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Direct Air Capture with Integrated Electrochemical Conversion through Combined Solid and Liquid Sorbents

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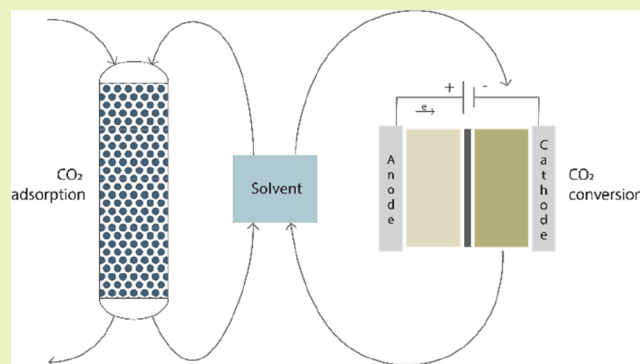
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Supporting Information

ABSTRACT: Direct air capture and CO₂ conversion will play vital roles in a future circular carbon economy. Here, we propose a novel system for integrated CO₂ capture and conversion through the combined use of solid and liquid sorbents and demonstrate its technical feasibility. CO₂ is initially captured from the air using an amine-functionalized solid sorbent that is regenerated by rinsing with an aqueous carbonate-rich solution. The resulting bicarbonate-rich solution is fed to an electrolyzer, converting the bicarbonate electrolyte to syngas. Through our experiments, we demonstrate that the dual-sorbent system is capable of capture from air as well as solid sorbent regeneration and CO₂ conversion. We use a simplified process model based on experimental results to explore the effects of scaling the components of the proposed system. We find that the pH-swing between the rich and lean solutions is a dominant design parameter, which is almost exclusively governed by the electrolyzer sizing. A small pH swing results in an improved electrolyzer performance and beneficial syngas composition, whereas a large pH swing results in efficient solid sorbent use and decreased water loss. Our results further highlight the fundamental trade-offs that are present when designing integrated capture and conversion systems.

KEYWORDS: direct air capture, carbon capture and utilization, electrochemical CO₂ reduction, bicarbonate electrolysis, process integration, sustainable syngas production



INTRODUCTION

Direct air capture (DAC) of carbon dioxide is expected to play a vital role in humanities roadmap to a net-zero CO₂ economy,¹ both as a CO₂ removal technology and as a carbon source for sustainable products. Efficiently capturing CO₂ from a stream as dilute as ambient air (≈ 425 ppm) presents a formidable engineering challenge. Since its introduction by Lackner in 1999,² the prevalent DAC routes have emerged: the solid sorbent process as developed by, among others, Gebald³ and Wurzbacher⁴ and commercialized by Climeworks and Global Thermostat and liquid-based processes such as those developed by, e.g., Zeman and Lackner⁵ and Keith⁶ and commercialized by Carbon Engineering. However, large barriers remain for the gigatonne-scale implementation of DAC, most notably its high costs (≥ 1000 \$/tonne⁷) and energy demands (> 500 kJ/mol⁸).

In the solid sorbent process, air is contacted with an amine-functionalized material, on which CO₂ is adsorbed. A combination of elevated temperature (≤ 120 °C), reduced pressure (≥ 30 mbar), and optional steam purging are used to release CO₂ and regenerate the sorbent. The major energy demand (up to 76%⁹) in this DAC process is associated with thermal cycling of the sorbent. Furthermore, the elevated temperatures are associated with accelerated degradation of the

sorbent, limiting its lifetime (1–3 years) and forming a major cost-driver.¹⁰

Contrarily, in the alkali scrubbing process, the air is contacted with a sodium or potassium hydroxide solution in a cooling tower-like structure. The resulting carbonate solution is causticized with calcium hydroxide to form calcium carbonate crystals, which are calcined to release the CO₂. A major drawback of this process is the high temperature required for the calcining process (≈ 900 °C).¹¹ Additionally, the direct contact of the capture solution and large volumes of air leads to evaporative loss of water from the system, up to 20 tonne of water per tonne of CO₂.¹² With half of earth's population predicted to face severe water stress by 2030,¹³ losses should be minimized to conserve this critical resource.

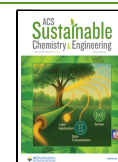
Among the possible routes to alleviate these challenges, the calcium loop and calcining step can be replaced by an electrochemical pH-swing to release the CO₂. This concept

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was, for example, demonstrated by Shu et al.,¹⁴ who achieved an energy consumption of 374 kJ/mol CO₂ through a H₂-recycling electrochemical system. Sabatino et al.¹⁵ modeled a similar system using bipolar membrane electrodesis instead and predicted a minimum energy consumption of 236 kJ/mol CO₂. Shu et al. proposed a dual sorbent system, where CO₂ is initially captured on an amine-functionalized solid, which is then rinsed with a sodium hydroxide solution to remove the CO₂, while the resulting bicarbonate solution is regenerated electrochemically.¹⁶ This eliminates the thermal cycling of the solid sorbent while also avoiding the direct contact between the air and the alkali solution.

