cases.

Figure 18 shows the levels of estimating capital costs, which consist of bare erected cost (BEC), engineering, procurement, and construction cost (EPCC), total plant cost (TPC), total overnight cost (TOC) and total as-spent cost (TASC). BEC comprises the cost of process equipment, on-site facilities and infrastructure that support the plant (shops, offices, labs, road), and the direct and indirect labor required for its construction and/or installation. The cost of EPC services and contingencies are not included in BEC. EPCC comprises the BEC plus the cost of services provided by the EPC contractor. EPC services include detailed design, contractor permitting, and project/ construction management costs. TPC comprises the EPCC plus project and process contingencies. TOC comprises the TPC plus all other overnight cots, including owner's costs. TOC does not include escalation during construction or interest during construction. TASC is the sum of all capital expenditures as they are incurred during the capital expenditure period including their escalation. TASC also includes interest during construction, comprising interest on debt and a return on equity.

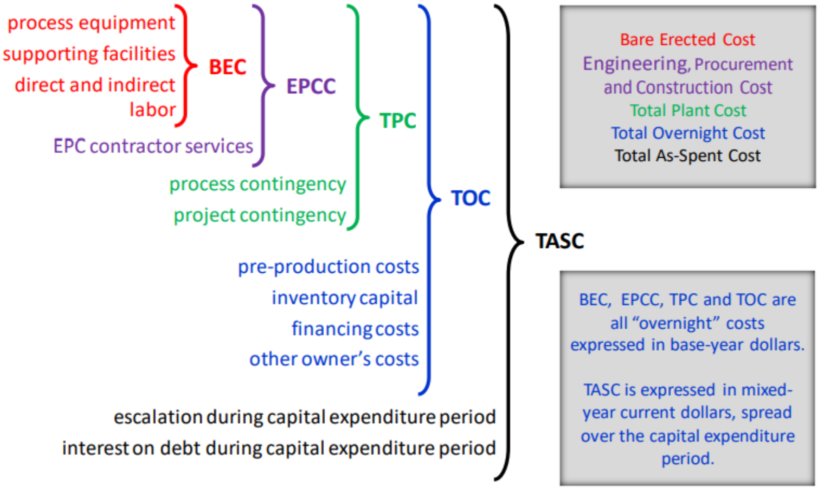

Figure 18: Capital cost levels and their elements

The capital charge portion of COE was calculated by multiplying a fixed charge rate (FCR – 0.068) by the plant total as-spent cost (TASC). TASC was calculated by taking the plant total overnight cost and multiplying by a TASC/TOC ratio (1.093). All costs are reported in 2019 dollars. The equation shown below provides the full calculation for the COC for the DAC plant:

$$Cost\ of\ CO_2\ capture\ (\frac{\$}{tonne}) = \frac{FCR \times TASC + OC_{FIX} + CF \times OC_{VAR}}{CF \times F_{CO_2}}$$

FCR = fixed charge rate taken from the referenced QGESS; TASC = total as-spent cost; OCFIX = the sum of all first year of operation fixed annual operating costs; CF = plant capacity factor (90%), assumed to be constant over the operational period; expressed as a fraction; OC_{VAR} = the sum of all first year-of-operation variable annual operating costs at 100 %; F_{CO2} = annual net CO_2 removed from the atmosphere.

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[2] Quality Guidelines for Energy System Studies: Performing a Techno-Economic Analysis for Power Generation Plants, National Energy Technology Laboratory, DOE/NETL-2015/1726, July 2015, https://netl.doe.gov/energyanalysis/details?id=711

Results of Preliminary TEA

The ionic liquid was shown to be a promising catalyst for CO_2 adsorption and desorption in the DAC process. This catalyst can be integrated into amine-based sorbents for improved sorbent CO_2 capacity and adsorption kinetics. The catalyzed sorbent developed in this work can meet the KPIs required for successful commercial deployment, such as

- Increased sorbent capacity for reduced sorbent mass requirements
- Increased rates of CO_2 adsorption and desorption to shorten cycle time
- Reduced energy consumption for the regeneration of the sorbent

Using the laboratory sorbent test data and initial monolith test results, we completed a preliminary process design and TEA to evaluate the estimated cost of CO_2 capture from our proposed DAC system with the assumptions shown in **Table 6** and using the assumptions and guidelines provided by DOE/NETL. The preliminary TEA results correspond to the "First Module" data. It is when the first large-scale module of multiple air contactors will be fabricated and is considered a first-of-a-kind unit.

Table 6: Assumptions used for TEA

Utility Costs		
Electricity cost	0.06	\$/kWh
Cooling water cost	0.05	\$/m3
Availability	90%	20-yr average
LP steam cost (~60 psig or 4 bar)	7.40	\$/tonne
Material Costs		
Fumed silica cost	10	\$/kg
PEI cost	15	\$/kg
Monolith substrate	200	\$/ft3
Sorbent replacement	23.5	\$/kg
Process Assumptions		
Sorbent CO_2 capacity	9.45	wt.%
Process cycle time	30	mins
Sorbent replacement	3	years
Plant lifetime	20	years
Sorbent productivity	0.31	t-CO_2/m^3/day
Air contactor ΔP	451	Pa

The catalyzed sorbent showed promising stability and provided a significant reduction in DAC cost. The drop in capture cost was achieved due to the catalyst impact on increasing the adsorption and desorption rate, consequently increasing the capture capacity. The catalyst also lowered the regeneration temperature which can lead to savings in steam and cooling water consumption. Overall, the Levelized cost of capture was reduced by 18.3% using this low regeneration (90°C) IL catalyzed (PEI + Si) sorbent coated structured DAC system compared to a baseline study at 120°C regeneration temperature as shown in **Table 7**.

