Thermokinetic/Mass-transfer Analysis of Carbon Capture for Reuse/Sequestration

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ABSTRACT

Effective capture of atmospheric carbon is a key bottleneck preventing non bio-based, carbon-neutral production of synthetic liquid hydrocarbon fuels using CO_2 as the carbon feedstock. Here we outline the boundary conditions of atmospheric carbon capture for recycle to liquid hydrocarbon fuels production and re-use options and we also identify the technical advances that must be made for such a process to become technically and commercially viable at scale. While conversion of atmospheric CO_2 into a pure feedstock for hydrocarbon fuels synthesis is presently feasible at the bench-scale – albeit at high cost energetically and economically- the methods and materials needed to concentrate large amounts of CO_2 at low cost and high efficiency remain technically immature. Industrial-scale capture must entail: (1) Processing of large volumes of air through an effective CO_2 capture media and (2) Efficient separation of CO_2 from the processed air flow into a pure stream of CO_2 .

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NOMENCLATURE

GJ	gigajoule
n	moles
R	ideal gas constant
Κ	Kelvin
Т	temperature
tonne	metric ton
ΔG_{unmix}	Gibbs Free Energy of unmixing
χ	mole fraction

1. INTRODUCTION

1.1. Rationale

Distributed recycling of atmospheric CO_2 into hydrocarbon fuels from stand-alone facilities operating on solar and wind energy would: (1) Contribute to US energy security by adding a new feedstock for domestic liquid fuel production; and (2) Increase the tooth to tail ratio^a of our armed forces by potentially enabling fuel production closer to where it is needed; while (3) avoiding further increases in the concentration of atmospheric CO_2 . For a carbon neutral liquid hydrocarbon fuels process, the CO_2 feedstock must come from a bio-refinery or from the atmosphere using energy not derived from fossil fuels. Capturing CO_2 from point sources such as power plants would allow the CO_2 to be recycled into a liquid fuel only once and mitigate some, but not all, CO_2 emissions. Note that distributed CO_2 capture would also allow on-site utilization of CO_2 to enhance and simplify algal biofuel production, enhanced oil recovery, and potentially the production of fertilizers, and solvents, and polycarbonate plastics.

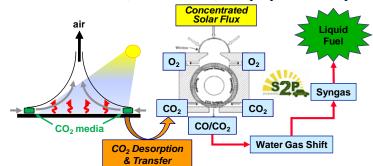


Figure 1. A concept for carbon neutral fuel production.

The burning of fossil fuels consumes roughly 10 to 15 GJ of energy for every tonne of CO_2 that is produced and released to the atmosphere.[1] The Gibbs free energy of un-mixing 100% of the CO_2 from the atmosphere is much less, at 0.5 GJ/tonne CO_2 .^b In comparison the energy required to separate 100% of the CO_2 from flue gas (12% CO_2) is marginally less energy intensive on an absolute basis at 0.2 GJ/tonne CO_2 . Despite small theoretical absolute energy difference, the key technical features of low cost, high efficiency, large-scale atmospheric carbon capture remain unresolved. The primary technical obstacles involved fall into three categories: (1) Air processing, (2) Designing better CO_2 sorbents, (3) Efficient integration to maximize efficiencies. The critical success metrics are: (1) efficiency (theoretical energy/actual energy,) (2) cost, (3) scalability or sustainability, and (4) durability.

^a Tooth to tail ratio (T3R) is a comparison of combat to non-combat (overhead and logistic support) elements in the armed forces.

^b Using $\Delta G_{unmix} = -nRT(\chi_{CO2}ln\chi_{CO2} + \chi_{atm}ln\chi_{atm})$ whereby $n = 5.98 \times 10^7$ mol air/tonne CO₂, R = 8.314 J/K mol, T = 288 K, $\chi_{CO2} = 3.85 \times 10^4$ and $\chi_{atm} = 1 - \chi_{CO2}$.