Rather than only releasing the CO₂, electrochemical approaches can be used to immediately convert captured CO₂, producing valuable products at a potentially lower energy penalty than separate capture and conversion processes. One such conversion pathway, bicarbonate electrolysis, has been demonstrated extensively in our own lab¹⁷ and other research groups.^{18,19} For bicarbonate electrolysis to CO, Li et al. demonstrated a Faradaic efficiency (FE) of 82% at 100 mA/cm² and a cell voltage of 3.5–3.8 V,¹⁹ corresponding to an energy consumption of 859 kJ/mol of CO produced. Simultaneously, process integration constrains the design space, leading to trade-offs, which must be carefully considered.²⁰ This was recently highlighted by Almajed et al.,¹¹ who marked the pH-mismatch between the capture (13.70 ≤ pH ≤ 14.00) and conversion (10.40 ≤ pH ≤ 11.15) steps as one of the biggest challenges for the integration of DAC and bicarbonate electrolysis.

In this work, we propose the integration of a dual sorbent system with bicarbonate electrolysis for direct air capture of CO₂ and conversion to CO. This process combines the aforementioned advantages of no thermal cycling and low water loss, while the use of an intermediate amine-sorbent works to close the pH-gap between the capture and conversion steps. We first outline the process and our setup. Next, we discuss our experimental results and use them in a simplified process model to explore the scaling of the integrated system. Finally, we provide a perspective on how this technology can be further developed and contribute to the sustainable development goals.

EXPERIMENTAL SECTION

Figure 1 shows the process flow diagram of our proposed system for integrated direct air capture (DAC) and electrochemical conversion using combined solid and liquid sorbents. A detailed instrumentation diagram of the setup is provided in Figure S1. First, the CO₂ is captured by adsorption (Figures 1a and S1a) using an amine-functionalized solid sorbent. This sorbent provides a chemical adsorption mechanism with a large adsorption capacity, rapid kinetics, and stability under the relevant DAC conditions.²¹ In this study LEWATIT VP OC 1065 is used, a commercially available, primary amine-functionalized sorbent frequently employed in DAC studies.^{8,22–25} It forms a carbamate species with the captured CO₂ according to reaction 1:²²



In this system, a mixture of (humidified) N₂ and CO₂ (425 ppm) was blown over the solid sorbent in the adsorber column until breakthrough was achieved. The outlet flow of the column was directly connected to a CO₂ gas analyzer, continuously measuring the CO₂ concentration. Both before and after the column, the relative humidity of the air was monitored (Figure S1a).

An alkaline 0.5 M K₂CO₃ solution was used to rinse the saturated sorbent (Figures 1b and S1b). In this step, the adsorbed carbamate is

