

Perspective

Translatable reporting of energy demand and rates in electrochemical carbon capture

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Electrochemical carbon capture has recently emerged as a viable alternative to temperature-swing carbon capture due to its comparatively low energy demands. However, as a new research area, the experimental and measurement practices have not been standardized, making it difficult to make comparisons among studies. Guided by questions of relationships, we critically review the energy and rate evaluation metrics in the electrochemical carbon capture literature to develop a set of guidelines to make new studies more meaningful and useful for future technology transfer efforts. We demonstrate the need both for more transparent reporting due to the ways that experimental choices such as feed and outlet gas compositions influence these metrics and for careful consideration of how experimental details translate to practical applications at scale. This work is centered on capture from stationary energy generators but briefly mentions special considerations when applying the technology to direct air capture.

INTRODUCTION

Carbon capture technologies are essential for addressing climate change,¹ but despite decades of progress, they remain too expensive for wide-scale implementation, largely due to high energy demand (high OPEX) and/or low CO₂ capture rates (large absorbers and consequently high CAPEX).^{2,3} Electrochemical carbon capture has shown promise in the recent decade due to its lower minimum energy demand.^{4–7} This process uses reducing and oxidizing conditions as analogs to the low and high temperatures in typical temperature-swing capture to drive CO₂ absorption and desorption (Figure 1). Consequently, the process avoids the limits of the Carnot efficiency and leverages existing knowledge from large-scale electrochemistry such as fuel cells and battery research.

However, in its infancy, electrochemical carbon capture has not yet established standard practices for its evaluation metrics and benchmarks. The lack of standards limits the ability of researchers to determine whether their developments improve upon prior CO₂ capture studies, both electrochemical and non-electrochemical, because measurements such as the energy required per mole of CO₂ can differ dramatically under different experiment conditions. It is therefore difficult to discern whether improvements in specific evaluation metrics are due to novel chemistries, new reactor designs, or artifacts of different assumptions built into the experiment designs. Additionally, the lack of clear and consistent benchmarks or targets means that even if direct study-to-study comparisons were possible, it is difficult to contextualize those studies with industry needs and wants. Both problems of ambiguity can limit the quality

and coherence of new research in the field due to the inherent uncertainties, perpetuating the issue.⁸

This perspective assembled the reported evaluation metrics, benchmarks, and targets for electrochemical carbon capture and evaluated their utility for comparing experimental results across chemical mechanisms (electrochemical studies to each other, electrochemical studies to non-electrochemical studies), scales (bench to pilot), and use cases (variable energy and CO₂ sources) with the aim of developing guidelines for future electrochemical capture studies. Drawing from the feminist and Indigenous/decolonial scholarship^{9–17} and IEEE's P7000 standard for ethically informed design,¹⁸ our focus was on relations: How do these metrics aid in relating one study to another^{11,12,14,17}? How do different system metrics interact^{11,16,18}? How can and do study designs imply the projected future environments, stakeholders, and infrastructures that electrochemical carbon capture will touch^{9,10,13,15,17}? Like much of the scholarship that inspired our approach,^{9,10,12,15} these questions were first addressed through critical theoretical interrogations, but these critical inquiries led us to systems engineering analyses that further substantiated our claims. In bringing our socioethical analyses in conversation with material (physical) analyses,^{17,18} we hoped to make our assertions more accessible to the electrochemical carbon capture community. With this audience in mind, the analysis we presented here focused primarily on the material systems analyses using analogous technoscientific terminology and only contextualized our conclusions with social (feminist, Indigenous/decolonial, ethical) theories when absolutely necessary (Table 1). While a thorough understanding of these concepts is not necessary to understand