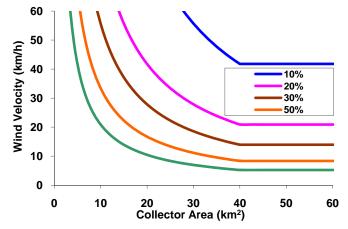
2. RESULTS & DISCUSSION

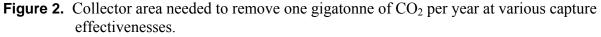
2.1. Air Processing

Since CO_2 is a trace component of the atmosphere (385 ppm by volume), the capture of industrially useful volumes of CO_2 will require processing exceedingly large volumes of air. CO_2 capture rates increase and scale linearly, with increases in the capture effectiveness of CO_2 , air flux, and collector area. This relationship is:

Captured CO₂ (kg/yr) \propto (Capture Effectiveness, %)(Air Flux, m/sec)(Collector Area, m²)([CO₂]_{air}, kg/m³)

Using the ideal gas law and conditions of 15 °C and 1 atm, the density of CO_2 in dry air is calculated to be 0.0007 kg CO_2/m^3 air. This means that a 1 m² collector operating at 10% capture percentage with an air speed of 3.6 km/h could remove 2.2 tonnes CO_2/yr . Figure 2 shows the collector area needed to remove one gigatonne of CO_2 per year, roughly 1/7 of annual CO_2 emissions for the US, as a function of capture percentage and air speed. Lower capture effective sorbents will require greater collector areas and vice versa. Higher air speeds, natural or engineered, would allow a smaller collector footprint (Figure 2).





The collector areas required to capture large amounts of CO_2 are relatively small, and may be a reasonable source for limited fuel production (i.e. forward military operations). However maintaining a constant air flow to prevent plant start/stop events and/or overcoming back pressures associated with moving air through CO_2 capture media is potentially more problematic. Natural wind requires no external energy input, but wind velocities vary diurnally and seasonally with solar radiation inputs and the alternative, controlling air flows with motorized fans, could be prohibitively energy intensive. The challenge to air processing appears to be to take advantage of passive or concentrating solar power to generate high, sustained and directed air velocities.

Solar chimneys (Figure 3) are one type of device identified for passive wind generation and potential integration with CO_2 adsorbent materials. Solar chimneys rely on a temperature gradient, created by passive or concentrated solar energy, to generate fluxes of air (wind) which turn a turbine to produce electricity. Coupling the solar chimney with the capture of CO_2 from air would result in a "negative-carbon" energy source, if the CO_2 is later sequestered or carbon neutral if the CO_2 is used as a feedstock for hydrocarbon fuels. The solar chimney would generate power with turbines AND provide the necessary fluxes of air to be passed over/through sorbents to remove CO_2 from the atmosphere. Both thermal and electrical energy generated from the chimney could be utilized for the CO_2 capture and release mechanism (thermal swing, pressure swing, etc...) and/or be delivered to the grid, assuming the availability of transmission lines.

How to best interface air fluxes to sorbent media; determining the effects and interactions amongst the critical operating parameters (temperature, air speed, chimney height, chimney design, and collector footprint) are elements that still need to be understood for a CO₂ capture form air device.



Figure 3. Solar chimney at Manzanares, Spain.

One method investigated to move large amounts of air would entail generating a vortex through the combination of buoyancy and shear forces. This concept is similar to a solar chimney, however air would enter tangential to the chimney. The size of the vortex is yet to be determined, but it is anticipated that the diameter would be on the order of 10s of meters and height on the order of 100s of meters, up to kilometers. The vortex can be created by either introducing air at a higher temperature than the surroundings in tangential entries in a circular base structure that anchors the vortex, or by heating a surface in the center of the base structure resulting in entrainment through the tangential ports. The circular base structure only needs to extend roughly a diameter in height since it is necessary only to allow the vortex to become established. The ability to generate such a vortex has been demonstrated at a relatively small scale (~1 m dia., ~7 m height).[2] Questions remain, however, regarding atmospheric interaction, energy supply requirements, and air intake potential. Thus, the outstanding research questions for development pertain to efficiency and stability for prototypic scales. Efficiency for the objectives of this research pertains to the ratio of intake air to the heating requirements both in terms of power.