Figure 19 shows the estimated cost at different scales and the road map of the DAC system to 1 million tonne CO_2 per year with implementation from pilot scale commenced in year 2025 to year 2040 with improvements in DAC sorbent absorption capacity, capture rate and reduction in regeneration energy along with the reduction in cost in transport and storage of CO_2 according to Smith (2015). Increasing plant capacity from 100 tonner per day to 3,052 tonne per day reduces the cost of CO_2 capture from 199 \$/tonneCO_2 to 81 \$/tonneCO_2.

Table 7: Comparison of ionic liquid catalyzed (PEI + Si) DAC system with SOTA technologies

Technology/Source	Capture cost (USD/tonne-CO_2)	Capture cost (USD/tonne-CO_2)
Baseline	\$171 (excl. T&S)	\$201 (inc. T&S)
Susteon new sorbent	\$140 (excl. T&S)	\$170 (inc. T&S)

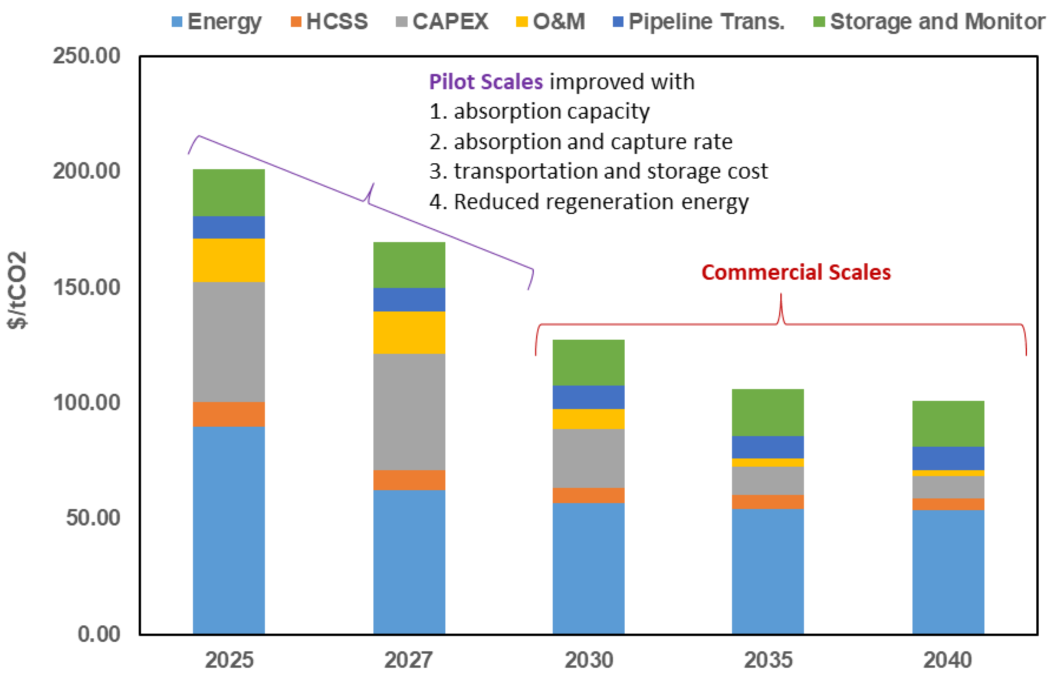

Figure 19: Roadmap of ionic liquid catalyzed (PEI + Si) coated sorbent DAC system to 1 MtonneCO_2/year

EH&S Analysis

The current project involved the synthesis and testing of PEI based DAC sorbents. A preliminary but comprehensive EH&S (environmental, health and safety) risk assessment (Appendix C) has been completed so that potential risk mitigation factors can be applied during the current and next phases of sorbent and process development. A topical report has been submitted that summarizes the key laboratory EH&S concepts and approach. This report summarized the key EH&S risks identified and the mitigating factors for management of the risks are summarized. Since no toxic chemicals were used in the preparation and use of the developed DAC sorbents, the EH&S analysis indicated that there are no un-surmountable risks associated with the manufacturing and use of the DAC sorbents.

Conclusions

Susteon, in partnership with UWy and SoCalGas, has successfully developed ionic liquid silica/PEI sorbents for DAC application. The catalyzed sorbents have been shown to have CO_2 working capacities double that of the non-catalyzed sorbents made from the same starting materials. Furthermore, the catalyzed sorbents have been shown to have adsorption rates that are at least 40% higher than those of the non-catalyzed sorbents.

Amine doped solid sorbents have been proven effective for DAC applications and can be regenerated by heat or by a combination of heat, steam and vacuum. The ionic liquid catalyzed amine sorbents developed by Susteon using polyethyleneimine (PEI) on fumed silica (FS) have shown significantly increased CO_2 capacity at more than 11 wt% of CO_2 captured from air at realistic ambient conditions (highest reported in the literature). Analyses by DAC industrial players indicate that the most effective strategy for drastically reducing DAC cost is increasing adsorbent capacity. The combination of an industrially utilized amine-based sorbent with a highly active catalyst to form a new class of materials for DAC provides a technically viable pathway for reducing the cost of DAC to <\$100/tonne of CO_2 and presents a low technical risk solution with high potential for success. Laboratory measurements showed that the silica/PEI sorbents with 100 ppm of ionic liquid catalyst have almost 100% higher CO_2 cyclic capacity and 40% higher adsorption rate. Generally, CO_2 desorption occurred at higher temperatures and the rate of desorption could be 10 times faster than adsorption which occurs at ambient conditions. Therefore, adsorption rate is a much more important factor in the cost of DAC because it is directly linked to the CAPEX of the total system and the cycle time (i.e., sorbent productivity in ton/day of CO_2 captured per unit volume of the air contactor).