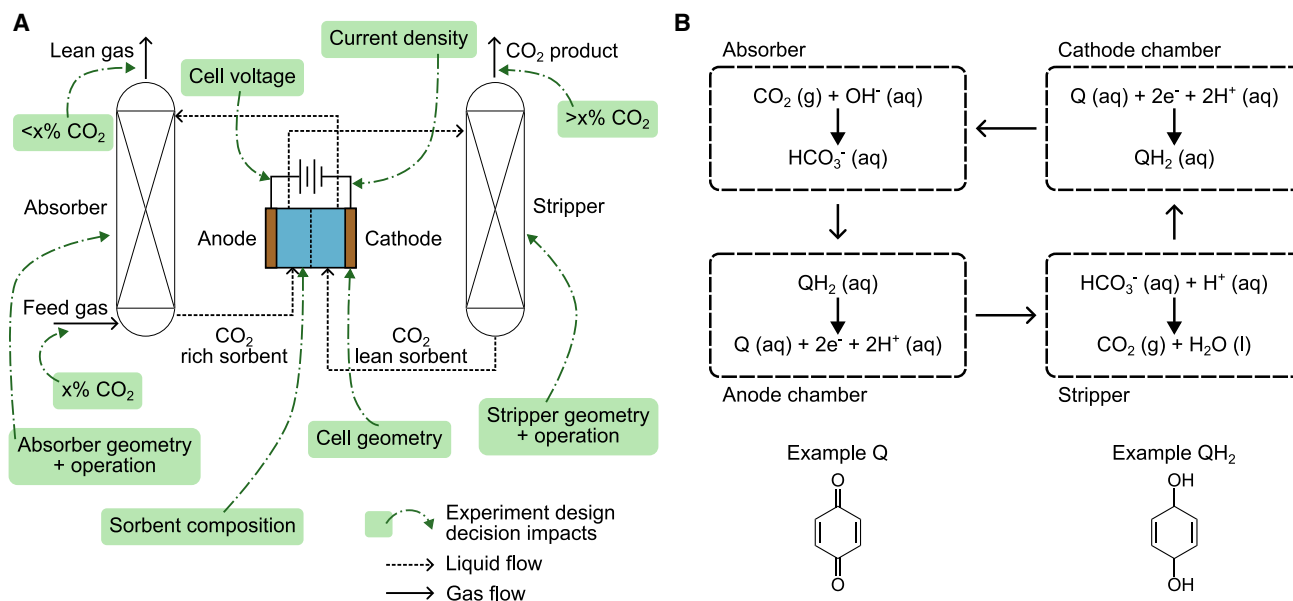


Figure 1. Electrochemical CO₂ capture process and relevant variables

(A) Example schematic diagram of an electrochemical carbon capture plant with key bench-scale experiment design decisions (green boxes) and their direct impacts noted.

(B) One example of the processes taking place in the absorber, anode chamber, stripper, and cathode chamber. For a complete description of possible mechanisms, see.^{4–7}

our analysis, we present these concepts here to serve as an example of the analytical transparency that we advocate for in this work.

Our relational analysis highlighted the shortcomings and opportunities for improvement within electrochemical carbon capture’s measurement and reporting practices, leading to our best practice guidelines. These guidelines are designed to be flexible to account both for the multiple contexts in which electrochemical carbon capture may be applied and for the field’s infancy and consequently, the wide variation in reactor designs and chemistries currently being researched. We therefore focused our attention on what gets reported and how those data are framed – that is, the situatedness of the knowledge produced by the studies.^{9,10,12,16} Metrics should serve as a bridge between experimental setups to showcase improvements and optimization, but haphazard metric selection, as is the current standard, prevents comparisons and limits interpretability, worsened further by a lack of transparent reporting. In keeping with our feminist and decolonial analytic, we have refused, rather than neglected,^{15,19} to impose universal and standardized rules for instrumentation, experimental setups, and experimental parameters as may be typical of other best practices analyses – that is, we do not advocate for every researcher to use identical experimental setups, conditions, and/or procedures. We instead focus on the necessary standards for what makes a study’s results translatable across different experimental setups and conditions while maintaining both scientific rigor and room for creative advancements. Many of our metrics reporting guidelines are based on common practices in the non-electrochemical literature, recognizing both that electrochemical capture has emerged from and will not likely change non-electrochemical capture’s

experimental practices and that non-electrochemical studies are vital benchmarks for comparison given their more robust literature, including technoeconomic assessments.^{2,3}

We focused on bench-scale metrics of energy demand and CO₂ capture rate because these are the two primary limitations of existing temperature-swing carbon capture processes due to their strong contribution to cost.^{2,3} Our attention to the CO₂ capture rate is motivated by the underreporting of this variable ($\approx 17\%$ of papers) despite (1) being the rate-limiting step in the overall process, (2) dictating the size of the absorber, which has the largest capital cost contribution of any system component, and (3) possessing an inherent trade-off with energy demand.^{2,20,21} While other bench- and industrial-scale metrics, such as potential public health and environmental justice impacts,^{22,23} emerge when examining this technology relationally, there are no published measurement practices to assess, and thus we have opted to ignore those other metrics in our analysis and guidelines for now. Similarly, while other bench- and industrial-scale metrics, e.g., electrode material and electrolyte choices, may have important impacts on the overall cost of carbon capture, the lack of data on how those choices affect overall cost, i.e., their direct contributions to the capital, fixed, and variable costs combined with their indirect impacts via affecting energy demand and rate, makes their analysis active avenues of research outside the scope of this perspective.