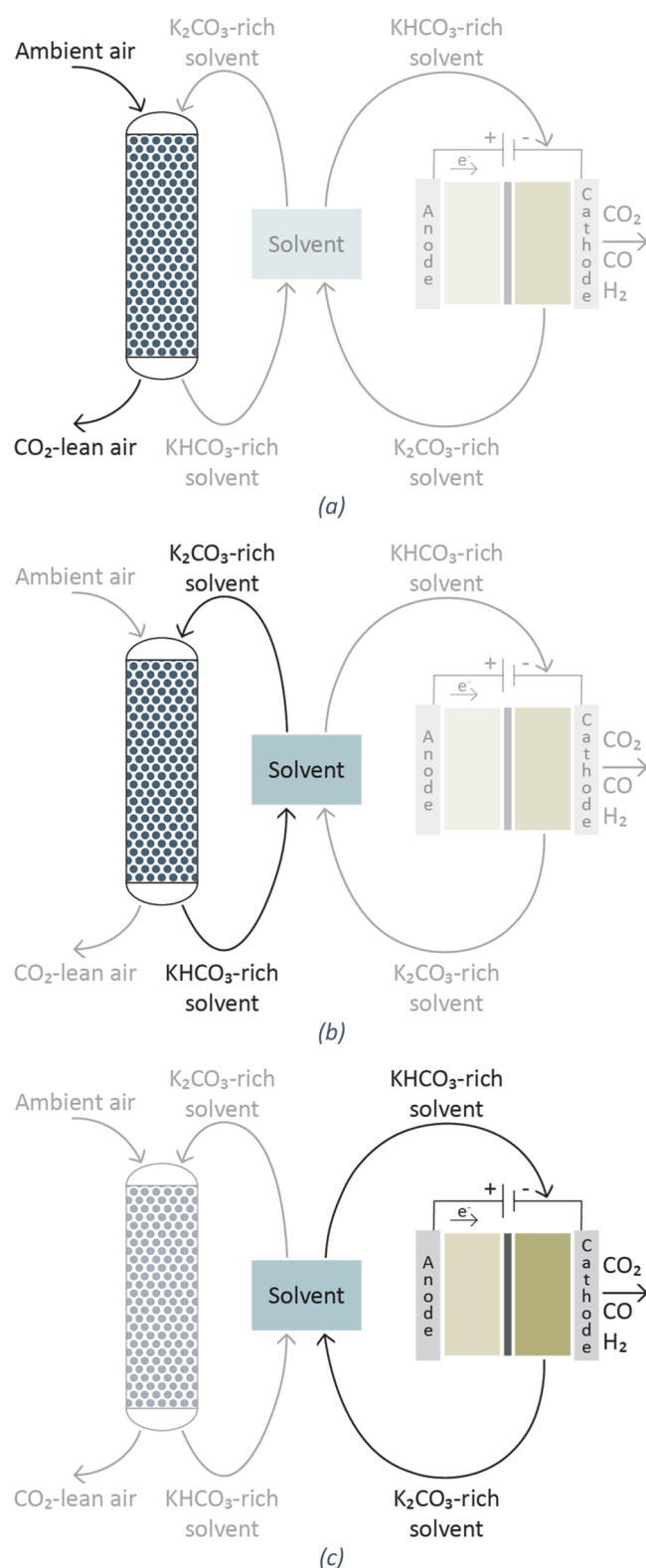


Figure 1. Process flow diagram of integrated DAC and electrochemical conversion. The faint parts indicate components inactive during each step. During adsorption (a), air is fed to the column and CO₂ is adsorbed on the sorbent. During desorption (b), the solvent is passed through the column and CO₂ transfers from the solid sorbent to the liquid solvent. For the conversion step (c), the loaded solvent is recirculated through the electrochemical cell, releasing CO₂ and producing CO and H₂ at the cathode and producing O₂ at the anode (not depicted).

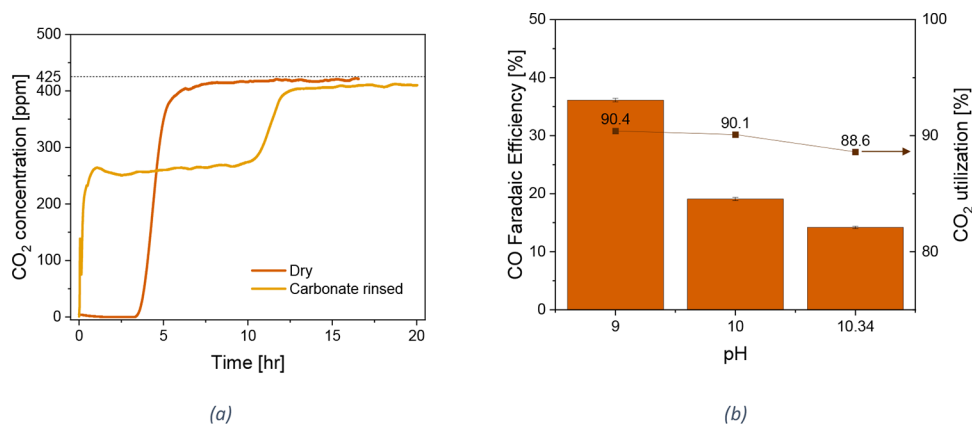
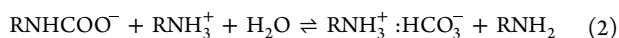
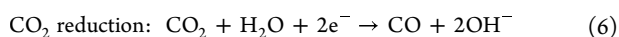
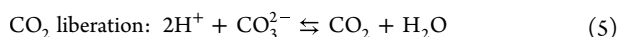
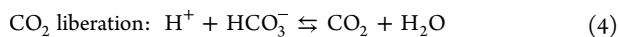


Figure 2. Experimental results for the integrated capture and conversion process. (a) Breakthrough curves for the fresh and dry adsorbent (orange line) and carbonate rinsed sorbents (yellow line). Electrolyzer performance using the obtained electrolyte after desorption (pH 10.3) and electrolyte at pH 9 and 10 presented in (b) Faradaic efficiency toward CO as a function of inlet pH (bars) as well as the CO₂ utilization (markers).

hydrolyzed and released from the resin to form KHCO₃, regenerating the sorbent according to reactions 2 and 3. Though mechanistic studies for similar systems exist (e.g., Said et al.,²⁶ Matsuzaki et al.²⁷), the exact mechanism through which LEWATIT VP OC 1065 is regenerated by the (bi)carbonate solution is unknown and is a subject for further study.