An initial process design coupled with techno-economic analysis, based on optimal experimental results and preliminary resulting from structured sorbent testing, showed a path to lower the DAC cost from the current more than $200/tonne CO_2 to less than $100/tonne when the technology is scaled up and matures with projected material and process improvements. These results demonstrate the effectiveness of the catalyst in silica/PEI sorbents in enhancing sorbents' CO_2 working capacity, in increasing the rate of adsorption and desorption, and in lower the CAPEX and OPEX of the DAC system employing the ionic liquid catalyzed sorbents.

To fully utilize the catalyzed DAC sorbents developed in this project, the next step of technology development is to apply the highly dispersed ionic liquid catalyzed silica/PEI sorbent on a monolith assembly. The adsorption-desorption operation for cyclic DAC process can be conducted by means of vacuum swing adsorption (VSA), or temperature swing adsorption (TSA), or a combination thereof, with the catalyzed high-capacity sorbents. The material (such as fumed silica) can be first coated onto a monolith structure, followed by coating of the active PEI and ionic liquid catalyst materials. The loss of active material during the sorbent regeneration step in a TVSA or TSA process can be minimized by using relatively low volatility PEI and the nonvolatile ionic liquid catalyst.

APPENDIX

Appendix A: Final Technology Maturation Plan

Low Regeneration Temperature Sorbents for Direct Air Capture of CO_2

DOE Cooperative Agreement: DE-FE0031965

SUBMITTED BY

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SUBMITTED TO

US Department of Energy
National Energy Technology Laboratory

September 30, 2022

Technology Readiness Level

Technology Readiness Level Assessment

Amine-based sorbents are the state-of-the-art (SOTA) materials for a large-scale DAC application. These sorbents are known for their high selectivity to adsorb CO_2 from the air. However, the primary bottleneck for large scale deployment of direct air capture (DAC) of CO_2 remains in electrical and heat energy consumption. The key to lowering the DAC cost rests on two factors: 1) the energy required for releasing CO_2 from the capture agents, and 2) the pressure drop needed to move the air through the capture unit. Major technology providers such as Global Thermostat, Climeworks, and Svante have developed and deployed novel pathways to reduce the CO_2 capture cost by mainly lowering the regeneration energy. The current project aimed to develop novel ionic liquid catalyzed solid sorbent materials that have enhanced adsorption and desorption kinetics and CO_2 working capacity than the current state-of-the-art materials. Increase in CO_2 adsorption and desorption rates, reduction of sorbent regeneration temperature and energy consumption were achieved by integrating ionic liquid catalyst in sorbent structure. The ionic liquid catalyst was proven effective when added in ppm concentrations in an aqueous monoethanolamine (MEA) solution.

At the beginning of the project, the ionic liquid catalyzed DAC sorbent technology readiness level (TRL) was assessed to be TRL 2. The basic principles of the ionic liquid catalysis to enhance CO_2 reaction kinetics with amines were observed in preliminary tests carried out to measure CO_2 absorption and desorption performance with 20 wt% MEA solution in the laboratory.

Table A1: Description of technology readiness level 2 (TRL 2)

Relative Level of Technology Development	Technology Readiness Level	TRL Definition	Description
Basic Technology Research	**TRL 2**	Technology concept and/or application formulated	Once basic principles are observed, practical applications can be invented. Applications are speculative, and there may be no proof or detailed analysis to support the assumptions. Examples are still limited to analytic studies. Supporting information includes publications or other references that outline the application being considered and that provide analysis to support the concept. The step up from TRL 1 to TRL 2 moves the ideas from pure to applied research. Most of the work is analytical or paper studies with the emphasis on understanding the science better. Experimental work is designed to corroborate the basic scientific observations made during TRL 1 work.

Description Of Technology

The first step in developing the TMP was to define the technology or "system" to be evaluated. The technology to be assessed was an ionic liquid catalyzed sorbent system for DAC applications. It is the key CO_2 adsorbing material used in the DAC process.

This project was aimed at using the ionic liquid catalyst to catalyze DAC sorbents with enhanced adsorption and desorption kinetics to lower the cost of DAC. The key to lower the cost of DAC rests on three factors: 1) the energy required for releasing CO2 from the capture agents, 2) reduce the size of the air contactor thus lowering the capital costs, and 3) the pressure required to move the air through the capture device.

To achieve this objective, we developed amine-doped solid sorbents catalyzed by an ionic liquid catalyst that have CO_2 adsorption and desorption rates orders of magnitude higher than state-of-the-art sorbents. In laboratory tests, these catalysts showed that the proprietary ionic liquid catalyst used in ppm levels in PEI based sorbents shown 100% increase in sorbent CO2 working capacity and 40% increase in adsorption rate.

In this project, we focused on (1) synthesizing the ionic liquid catalyst for amine-based sorbents for improved desorption and absorption kinetics, (2) evaluating the catalyzed amine-based sorbents for direct CO2 capture process to determine CO_2 desorption rates and energy requirements, and (3) based on the experimental results, developing a conceptual process design to perform a preliminary economic assessment to evaluate the potential for DAC process cost reduction using the catalyzed sorbents.

The ionic liquid catalyst was also tested in amine-based solid sorbent under direct air capture conditions. We prepared polyethyleneimine (PEI) on fumed silica with PEI content from 20 to 50 wt% and catalyst concentrations from 100 to 300 ppm by weight. Using 400 ppm CO_2 in nitrogen as simulated air with 75% RH at 23°C, the CO_2 capture performance and rate of adsorption were measured. The results are shown in **Figure A1** and **Table A2**.