Our relational approach highlighted the impact of specific experimental conditions on energy and rate across scales and use cases, which we demonstrated quantitatively using existing thermodynamic and systems models. With these experimental impacts in mind, we conclude that the most translatable energy and rate metrics are either condition-invariant or transparent in

Table 1. Concepts and approximate technoscientific analogs guiding our analysis

Social theory concept	Definition	Technoscience analog	References
Situated knowledge	Knowledge is produced by biased actors in specific circumstances, and should be interpreted through those contextual lenses. Contrast with: Universalism	Science and engineering assumptions	Haraway ¹⁰ ; Oliver ¹²
Strong objectivity	Due to the situatedness of knowledge, objective truth is only possible through transparency of contexts, including personal and epistemic standpoints. Contrast with: the “God trick”	Transparent reporting	Haraway ¹⁰ ; Oliver ¹² ; Harding ¹⁶
Relational thinking	Asking questions about and considering systems as wholes, including, but not limited to: physical environments, internal systems, social circumstances, temporal changes. Contrast with: Atomistic paradigm	Systems analysis	Liboiron ⁹ ; Walker ¹⁴ ; Duarte ¹⁷
Refusal	The practice of (1) deciding, based on ethical principles, that certain questions should remain unanswered or certain answers should be withheld at this time and (2) redirecting attention to alternative questions and answers that align with those ethical principles. Contrast with: Nescience (unknown unknowns)	None	Flores ¹⁵ ; Tuck and Yang ¹⁹

their dependencies, showing unambiguous relations to experimental choices.^{16,24} Through these relations among studies, experimental scales and circumstances, and energy and rate metrics’ use and discussion, we highlighted some of the inherent limitations of singular, context-independent universal metrics and benchmarks, consistent with the science and technology studies literature,^{10,12,24,25} supporting our refusal to suggest standardized rules. However, while universal metrics are limited, we show that the following measurement and reporting practices will improve the electrochemical capture literature regardless of contextual use case if universally adopted.

- Energy consumption and CO₂ absorption kinetics must be studied in tandem to be useful due to inherent energy-rate trade-offs.
- The feed, lean, and product gas concentrations must be reported because both energy and rate can be artificially manipulated by using industrially irrelevant concentrations.
- Energy consumption comparisons between electrochemical and temperature-swing capture must include situation-specific heat-to-electricity efficiency losses.
- Rate measurements are most useful when either scale-independent (reaction rate constants) or reactor geometry and scale explicit (capture flux).

These guidelines are designed to navigate multiplicity and specificity such that fair comparisons can be made without resorting to the common problem of dislocation, which has been shown to weaken objectivity claims.^{16,24,25} By following these guidelines, researchers will make transparent their study’s most impactful experimental and epistemic relations that could both explain inter-study variability and emplace envisioned use for more targeted technology transfer. It is only through transparency that different relations can be accounted for when making

comparisons across use cases and embedded assumptions. While not all of these guidelines may be relevant for every study depending on research goals and usage contexts, the choice to ignore a guideline should be rationalized and articulated, particularly when assessing and comparing study results. We showcased the importance of emplacement by focusing our analysis on point-source capture from power plants, the most commonly studied scenario, followed by a brief adaptation of our analysis for direct air capture based on its unique relationships.