Second, the regeneration of the liquid capture solvent is achieved by electrochemical conversion of the solution using a bicarbonate electrolyzer (Figures 1c, S2, and S3). A full description of the electrochemical setup and catalyst preparation is given in Supporting Information S3 and S4, respectively. In the electrolyzer, the CO₂ is first liberated in situ by the acidic environment close to the bipolar membrane (reactions 4 and 5). Next, liberated CO₂ is reduced on the silver catalyst (reaction 6).



The bipolar membrane provides a constant flux of H⁺ to the cathodic compartment, creating the acidic environment required for CO₂ liberation. This results in a higher local CO₂ concentrations near the catalyst surface as compared to the system using dissolved CO₂, as these systems are limited by the CO₂ solubility in water.^{19,28} By removal of the CO₂ from the solution, the pH is increased such that it can be recycled back to the capture step for a new cycle of resin regeneration. In Figure S4, the full experimental setup that was used is depicted.

RESULTS AND DISCUSSION

Experimental Results

The integrated capture and conversion process was tested consecutively for direct air capture, both adsorption and desorption, and the conversion process. In Figure 2a, the breakthrough curve for the amine-functionalized solid sorbent is presented for the initial dry sorbent (orange line) and the carbonate rinsed sorbent (yellow line). Breakthrough was obtained after 5–6 h, with a total capture capacity of 0.91 mmol CO₂/g sorbent using a fresh batch of the solid sorbent, in good correspondence with previous literature data.²³ After regeneration by rinsing with a carbonate solution, the measured breakthrough capacity was 0.87 mmol of CO₂/g,

indicating the sorbent bed maintained its absorption capacity. However, after regeneration, a significant decrease in the adsorption rate was observed. We observed that the particle bed remains saturated with liquid after draining due to the small particle size and resulting strong capillary forces and then dries over the course of the adsorption time. Therefore, we hypothesize that the CO₂ mass transfer is slowed down due to the liquid blocking the pores. This effect increases the adsorption time for the second and any consecutive cycle. We also observed some initial (<10 min) fluctuations in the wet adsorption experiment, presumably due to hydrodynamic effects in the wet bed (i.e., channeling flow), though their effect on the measured capacity is small (<1%).

Initial desorption experiments show a minimal decrease in pH of the solvent due to the relatively low mass of CO₂ capture in the lab-scale capture column (using a total of 20 g of LEWATIT VP OC 1065 sorbent). To obtain a sufficient volume of loaded solution for conducting electrochemical experiments, the same solvent was ran through multiple desorption cycles, reloading the column of CO₂ between each and progressively lowering the solvent pH. The optimal operating conditions for the bicarbonate electrolyzer are at a near-neutral pH of around 8.5.¹⁷ In Figure S5a, the dissolved inorganic carbon (DIC) concentration and pH of the solvent are presented as a function of four adsorption–desorption cycles. As the pH decreases, the CO₂ capture capacity of the solvent decreases, leading to less deep regeneration of the solid sorbents with each cycle and simultaneously reducing the capture capacity of the solid sorbents. The solvent initially had a pH of 11.9 and a dissolved inorganic carbon (DIC) concentration of 0.49 M. After the four cycles, the pH decreased to 10.3 and the DIC concentration increased to 0.74 M.

The final capture solution obtained after four adsorption–desorption cycles with a pH of 10.3 was then used directly as input for the electrochemical conversion in the bicarbonate electrolyzer. The performance of the bicarbonate electrolyzer, shown in Figure 2b, is highly dependent on the electrolyte pH; therefore, two other solvent pH points were included to highlight the change in performance as a function of pH. A Faradaic efficiency (FE) toward CO of 14% is obtained at an inlet pH of 10.3, resulting in an energy consumption of 5383 kJ/mol CO produced and a voltage efficiency of 39.2% (see Supporting Information S6 for calculation). The performance