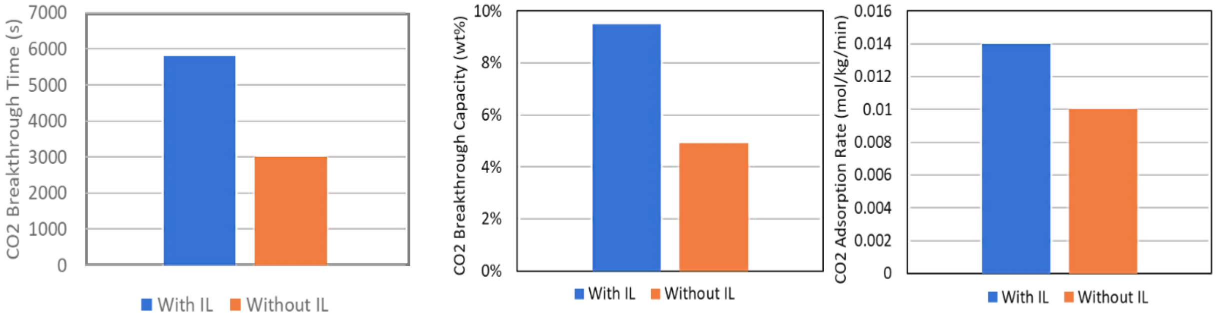

Figure A1: PEI on silica sorbent CO_2 capture performance under direct air capture conditions.

Table A2: PEI on silica CO2 capture performance under direct air capture conditions

Parameters	Unit	With Catalyst	No Catalyst	% Increase
Catalyst concentration	*ppmw*	100	0	-
Breakthrough time	*sec*	5800	3000	93%
CO_2 capacity at breakthrough at 0 ppm	*wt%*	9.49%	4.91%	93%
Rate of CO_2 adsorption	*mol/kg/min*	0.014	0.010	40%

It is clear from these data that the catalyst is also effective in catalyzing CO_2 reaction with amine in solid sorbents. In fact, there is almost a 100% increase in CO_2 breakthrough capacity and a 40% increase in the rate of CO_2 adsorption. These improvements can translate to large reduction in CAPEX due to higher sorbent CO_2 working capacity and productivity.

Critical Technology Elements

This technology has two critical elements:

1. Ionic liquid technology synthesis and characterization, and
2. Polyethyleneimine (PEI) on fumed silica CO_2 sorbent catalyzed by ionic liquid for DAC.

Technology Readiness Level

At the completion of the 2-year long research and laboratory testing work, we assessed that both of the critical technology elements have reached a **TRL level of 3** (**TRL 3**) based on the description shown in **Table A3** below. We scaled up the ionic liquid synthesis process from gram level in the laboratory to kilogram level in pilot scale. In fact, we prepared about 20 kg of the ionic liquid for characterization and testing under this project. As stated above, the ionic liquid catalyzed PEI/fumed silica sorbent has been tested in the laboratory using simulated air with about 400 ppm CO_2 for CO_2 breakthrough capacity and cyclic absorption/desorption behavior.

Table A3: Description of technology readiness level 3 (TRL 3)

Relative Level of Technology Development	Technology Readiness Level	TRL Definition	Description
Research to Prove Feasibility	**TRL 3**	Analytical and experimental critical function and/or characteristic proof of concept	Active research and development (R&D) is initiated. This includes analytical studies and laboratory-scale studies to physically validate the analytical predictions of separate elements of the technology. Examples include components that are not yet integrated, or representative tested with simulants. Supporting information includes results of laboratory tests performed to measure parameters of interest and comparison to analytical predictions for critical subsystems. At TRL 3 the work has moved beyond the paper phase to experimental work that verifies that the concept works as expected on simulants. Components of the technology are validated, but there is no attempt to integrate the components into a complete system. Modeling and simulation may be used to complement physical experiments.

Target Commercial Application

The ionic liquid catalyzed sorbents with enhanced adsorption and desorption kinetics are targeted for DAC applications. The key to lowering the cost of DAC rests on three factors: 1) the energy required for releasing CO_2 from the capture agents, 2) the pressure drop required to move the air through the capture device, and 3) the capital cost of the capture system. The current project addressed all these factors by developing solid sorbent materials which have much faster adsorption and desorption kinetics than the current state-of-the-art materials and can be made into structured sorbent beds for low pressure drop operation to significantly high cyclic CO_2 capacity to lower the cost of DAC.

The Direct Air Capture Utilization and Storage (DAC-US) is a complementary technology to point-source CCUS and vital to meet CO_2 mitigation targets of 1.5°C scenario. DAC-US captures CO_2 from ambient air and subsequently converts or stores it. DAC is potentially a scalable negative CO_2 emissions technology. Extensive deployment of DAC can mitigate distributed emissions such as transport and aviation. Beyond negative emissions, DAC units can be placed ubiquitously to capture and utilize CO_2 for a wide variety of applications. Installing DAC at the sequestration hub's vicinity can reduce transport and storage costs by eliminating expensive pipelines. Expanding DAC deployment on a large scale unlocks significant potentials to convert CO_2 to high-value products while reducing CO_2 emission levels. Some of the CO_2 utilization pathways that have been integrated to point-source CO_2 capture can be integrated downstream of the DAC unit. Successful examples of such integrations are converting CO_2 to methanol and acetic acid or fixing CO_2 in building block materials. Methanation is another alternative that can be integrated downstream of the DAC unit to produce renewable natural gas, which is considered as a promising pathway for renewable energy source storage and supply.

These advantages of DAC technologies become a strong promoter for the commercial deployment of these systems. Most DAC technologies are modular, which extends the prospect of more rapid scaling by numbering them up. The regeneration can be done at low temperatures (significantly below 100°C), enabling use of industrial waste heat. The CO_2 capture and regeneration rates are expected to be higher, thereby providing a shorter cycle time and high capital productivity.