ENERGY DEMAND

Presently used metrics and benchmarks

The low minimum energy demand of electrochemical carbon capture has been the primary motivation for studying this technology over its temperature-swing counterpart. This has consistently been presented in terms of energy consumed per mole of CO₂ captured (kJ/mol C) in both the electrochemical and non-electrochemical carbon capture literature.^{4–7,26} To compare these technologies, the temperature-swing process has often been cited as approximately 100 kJ thermal energy (kJ_t) per mole of CO₂ (e.g.,^{27,28}), albeit without substantive evidence. Recent articles remedy this issue by citing experimental measurements of the temperature swing process in the range of 90–185 kJ of thermal energy (kJ_t)/mol C^{29–31} or 35–60 kJ of electrical energy (kJ_e)/mol C.³²

Because the wide range of temperature-swing energy demands comes from both variability in experimental setups and differences in assumptions of the amount of process heat that can be leveraged,^{33–35} it is difficult to compare the energy demand of temperature-swing capture directly to that of alternatives such as electrochemical capture. A better option would be to use a target energy demand, such as the US Department of Energy’s estimate that retrofitting a power plant with carbon capture

Table 2. Power plant type and target energy demand

	Power plant efficiency (%) [38–42]	Flue gas CO ₂ % [38, 43–50]	W _{min} (kJ/mol C) 90% removal, 95% purity	Scaling factor (median ^a)
Coal	Subcritical: 33–39	12–15	5.85–6.44	1.0
	Supercritical: 38–44			1.1–1.3 (1.2)
	Ultrasupercritical: 43–47			1.2–1.3 (1.3)
Integrated gasification combined cycle	39–43	12–15	5.85–6.44	1.1–1.6 (1.2)
Natural gas combined cycle	45–55	3–10	6.91–9.98	1.5–2.7 (2.0)
Biofuel	23–45 (Bioliquid/biogas: 30–45) (Direct biomass: 23–37)	4–15	5.85–9.26	0.7–2.0 (1.3)

^aUsing the median power plant efficiency and the median flue gas concentration

would be viable if it causes the cost of electricity to increase by less than 35%. Two different calculation methods – one based on existing technology benchmarks and another based on power plant energy and CO₂ emission rates – find that achieving this cost target requires the energy demand for carbon capture to decrease by approximately 55% relative to the current state-of-the-art,^{36,37} although they disagree on the exact value (≈ 30 vs. 66 kJ_e/mol C). Notably, these targets are similar to the previously mentioned experimental energy demands. This shows that the literature’s obsession with decreasing the energy demand due to its strong influence on OPEX^{4–7,26–32} is insufficient to overcome the cost barrier for adoption, in large part due to carbon policy assumptions embedded within the energy target estimation.^{36,37} This range of 30–66 kJ_e/mol C, which encompasses both the Department of Energy’s energy demand target and present-day temperature-swing energy demand estimates, is only useful as a benchmark, not as a target. That is, these values serve as useful context, but an electrochemical capture process with an energy demand similar to or less than this range is not a guaranteed breakthrough.

Relation to energy and CO₂ source

Some of the inconsistency in the energy demand of the temperature-swing benchmark can be attributed to an often-invisible unit conversion from thermal energy (kJ_t) to electrical energy (kJ_e) due to reporting simply as energy (kJ). In the temperature-swing process, heat is directly sourced from the combustion reactions that the power plant uses to generate electricity, so the thermal energy can be easily converted to the equivalent amount of electrical energy had carbon capture not occurred by using the power plant’s efficiency. This efficiency varies depending on the fuel and power plant configuration (Table 2), typically 30–50%. Using 35% efficiency, a rough estimate for a subcritical coal power plant, the temperature-swing benchmark measurements of 90–185 kJ_t/mol and 35–60 kJ_e/mol C are roughly equivalent.

This conversion is important when comparing temperature-swing capture to electrochemical capture because the electrochemical reaction is driven by electrical energy, not heat. While electrochemical carbon capture is often claimed to be motivated by its capacity to use renewable electricity, doing so is impractical for capture from power plants, where electricity is already being generated internally. While the variation in power plant efficiencies

adds further uncertainty to the energy target, both the experimental measurements and the DoE target were based on pulverized coal, the least efficient fuel. In a more efficient power plant, the electrical energy produced by the same amount of heat would be higher, and thus we expect a greater energy demand target that electrochemical capture must fall below for newer facilities. Importantly, if we assume application in a coal-fired power plant, bench-scale electrochemical capture is roughly equivalent to the temperature-swing benchmark in energy consumption, not substantially lower as is often reported in the literature.