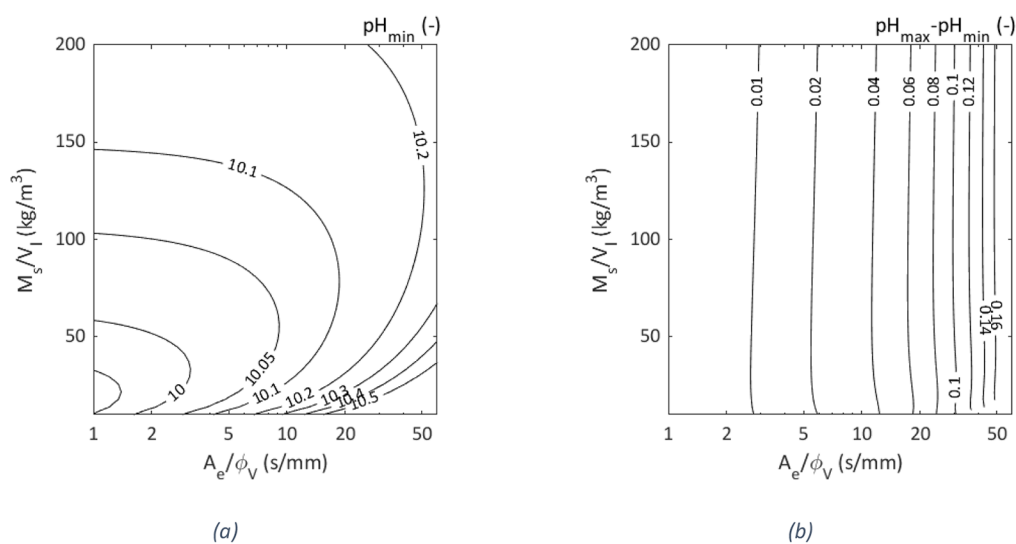


Figure 3. Calculated results of pH of the (bi)carbonate-rich solution (pH_{\min} , (a)) and pH-difference between (bi)carbonate-rich and (bi)carbonate-lean solutions ($pH_{\max} - pH_{\min}$, (b)) as a function of electrolyzer size (A_e/ϕ_V , electrode area per volumetric solvent flow rate) and column size (M_s/V_b , sorbent mass per solvent volume). Results were obtained from the model described in the [Supporting Information S7](#).

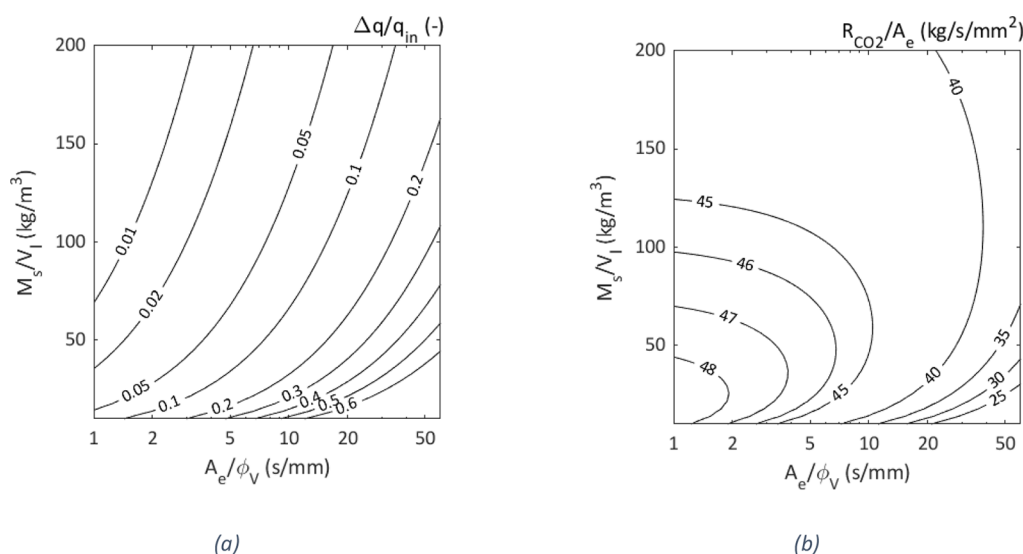


Figure 4. Calculated results of regeneration efficiency ($\Delta q/q_{in}$, loading difference before and after regeneration over initial loading) (a) and electrolyzer productivity (R_{CO_2}/A_e , CO₂ capture mass rate over electrode area) (b) as a function of electrolyzer size (A_e/ϕ_V , electrode area per volumetric solvent flow rate) and column size (M_s/V_b , sorbent mass per solvent volume). Results were obtained from the model described in the [Supporting Information S7](#).