Technology Maturation Activities

Activities Performed in the Project

By performing the following activities, Susteon has taken the ionic liquid catalyzed solid sorbent technology from **TRL 2** at the beginning of the project to **TRL 3** at the completion of the project which makes cost-effective DAC deployment more achievable. The activities were aimed at developing the ionic liquid catalyzed sorbent materials for application in commercial-scale processes to address scale-up and operational uncertainties for large-scale DAC deployment. In the first phase of the project, the focus was on understanding the reaction mechanism, developing a kinetic model, and determining the impact of operating conditions on material stability and capture performance. The activities are summarized in **Table A4**.

Table A4: Technology Maturation Activities

Activity	Description
Proof of concept for integrated sorbent with catalyst	The laboratory data summarized in the previous section provided strong evidence of the efficacy of the ionic liquid catalyst in reducing energy consumption. Those preliminary tests have been done in an amine solvent system. There is a strong analogy between the regeneration mechanism in amine-based solvents and amine-based sorbents for CO_2 capture. The ionic liquid's catalytic impact was verified under DAC operating conditions with the candidate sorbents.
Synthesize the catalyst and investigate alternative pathways to be able to produce catalyst at a large scale	Catalyst required for CO_2 capture tests was synthesized following known procedures from our lab synthesis. While preparing the materials, the team investigated the pathways to optimize the catalyst synthesis to develop a commercially reliable process.
Integrate the developed catalyst in commercially available sorbent for DAC	The team integrated the ionic liquid catalyst in commercially available DAC sorbents and tested them for effectiveness. During this activity, the integration procedure was investigated, which helped define the development required for commercial-scale sorbent production.
Ensure the quality, stability, and performance of the new ionic liquid-enhanced sorbent at real conditions and its strength while being exposed to highly variable feedstock conditions	During catalyst production and integration of ionic liquid to sorbent structure, the team developed the QA/QC procedures to ensure the produced material meets the KPI's expected for the sorbent. The performance and stability of the new sorbent was tested under real conditions.
Commercialization assessment of the project	Project team evaluated the new sorbent's performance and the procedures to produce the catalyst and its integration to commercially available sorbents for large-scale applications. This activity helped in identifying technological drawbacks and economic drivers and in defining the milestones for the next stage maturation of the technology

Post-Project Plans

Susteon's approach to combining known amine-based sorbents with a proven catalyst forms a new class of materials for DAC. It provides a technically viable pathway for reducing the cost of DAC to <$100/tonne-of-$CO_2$ and presents a low technical risk solution with high potential for success.

After completing this project, the next step in developing the "low regeneration-temperature DAC technology" is a pilot plant campaign using a multi-segment rotating packed bed. This project will do its best to ensure that as much of the groundwork to support this pilot plant test is completed. As part of taking this technology to the next scale, the team will actively work with a commercial catalyst manufacturer to define and evaluate scale up for catalyst production. Additionally, feedback from potential end-users of the technology, on integrating the new sorbent in widely used rotating packed bed and monolith technologies for DAC will help establish testing plans and goals for the pilot campaign. Successful pilot plant testing would be followed by efforts to locate potential opportunities for using the technology at sites for small scale commercial applications. Major post-project activities are summarized in **Table A5**.

Table A5: Post Project Activities

Activity	Description
Deployment of the confirmed pathway to produce the catalyst in large quantity for commercial application.	Applying the new catalyst on a large scale requires a technically reliable and cost-effective process to supply and support the market demand. Susteon will investigate the available pathways and propose a commercially viable option to ensure the project's success. The proposed pathway will include reliable quality control and a quality assurance philosophy that guarantees product performance.
Development of structured material systems and component designs incorporating the ionic liquid catalyzed sorbent specifically tailored for DAC processes that are capable of more efficient mass and heat transfer while reducing pressure drop.	Employ monolithic or 3D printed designs and fabrication techniques to produce structured material system and achieve low-pressure drop and DAC cost reduction using the ionic liquid catalyzed DAC sorbent. Maximize CO_2 capture volumetric productivity of DAC components to reduce system size, land requirements and environmental footprint of DAC systems.
Integrated bench scale testing of the ionic liquid catalyzed structured sorbent system.	Test the ionic liquid catalyzed structured sorbent system using integrated bench-scale DAC systems under environmentally relevant conditions for DAC. Establish improved material stability towards oxidative and hydrothermal degradation with long-term testing. Determine auxiliary power requirements Maximize CO_2 capture volumetric productivity of components to reduce air contactor size, land requirements and environmental footprint of DAC systems.
Design the procedure for the integration of the proposed catalyst on commercially available amine-doped sorbents	Several companies such as Svante, Climeworks, and Global Thermostat have commercialized amine-based sorbents for DAC application. Susteon has a strong relationship with these major players and will explore and define the modifications required in their process for the catalyst integration and production of the new sorbent for commercial-scale application.

Ensure the stability and performance of the new ionic liquid-enhanced sorbent at real conditions	The pilot campaign will prove the new sorbent's stability, long-term performance and determine O&M cost for a detailed TEA.
Investigate modifications on the end-to-end capture process	Deployment of this new sorbent to reduce overall energy consumption may require redesign of other unit operations upstream and/or downstream of the capture unit.

Appendix B: State Point Table

Final state point table for ionic liquid catalyzed direct air capture sorbent based on test data is shown in **Table B1** below.