An initial evaluation has been performed using the Computational Fluid Dynamics (CFD) code Vulcan in order to determine the potential to utilize small temperature differentials to create a vortex. A simulation of a 10 m diameter base structure with a height of about 10 m and a heated plate of about 5 m in diameter at the bottom of the base structure was performed. The plate was specified at a temperature 30 K above ambient. Figure 4 showing temperature

contours and velocity vectors indicate that even at this low temperature differential a vortex five diameters in height can be created. The intake velocities are approximately 1.5 m/s, providing a volumetric air intake of about 60 m³/s through the ports in the base structure. Note that this volumetric air intake is several orders of magnitude below what is required for the objective of this project (~ $2 \times 10^5 \text{ m}^3$ /s). However, it is anticipated that future simulations would reveal much higher intake capacities at prototypic scales, particularly for optimized base structures that could be designed using CFD.

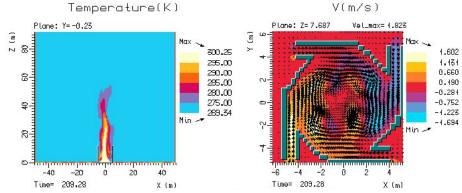


Figure 4. Temperature contours and velocity vectors for vortex based air movement. CO₂ capture media would be placed in the tangential entry points at the base of the vortex.

2.2. CO₂ Selective Sorbents

Given sufficient air fluxes, sorbents are needed that can efficiently, cost-effectively, and reliably catch and later release CO_2 with minimal energy inputs. There are commercial processes that remove CO_2 from concentrated streams associated with flue gas and hydrogen production. Metal oxide and hydroxide chemistries to capture CO_2 from flue gas have also been proposed. However, chemistries/materials to remove CO_2 from air have been less well studied. A notable exception are small scale systems developed for use in space life support.[3] The following discussion highlights technologies that might be adapted to separate CO_2 from air. The separation can take place in a liquid or solid media.

2.2.1. Liquid Sorbents

Aqueous amine solvents, such as monoethanolamine (MEA), Econamine (Fluor) and KS-1 (Mitsubishi), are commercially available to remove CO_2 from flue gas. Aqueous solutions of amine compounds react with CO_2 from flue gas at near atmospheric pressure to produce ammonium salts.[4] The ammonium salts are subsequently heated to recover CO_2 and regenerate the amine:

High energy demands are largely due to CO_2 release from the large volumes of water; dilution by water, for example, is required to mitigate the corrosive power of the amine on pipes, pumps, etc. The dilution increases the energy required in the regeneration step since a substantially larger volume of solvent, that doesn't participate in CO_2 capture, must be heated. Solvent losses due to evaporation during operation add to costs. The volume of aqueous amine required for CO_2 flue gas capture can be reduced by using a membrane contactor[5], however dilute amine solutions are still needed. Ionic liquids[6] show promise as they eliminate evaporative loss encountered with amine systems, but are usually considered for physical absorption (i.e., at high pressure in fuel gas). Material costs of ionic liquids are currently high and further development is needed to make these solvents suitable for low-pressure applications.

Other aqueous CO₂ sorbents contain alkalis, such as NH₄OH, NaOH, or KOH, and rely on the very high solubility of CO₂ in high pH solutions (> 100 g/L at pH 11 and 25 °C). CO₂ is then released by either: (1) (for ammonia-based solutions) raising the temperature and relying on temperature-dependent ammonium acid-base equilibria to lower the solution pH (ammonium acidity is unique in being peculiarly sensitive to temperature), or (2) using lime to convert the dissolved CO₂ to calcium carbonate that is subsequently calcined to recover the CO₂. Ammoniabased approaches require energy inputs associated for shifting the temperature. Also, ammonia is hazardous and caution must be taken so that ammonia is not released in the process. The calcination step in the second process occurs at ~700°C and requires high energy inputs. In addition, all liquid sorbents must overcome kinetic resistance to the hydration/dehydration of CO₂ as it crosses the gas-liquid interface. In biologic systems the enzyme carbonic anhydrase dramatically accelerates CO₂ equilibration. Carbonic anhydrase has been suggested as a potentially important enabler for achieving the same effect in atmospheric carbon capture schemes, though no pilot scale testing has been done.[7]