can be improved to 36% FE_{CO} (2131 kJ/mol of CO, 41% voltage efficiency) by decreasing the pH to 9. These voltage efficiencies are aligned with the range commonly encountered for low-temperature CO₂ electrolysis (35–65% at 100 mA/cm²²⁹). The total output gas flow of the electrolyzer includes a mixture of CO, H₂, and unconverted CO₂, creating a relevant syngas mixture. Assuming a renewable electricity price of 10.8 \$/MWh,³⁰ electricity costs are 8 \$/GJ_{syngas},³¹ representing roughly half of the current total production cost of syngas from fossil sources (0.2 \$/Nm³ \approx 16 \$/GJ_{syngas}³¹). Here, we represent syngas costs based on heating value to normalize for its composition, while actual value varies with composition based on the desired application. In [Figure S5b](#), the total concentrations of CO and CO₂ measured at the outlet of the electrolyzer are presented as a function of pH. These values were used to calculate the CO₂ utilization. Here, the utilization

of CO₂ is defined as the fraction of CO over the sum of CO and CO₂.¹⁷ The utilization of CO₂ is approximately 90% for all three tested solvent pH. Therefore, the concentration of unconverted CO₂ in the outlet is limited.

MODEL RESULTS

Next, we used our experimental electrochemical results obtained as input for a simplified process model. These calculations evaluate the process conditions at steady state when the capture and conversion steps are in equilibrium with each other. They are based on the chemical equilibria between the dissolved and adsorbed carbon species during sorbent regeneration and the electrolyzer performance metrics (Faradaic efficiency and CO₂ utilization) observed in our experiments. The purpose of these calculations is to explore the effects of scaling of the capture and conversion devices on

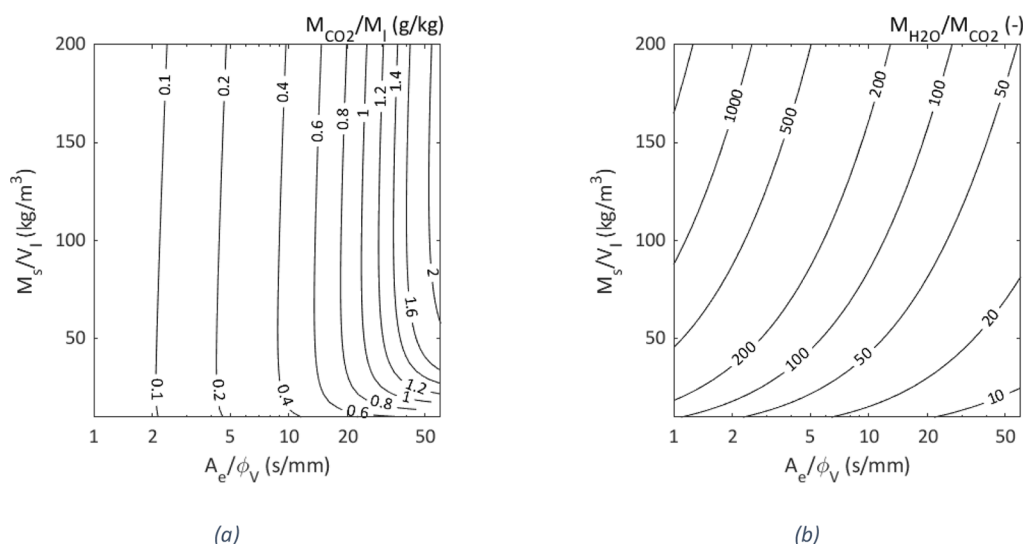


Figure 5. Calculated results of solvent productivity (M_{CO_2}/M_s , CO₂ capture mass rate per solvent mass flow rate) (a) and water loss (M_{H_2O}/M_{CO_2} , water mass per CO₂ mass) (b) as a function of electrolyzer size (A_e/ϕ_V , electrode area per volumetric solvent flow rate) and column size (M_s/V_l , sorbent mass per solvent volume). Results were obtained from the model described in the Supporting Information S7.

the integrated system. While a more detailed, rate-based approach would be required to, e.g., develop a full process design, the current simplified method provides useful a priori insight in the feasibility of integration. A detailed description of the model calculations is provided in Supporting Information S7. All parameters and their justifications are described in Table S1.

Here, we present our model results as a function of the normalized electrode area and normalized solid sorbent mass. The horizontal axis represents the electrode area available per volumetric liquid flow rate and should be interpreted analogous to the electrolyzer size or the residence time of the liquid in the device. The vertical axis represents the liquid–solid ratio during the regeneration step and should be interpreted as the adsorption column size relative to the liquid flow. Our experimental conditions correspond to the central point in each figure ($A_e/\phi_V = 9$ s/mm, $M_s/V_l = 100$ kg/m³).