Table B1: State-Point Data for Sorbent System

	Units	Measured Performance (Granule, Lab)	Projected Performance (Structure, Bench)
True Density @ STP	*kg/m*^{3}	1700	N/A
Bulk Density	*kg/m*^{3}	380	50 to 198[a]
Average Particle Diameter	*mm*	0.25	0.1[b]
Particle Void Fraction	*m*^{3}*/m*^{3}	0.375	0.460[c]
Packing Density	*m*^{2}*/m*^{3}	200	2,025-2,400[d]
Solid Heat Capacity @ STP, Sorbent	*kJ/kg·K*	1.15	1.15
Solid Heat Capacity @ STP, Monolith	*kJ/kg·K*	N/A	0.83
Crush Strength	*kg*$_f$	N/A	N/A
Attrition Index	-	9%	N/A
Thermal Conductivity, Sorbent Thermal Conductivity, Monolith	*W/(m·K)*	0.045	0.045
	W/(m·K)	N/A	0.4 – 2.5
Adsorption			
Pressure	*bar-a*	1.000405	1.000405
Temperature	*°C*	25.0	Ambient
Equilibrium Loading	*gmol* CO_2*/kg*	1.6 – 2.5	1.6 – 2.5
Heat of Adsorption	*kJ/gmol* CO_2	70 - 85[e]	70 - 85[e]
CO_2 Adsorption Kinetics	*gmol/kg-min*	0.014[f]	0.03 – 0.10
Desorption			
Pressure	*bar-a*	0.004	0.8 – 1.0
Temperature	*°C*	100 - 120	90 - 120
Equilibrium Loading	*gmol* CO_2*/kg*	0.1	0.1
Heat of Desorption	*kJ/gmol* CO_2	70 - 85[e]	70 - 85[e]
CO_2 Desorption Kinetics	*gmol/ kg-min*	0.12[f]	0.2 – 0.3
Overall Performance			
Space Velocity	*hr*$_{-1}$	20,600	100,000
Volumetric Productivity	*gmol*$_{CO2}$*/ (hr l*$_{adsorber\ bed}$*)*	0.31	0.5 – 2.5
CO_2 Capture Efficiency (single pass)	*%*	90	90
Pressure Drop	*Pa*	200	100
Degradation	*(% capacity fade/cycle)*	Stable in 25 cycles	<5*10^{-6} g

Notes:

[a] Bulk density of monolith

[b] Sorbent washcoat thickness

[c] Porosity of the monolith

[d] Packing density calculated as the surface area of the active sorbent divided by the overall monolith volume including voidage.

[e] Heat of adsorption and desorption for the sorbent were experimentally determined.

[f] Overall average kinetic rate measured in lab over period of 30 minutes. It is to be noted that initial rate where process cycle will be designed could be 4-5 times higher than this rate. Furthermore, in the lab setup we are mass transfer limited while in a commercial embodiment mass transfer limitation will be minimal.

[g] This represents 5% degradation per year, and roughly 4,000 cycles per year

Appendix C: Preliminary Environmental, Health and Safety Risk Assessment

Low Regeneration Temperature Sorbents for Direct Air Capture of CO_2

October 1, 2020 – September 30, 2022

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Disclaimer

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Executive Summary

This topical report presents the preliminary environmental, health and safety (EH&S) risk assessment for the project DE-FE0031965 involving the synthesis and use of an ionic liquid catalyst in polyethyleneimine (PEI) based sorbents for direct air capture (DAC) of CO_2. Ionic liquid catalyzed DAC sorbents have demonstrated ~100% increase in breakthrough CO_2 capacity and 40% increase in adsorption rate.

This topical report summarizes the key EH&S risks. Key EH&S risk are provided in tabular form including the risk mitigation factors.

Ionic Liquid Catalyzed Sorbent Development

This project was aimed at using the ionic liquid catalyst to catalyze DAC sorbents with enhanced adsorption and desorption kinetics to lower the cost of DAC. The key to lower the cost of DAC rests on three factors: 1) the energy required for releasing CO_2 from the capture agents, 2) reduce the size of the air contactor thus lowering the capital costs, and 3) the pressure required to move the air through the capture device.

To achieve this objective, we developed amine-doped solid sorbents catalyzed by an ionic liquid catalyst that have the potential to increase the CO_2 adsorption and desorption rates by orders of magnitude. In laboratory tests, these catalysts showed that the proprietary ionic liquid catalyst used in ppm levels in PEI based sorbents shown 100% increase in sorbent CO_2 working capacity and 40% increase in adsorption rate.

In this project, we have been focused on (1) synthesizing the ionic liquid catalyst for amine-based sorbents for improved desorption and absorption kinetics, (2) evaluating the catalyzed amine-based sorbents for direct CO_2 capture process to determine CO_2 desorption rates and energy requirements, and (3) based on the experimental results, developing a conceptual process design to perform a preliminary economic assessment to evaluate the potential for DAC process cost reduction using the catalyzed sorbents.

The benefits of this novel sorbent process are 1) greater than 20% reduction in the energy required for sorbent regeneration, 2) the potential to use waste heat for sorbent regeneration, 3) drastic increase in sorbent lifetime/stability due to lower regeneration temperature, 4) lower sorbent replacement cost due to the increased stability, and 5) >40% reduction in CAPEX for DAC processing using the ionic liquid catalyzed sorbents.

Sorbent Preparation Process

PEI dissolved in ethanol at desired concentration was mixed with a known amount of fumed silica support. Ethanol was removed by evaporation at ~70°C with stirring resulting in a very even distribution of PEI onto the fumed silica. The resulting sorbents were further dried under vacuum (~20 mtorr) at ~60°C for 6-8h.

The ionic liquid catalyst was dissolved in ethanol in desired concentration and deposited onto the PEI/Silica sorbent by incipient wetness method.