2.2.2. Solid Sorbents

Many of the candidate solid sorbents have been borrowed from what has been proposed or commercialized for CO_2 capture from flue gases.[8-9] These rely on metal oxide sorbents and amine functionalized mesoporous sorbents. The metal oxide and mesoporous sorbents capture CO_2 and are later regenerated producing a pure stream of CO_2 through a thermal, pressure, and/or electrothermal swing process.

Metal oxide materials have been extensively investigated for removing CO_2 from air. Other potential adsorbents include alkali carbonates and minerals containing metal oxides or hydroxides of calcium or magnesium.[10-11] A metal oxide reacts with carbon dioxide from the air to form a carbonate. The carbonate is then heated to regenerate the metal oxide and a pure stream of CO_2 .

Table 1 shows the CO_2 capacity and regeneration energy for various metal oxides.[12] Calcium oxide has been studied the most since converting calcium carbonate to calcium oxide is a commercial process used by the paper and pulp industry. Silver and zinc oxides have been evaluated on a smaller scale to scrub CO_2 from air in submarine and space applications. While magnesium oxide appears to be the best candidate for CO_2 adsorption from air based on CO_2 capacity and regeneration energy, other processing parameters including humidity of the air to be

scrubbed, available heat/energy for oxide regeneration and carbonate forming/decomposition characteristics must also be considered as additional critical metrics.

Metal Oxide	Density	CO ₂ Capacity	CO ₂ Capacity	Regeneration Energy
	(g/cm^3)	$(g CO_2/g oxide)$	$(g CO_2/cm^3 oxide)$	$(kJ/g CO_2)$
Ag ₂ O	7.14	0.189	1.349	1.865
BaO	5.72	0.287	1.642	6.081
CaO	2.62	0.785	2.057	4.042
Cs_2O	4.36	0.156	0.680	9.279
K ₂ O	2.32	0.468	1.086	8.895
Li ₂ O	2.01	1.471	2.957	5.146
MgO	3.65	1.092	3.986	2.261
Na ₂ O	2.27	0.709	1.609	7.309
Rb ₂ O	3.72	0.235	0.874	9.172
SrO	4.70	0.425	1.998	5.249
ZnO	5.47	0.540	2.954	1.616

Table 1. Theoretical CO_2 capacity and regeneration energy of metal oxides.[12]

The main advantage of metal oxides is their low cost, abundance and relatively benign environmental impact. Carbon dioxide can be stored indefinitely within these materials or regenerated on a continuous basis. Regeneration of the oxides can be problematic though. High temperatures, 200 to 1000 °C depending on the oxide, must be used and ideally regeneration of the oxide should occur in a vacuum or atmosphere of carbon dioxide to produce a pure CO_2 stream. This is contrary to how an industrial kiln operates in which the hot gases from combustion are in direct contact with the oxide/carbonate materials. Most envisioned CO_2 capture plants have an oxygen fueled kiln to do the oxide regeneration, thus requiring an oxygen separation plant.[13-14] Another issue associated with metal oxides is a loss in capacity after repeated cycles of carbonation/decarbonation.[12] The loss in capacity is due in part from an intractable build up of oxide crystallites which effectively decreases the reactive surface area for $CO_2.[15]$

Solid amine-sorbents work in much the same way as aqueous amine systems, but with much lower water and hence potentially lower energy requirements. A number of amine-modified materials have been examined for reversible CO₂ adsorption/desorption behavior, however, the bulk of them suffer from low CO₂ capacities, instability during cycling, high temperature requirements, and are prohibitively expensive for more general use. This has focused research onto improved sorbents.