Figure 3a shows the pH of the (bi)carbonate-rich solution at the adsorber outlet. We observe almost semicircular isolines; for a constant electrode area, increasing the sorbent mass first decreases the outlet pH to a minimum value and then increases the pH again. For a small sorbent mass, the solvent pH is limited by the availability of CO₂ on the loaded sorbent. The lowest pH at the electrolyzer inlet is achieved when the capture and conversion steps are carefully balanced. When the column is further oversized, the sorbent working capacity (Figure 4a) is limited by regeneration in the electrolyzer, again raising the pH and resulting in a decrease in the electrolyzer performance. The achievable pH swing between the rich and lean solutions (Figure 3b) is governed almost exclusively by the electrolyzer sizing and is mostly invariant to the sorbent mass.

The most efficient use of the sorbent (Figure 4a) is found at a large electrode area, capable of achieving a large pH swing. For a small electrode area, the regeneration efficiency (fraction of adsorbed CO₂ removed from the sorbent during regeneration) can be as low as 1%, whereas a large electrode area can boost the regeneration efficiency as high as 60%, comparable to TVSA processes.⁸ Under these conditions, 0.87 mmol/g capacity and 12 h cycle time (Figure 2) and assuming sorbent lifetime and cost of 2 years³⁰ and 30 \$/kg³⁰,

respectively, sorbent costs are 900 \$/tonne_{CO₂}, compared to 540 \$/tonne_{CO₂} for TVSA.³⁰ Previous results have shown that eliminating regeneration at elevated temperature in the presence of oxygen (100 mbar air, 80 °C) decreases the deactivation rate by a factor 6,³² potentially bringing down sorbent costs to 150 \$/tonne_{CO₂}.

High regeneration efficiency comes at the cost of decreased electrolyzer productivity (Figure 4b). As described previously, the electrolyzer performance strongly depends on the electrolyte pH. An excessively large electrode area results in a diminishing Faradaic efficiency and CO₂ conversion, corresponding to a decreasing CO content in the product stream, both from increasing H₂ production and decreasing CO₂ conversion (Figure S7). Combined, these effects confirm that careful balancing between capture and conversion steps is needed to make efficient use of the equipment.

Figure 5a shows the solvent productivity, which is governed by the (bi)carbonate-lean and rich stream pH values. It is largely determined by the pH swing between them and therefore only dependent on the electrode area. A notable exception is found for the combination of a large electrode area and small sorbent mass, in which case the solvent productivity decreases sharply. The high pH under these conditions places the process in a window where little bicarbonate–carbonate interconversion occurs, leading to a decreased uptake/liberation capacity and, by consequence, poor electrolyzer performance.

One of the potential advantages of the presented process is a lower water loss compared with liquid-DAC, as it avoids direct contact between the solvent and air streams. In our experiments, we observed a water loss equal to the entire interstitial volume of the adsorber column. Applying this to our model, Figure 5b shows that in the current unoptimized column design, water loss is significantly higher than in liquid-DAC, by a factor ≈ 8 . By changing the sizing of adsorption and conversion steps, it can be reduced to 10–20 kg/kg, comparable to liquid-DAC.¹² The minimum water loss is found at maximum sorbent utilization, meaning the sorbent goes through fewer regeneration cycles per kg of CO₂

captured. In closed-loop operation, the continuous water loss would lead to concentration of the solution, increasing its pH and negatively affecting electrolyzer performance. Therefore, makeup water should be introduced before rinsing the adsorbent, such that the pH of the (bi)carbonate-rich solution remains constant between rinsing cycles. Assuming the cost for seawater desalination using renewable energy to be $\$5/\text{m}^3$,³³ this incurs costs of 50–100 $\$/\text{tonne}_{\text{CO}_2}$.

Future Perspective

The proposed novel integration of direct air capture using a dual sorbent system with the electrochemical conversion in a bicarbonate electrolyzer is a promising yet challenging method. As previously discussed by Almajed et al.,¹¹ the most challenging part of integrated carbon capture and conversion is the coupling of the electrolyte streams at a pH condition that is suitable for both systems. The electrolyzer product selectivity decreases with an increasing electrolyte pH, whereas the CO_2 capture capacity increases for an increased electrolyte pH. The integrated nature of the process provides a constrained design space.²⁰ A trade-off between optimal capture and conversion conditions is therefore inevitable.