Sorbent Testing Process

A direct air capture setup shown in detail in **Figure C1 and C2** has been designed and built to evaluate performance of the sorbents for DAC. To simulate air CO_2 concentration (400 ppm), the flow rates of 1% CO_2/N_2 mixed gas (1) in combination with pure N_2 (2) were passed through mass flow controllers (4) which were calibrated to allow a total flow rate of 500 to 2500 mL/min containing 400 ppm CO_2 ($C_{a,0}$, the gas analyzer (14) was calibrated to measure the CO_2 concentration in the gas stream). 0.5 grams of lab synthesized sorbent (12) were held in a steel or quartz tube reactor (10) between two layers of quartz wool (11). An experimental cycle on this setup consisted of three steps: pretreatment (T: 120 °C to release adsorbed moisture and CO_2), adsorption ($C_{a,0}$= 400 ppm CO_2; T: 25 °C), and desorption (T: 85 - 120 °C). After pretreatment at 120 °C, the temperature was cooled to absorption temperature. Measurement and recording of CO_2 concentration in the reactor exit stream started when simulated air was introduced into the reactor. CO_2 breakthrough was observed when this CO_2 concentration started to rise from zero. CO_2

breakthrough capacity was calculated from the breakthrough time, inlet gas flow rate, and the total weight of sorbent in the reactor. When exit stream CO_2 concentration reached that of the inlet stream, sorption equilibrium was said to be reached. Sorbent equilibrium CO_2 capacity was calculated from the time it took to reach equilibrium. After the adsorption experiment, temperature of the reactor was set to the desorption temperature. A 500 ml/min of N_2 gas was used as carrier gas during desorption.

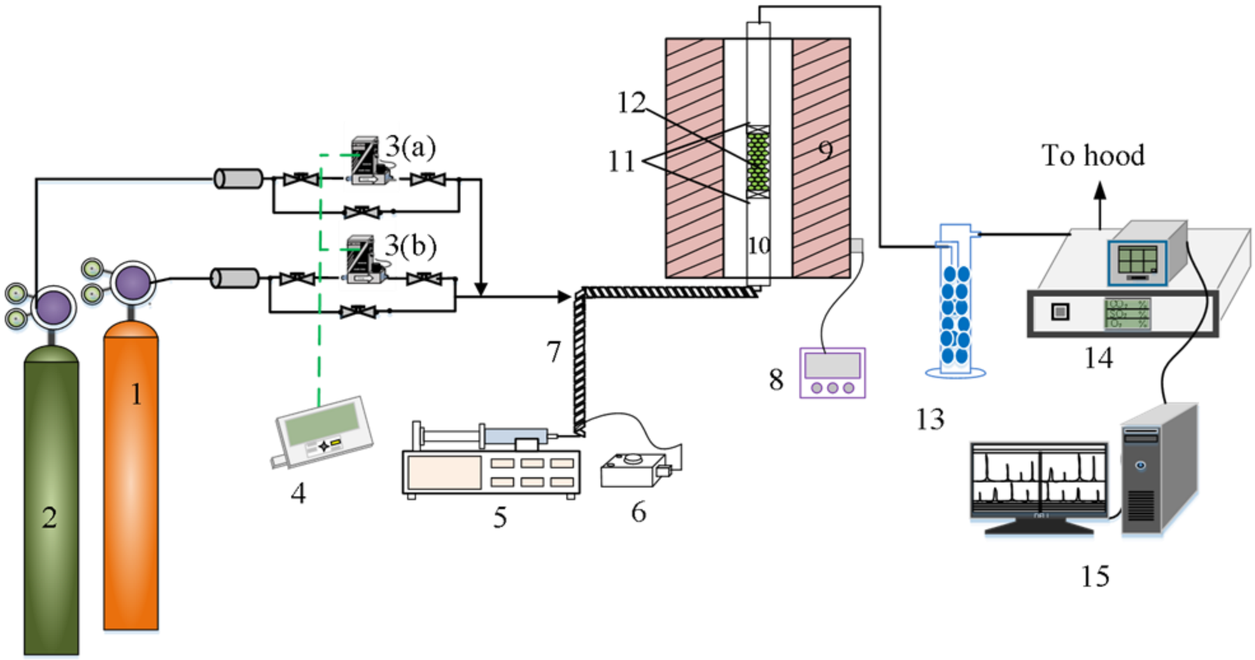

Figure C1: Diagram of air capture setup

1. 1 vol% CO_2 gas cylinder
2. nitrogen cylinder
3. mass flow controllers (3a and 3b); 4. control module of mass flow controller
4. pump
5. syringe
6. temperature controller of heating tape
7. heating tape
8. temperature controller of the tube furnace
9. tube furnace
10. tube reactor
11. quartz wool
12. sorbent bed
13. moisture removal unit
14. gas analyzer
15. data recording unit/pc

Figure C2: Photo of the air capture setup

Environmental and Health Risk Assessment Methodology

The preliminary EH&S risk assessment for the ionic liquid catalyzed PEI/Silica DAC sorbents was performed using Job Safety Analysis as a tool. The EH&S risk assessment was used as a formal process to identify potential environmental; health and safety hazards related the development and use of the ionic liquid catalyzed PEI/silica DAC sorbents. This EH&S risk assessment identifies which risks pose the greatest threat and identify controls to reduce the level of risk. In addition, as per DOE-NETL requirements, a National Environmental Policy Act (NEPA) environmental questionnaire has been completed for this project. A summary of risk factors and mitigating actions was prepared and shown below in **Tables C1 and C2**.

Summary of EH&S Risks and Mitigating Approaches

The following tables summarize the major EH&S issues arising from preparation and testing of the ionic liquid PEI/Silica DAC sorbents, along with a list of related mitigation approaches and actions to satisfy all existing EH&S regulations and guidelines. **Table C1** focuses on potential risks related to chemicals and potential exposures, while **Table C2** focuses on the preparation and testing process.