A number of amine functionalized mesoporous sorbents have also been proposed for use in capturing CO₂. These sorbents include carbon[16] and silica[17] based sorbents and zeolites[16]. The sorbents are typically activated (by heat) or functionalized with amines to increase carbon dioxide selectivity. Sorption can occur via chemi- or physisorption at ambient temperatures and pressures. The high CO₂ capacity sorbents are functionalized with amines and work in a similar manner to the solvent based amine system. In such a system, CO₂ can react in two different ways. In the first pathway, CO₂ reacts in the presence of water and an amine to form a bicarbonate and ammonium species. Two amines may also react with CO₂ form a carbamate and ammonium species. Both reactions are reversible upon heating to temperatures up to 100 °C.

$$R_2 NH + CO_2 + H_2 O \longrightarrow R_2 NH_2 + HCO_3^{\ominus}$$

$$2 R_2 NH + CO_2 \longrightarrow R_2 NH_3 + R_2 NHCO_2^{\ominus}$$

The amine functionalized sorbents are attractive because of their specificity for CO_2 . However improvements must be made in sorbent CO_2 capacities, stability during cycling, and synthesis costs for an effective and economical process to capture CO_2 from air.

2.2.3. Integrating CO₂ Sorbents and Processed Air Flows

Efficiently integrating a carbon dioxide selective sorbent and air processing unit will be critical for success. There have been several laboratory/bench scale analyses that suggest what a CO_2 processing plant would look like. Keith *et al* has proposed using "spray tower" technology to remove CO_2 from air.[18] In this scheme, air is passed using a motorized fan through a mist of aqueous sodium hydroxide to remove the CO_2 . The sodium bicarbonate generated from this process is then geologically sequestered; no effort was made to consider recovering the CO_2 as a gas for use in fuels production or reuse for algae production, industrial, or enhanced oil recovery purposes. Researchers at Columbia University use sheets of a basic ion exchange resin to capture CO_2 from air.[19] Hydroxide anions bound to the resin react with CO_2 to form a bound bicarbonate and one equivalent of water. Carbon dioxide catch and release can be performed by simply changing the water content of the ion exchange resin. Few details of this proprietary system have been revealed (i.e., how air is moved through the resin, overall efficiencies). A process that uses calcium oxide to capture CO_2 and thermal energy from concentrating solar power to produce a pure stream of CO_2 has also been described, but the scalability, economics, and mechanism for air flow have not been reported for this process.[20]

4. CONCLUSIONS

Our analysis suggests the following technical challenges must be met before capture of atmospheric CO_2 for conversion and recycle to hydrocarbon fuels or for re-use options can be considered plausible at the industrial scale.

- 1. Low-energy input air processing approaches must be pioneered to assure regular and effective air flows through CO₂ sorbent media to ensure high production rates. Ideally these will take advantage of, or be engineered enhancements of, natural wind-generating thermal gradients. Air flows must be over-pressured to compensate for pressure drops or deflection that will naturally occur during passage through or around the media.
- 2. For aqueous CO₂ sorbents, air-water CO₂ transfer may be the step that limits overall process success. Using carbonic anhydrase or a similarly effective CO₂ hydration/dehydration catalytic agent is a potential enabler for an industrial scale CO₂ capture process.
- 3. Less expensive solid CO₂ sorbents must be developed that are stable over very many catch and release cycles, have higher CO₂ capacities, and lower non-recuperable swing energies. Media and/or its support must be designed to minimize the pressure drop associated with the air processing.
- 4. Atmospheric CO_2 capture systems must be objectively evaluated first at the bench scale and later at the pilot scale. A host of bench and pilot-testing has built confidence in flue gas capture approaches and driven significant progress. No such track record exists to benchmark and accelerate the development of atmospheric CO_2 capture approaches.

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