In the presented study, the electrolyzer produces a mixture of CO , H_2 , and unconverted CO_2 , creating a syngas. The ratio in which this gas mixture is obtained determines the application of the syngas and therefore the relevance of the product. Typically, a 2:1 H_2 : CO syngas ratio is required for Fischer–Tropsch synthesis of long-chain hydrocarbon products, such as synthetic fuels.³⁴ Correction of the product gas composition can be achieved through, for instance, a high-temperature reverse water–gas shift step. However, preferable over adjusting the syngas ratio, we believe it is crucial to improve the performance of the electrolyzer, specifically at a higher pH. The electrochemical step is dominating the energy consumption of the overall process, as estimates of the fan and pump power are at least 2 orders of magnitude smaller⁸ than the conversion energy reported here. By improving the FE at a higher pH, the energy consumption is brought down, while simultaneously the pH swing can be enlarged, allowing for a more efficient use of the sorbent. We therefore see a role for further development of electrocatalysts for bicarbonate reduction. Additionally, increasing the operating temperature is expected to improve the electrolyzer performance as the CO_2 liberation from the (bi)carbonate solvent is enhanced and the reaction kinetics increased.³⁵

One of the additional potential benefits of the proposed system is the decrease in water loss by elimination of direct contact between the liquid solvent and the air. However, our present results instead show a remarkably higher water loss, roughly 8 times higher than liquid-DAC. We have identified that this can be reduced to levels comparable to liquid-DAC by adjusting the electrolyzer sizing. However, more importantly than sizing, water loss can be reduced through improved column design. In our experiments and calculations, the water loss was equal to the static liquid holdup in the column after regeneration. Without any additional purging or drying steps, this is the water volume evaporated from the atmosphere during the subsequent adsorption cycle. In our current column design using the commercial LEWATIT VP OC 1065 sorbent, we observed the static holdup to be equal to the entire interstitial volume (37% of the column volume), due to the small particle size ($\approx 500 \mu\text{m}$ ³⁶) and resulting strong capillary forces. In contrast, structured packings for trickle columns,

such as the Mellapak Y packing often employed in liquid-DAC set-ups,³⁷ can have a static holdup as low as 3%.³⁸ We envision that water loss can be reduced up to an additional factor 10 by optimizing the sorbent material and shape (increasing particle size and contact angle) and operating procedure. In addition to reducing water loss, we believe that these measures would also contribute to reducing the pressure drop and alleviating the mass transfer limitations observed during adsorption on the wet column.

Lastly, one parameter left untouched in our analysis is the type of solid sorbent material used. Our choice was based on a commercially available ion-exchange resin (LEWATIT VP OC 1065), functionalized with benzylamine groups ($\text{p}K_{\text{a}} = 9.34$ ³⁹). A less hydrophilic support material is desirable for reducing the water loss and mass transfer limitations. Choosing a material with a lower $\text{p}K_{\text{a}}$ will likely result in a lower absorption capacity but will also allow for a deeper regeneration at lower pH. This once again underlines the fundamental trade-off between the capture and conversion steps. The current work has provided a proof-of-concept for the dual-sorbent capture and conversion process and has highlighted focus areas for future development. Vital future research should include experimental demonstration of the closed-loop process to show the equilibrium between capture and conversion steps, long-term stability of materials, and forming the basis of more elaborate predictive process models and techno-economics, as well as an engineering design study on the air contactor, optimizing it for low liquid hold-up and efficient airflow.

CONCLUSION

In this work, we describe a novel dual-sorbent system for integrated direct air capture and conversion of CO_2 for the production of syngas from ambient air and (renewable) electricity. Through the combination of a solid, amine-functionalized sorbent and an alkalic solvent, this system is able to bridge the pH-gap between the capture and conversion steps found in single-sorbent systems. Based on our experimental and model results, we conclude that the proposed system is technically feasible and capable of producing syngas with limited ($<13 \text{ mol } \%$) CO_2 content, albeit at relatively high ($\text{H}_2/\text{CO} \approx 4\text{--}5$) syngas ratios. The pH swing between the capture and conversion steps is a vital design parameter and is mainly governed by the electrolyzer sizing. A large pH swing results in diminishing electrolyzer productivity through decreased Faradaic efficiency and CO_2 utilization. Conversely, a small pH swing results in inefficient use of the solid sorbent and excessive water loss. As such, a careful trade-off between these opposite effects must be made when considering the design of this integrated capture–conversion process. We identified the electrolyzer Faradaic efficiency at high pH (>9) as the main constricting factor for the process performance, highlighting a focus area for future electrocatalysis development. Furthermore, to reduce water losses to feasible levels, it is vital to revisit the packing material and design and minimize capillary effects.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.5c07683>.

Experimental setup including photographs, additional experimental results, details on efficiency calculation, model description, and additional model results (PDF)

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Notes

The authors declare no competing financial interest.

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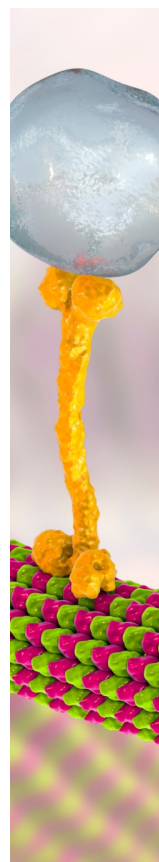
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