Table C1: EH&S Risks and Mitigation Approaches

Risk	Mitigation Approach	Comments (max. concentrations, exposures etc)
Waste solvent	Handled by appropriate disposal company off-site	Up to 5 gallons of ethanol (estimated amount a year) of used solvent
Accumulated used sorbent (solids)	Disposal of used sorbent by appropriate disposal company off-site	Maximum about 1 kg per year
Wastewater	Disposal of wastewater by appropriate disposal company off-site	Waste generated from ionic liquid synthesis is handled by appropriate for the off-site disposal, which may be characterized as “special waste” or “hazardous waste”

Table C2: EH&S Risks and Mitigation Approaches (Design, operations, and maintenance)

Risk	Mitigation Approach
Safe operation of lab equipment	Applied Susteon and University of Wyoming's comprehensive "Laboratory Safety Practices" guidelines for safety and operator training. Perform "Job Safety Analysis" prior to start of operation.
Safety issues arising from improper design and operations/maintenance requirements not identified at design	Comprehensive Process Safety Reviews (PSR)
Chemical exposure	Multiple eye wash and emergency showers Chemical hood for synthesis of catalyst and sorbents
Solvent handling	Rigorous operating procedures including mandatory usage of Personal Protection Equipment (PPE)
Solvent storage (regulatory requirements)	All laboratory chemicals are stored according to their storage requirements

Chemical properties and Toxicological effects

The chemicals used in the preparation of ionic liquid catalyzed PEI/Silica sorbents are: (1) Ionic liquid catalyst, (2) PEI and (3) Silica.

Table C3: Chemical EH&S Risks

Property/Effect	Solvent Data/Guidelines
State of matter/Color/Odor	Solid, amine-like
Emergency overview/Precautions	May cause eye irritation Severely irritating to eyes and skin May be harmful if swallowed May be harmful if absorbed through skin. - May cause sensitization by inhalation. May cause sensitization by skin contact Avoid contact with eyes, skin, and clothing Avoid inhalation of mists/vapors/dust
Potential health effects	**Primary routes of exposure:** Routes of entry for solids and liquids include eye and skin contact, ingestion and inhalation. Routes of entry for gases include inhalation and eye contact. Skin contact may be a route of entry for liquefied gases.
Degradation/Environmental fate	No data available concerning biodegradation
Flammability	**Nonflammable**
Accidental release measures	**Personal precautions:** Avoid inhalation. Avoid contact with the skin, eyes, and clothing. **Environmental precautions:** Do not disposal as regular solid waste, do not disposal into drains/surface waters/groundwater. **Cleanup:** Clean contaminated area by sweeping up all solid particles and seal in plastic bags for disposal by appropriate hazard solid waste disposal company off-site.
Handling and storage	**Handling:** Ensure thorough ventilation of storage and work areas. **Storage:** Keep container tightly closed and in a cool place. Keep container dry. **Storage incompatibility:** Segregate from strong acids and oxidizing agents.

Temperature tolerance	25 to 80°C. It is not necessary to protect the packed product against exceeding the temperature indicated.
Exposure control and personal protection	**Personal protective equipment** **Respiratory protection:** Wear a NIOSH-certified (or equivalent) respirator as necessary. Observe OSHA regulations for respirator use (29 CFR 1910.134). **Hand protection:** Wear chemical-resistant protective gloves. Manufacturer's directions for use should be observed because of great diversity of types. **Eye protection:** Tight fitting safety goggles (chemical goggles). Wear face shield if splashing hazard exists.
General safety and hygiene measures	Handle in accordance with good laboratory hygiene and safety practice. Avoid contact with eyes and skin. Do not breathe dust. Eye wash fountains and safety showers must be easily accessible.
Stability and Reactivity	**Substances to avoid:** Oxidizing agents and Acids **Hazardous reactions:** Evolution of smoke in contact with oxidizing agents. Evolution of heat in contact with acid. **Decomposition products:** Possible thermal decomposition products: carbon monoxide, carbon dioxide, ammonia, volatile organic compounds, nitrogen oxides, carbon oxides, hydrogen fluoride **Thermal decomposition:** No decomposition if stored and handled as prescribed/indicated. **Corrosion to metals:** Corrosive effects to metal are not anticipated. **Oxidizing properties:** Not fire-propagating
Toxicological Information	**Acute toxicity:** Low toxicity after single ingestion. Virtually nontoxic after a single skin contact. **Irritation / corrosion:** Irritating to skin. May cause severe damage to the eyes and skin. Corrosive to skin, eyes and respiratory system. **Sensitization:** May cause sensitization of the respiratory tract. Sensitization after skin contact possible. **Repeated dose toxicity:** After repeated exposure, the prominent effect is limited to local irritation. **Genetic toxicity**: Non data available **Carcinogenicity:** No data available concerning carcinogenic effects. **Reproductive toxicity:** No reliable data are available concerning reproduction toxicity.
Aspiration Hazard	No aspiration hazard expected.
Ecological Information	**Aquatic toxicity:** Not likely to be toxic to environmental biota. **Environmental mobility:** The substance will not evaporate into the atmosphere.
Other adverse effects	Do not disposal of product into the environment
Disposal considerations	**Waste disposal of substance:** Incinerate in suitable incineration plant, observing local authority regulations.

Conclusion

The current project involved the synthesis and testing of PEI based DAC sorbents. A preliminary but comprehensive EH&S (environmental, health and safety) risk assessment has been completed so that potential risk mitigation factors can be applied during the current and next phases of sorbent and process development. This topical report summarized the key laboratory EH&S concepts and approach. Key EH&S risks were identified and the mitigating factors for management of the risks were summarized.