We note, however, that the type of fuel also affects the concentration of CO₂ in the flue gas, in turn influencing the energy consumption by affecting the minimum work of separation (Equations 1 and 2)³⁸:

$$W_{\min} = RT \left(\sum_{\text{outlets}} (n_{\text{CO}_2} \ln y_{\text{CO}_2} + n_{-\text{CO}_2} \ln y_{-\text{CO}_2}) - \sum_{\text{inlets}} (n_{\text{CO}_2} \ln y_{\text{CO}_2} + n_{-\text{CO}_2} \ln y_{-\text{CO}_2}) \right) \quad (\text{Equation 1})$$

$$E_{\min} = \frac{W_{\min}}{\Delta[\text{CO}_2]} \quad (\text{Equation 2})$$

where R is the ideal gas constant, T is the absolute temperature, n_i and y_i are respectively the moles and mole fraction of gas i in the relevant gas stream, noted as CO₂ or non-CO₂, and $\Delta[\text{CO}_2]$ is the amount of CO₂ captured. Based on the direct air capture literature, to a first approximation, we assume that the temperature-swing energy demand scales proportionally with the increase in the minimum work of separation, so the impact of energy and CO₂ source on the benchmark electrical energy demand should be adjusted by a proportional scaling factor, SF:

$$\text{SF} = \frac{E_{\min}}{E_{\min}|_{\text{coal,sub}}} \frac{\eta}{\eta_{\text{coal,sub}}} \quad (\text{Equation 3})$$

where η is the heat-to-electrical energy conversion efficiency of the process of interest. While the assumption of a constant

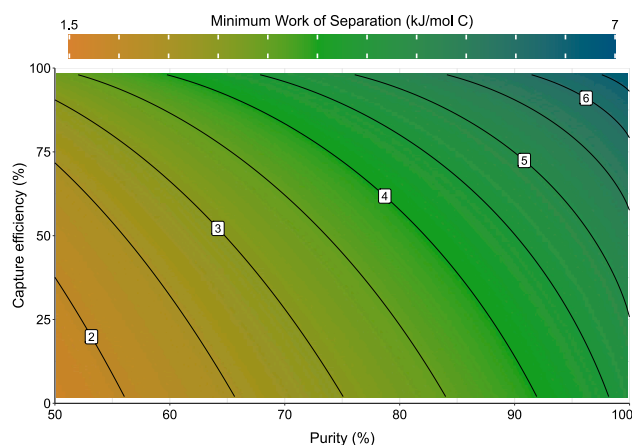


Figure 2. Minimum energy demand depends on CO₂ partial pressures

Minimum work of separation as a function of the CO₂ capture efficiency and product gas purity calculated assuming a 15v% CO₂ feed gas.

2nd law efficiency is not supported when examining across different separations,³⁹ the differences among 2nd law efficiencies of point sources listed in Table 2 are expected to be small relative to the variability among reported energy demands when capturing from the same source concentration. In this equation, a greater scaling factor would mean that the energy target is higher relative to the coal baseline, so electrochemical carbon capture would be competitive at a higher (i.e., less efficient) electrical energy demand. The scaling factor for a natural gas combined cycle shows that while carbon capture would require more energy to capture CO₂ from the more dilute stream, the energy efficiency improvement is more impactful (Table 2). Given the current transition away from coal toward natural gas,^{40,41} electrochemical capture will become more competitive over time due solely to changes in energy sources even if this technology does not become more energy efficient. In other words, the presently collected data (i.e., the experimentally implied future), which assumes coal conditions, is not consistent with likely energy transitions predicted by sociopolitical circumstances, and this inconsistency means electrochemical capture does not appear as promising as it would under a more likely future scenario. This also does not account for how natural gas emissions have lower SO_x, which inhibits most carbon capture sorbents,⁴² meaning long-term stability and efficiency will improve as well. Biofuels, which are expected to become more common due to the negative emissions potential of BECCS,^{1,41} have much more uncertainty due to variability in fuel and plant design from their relative infancy, though they generally provide more favorable conditions for electrochemical capture than coal.

Relation to output gasses

As noted in the minimum thermodynamic work calculation, characteristics of the nearly pure product gas and the CO₂-depleted lean gas also substantially impacts the energy demands (Figure 2). The carbon capture literature has settled

on a CO₂ mole fraction > 0.95 in the product gas with > 90% removal efficiency from the feed gas. While the purity of the product gas cannot fall below \approx 95% without substantial impact on the efficiency of CO₂ compression and pipeline transport,³⁸ the target removal efficiency of 90% seems arbitrarily selected, particularly knowing industrial-scale capture plants rarely achieve \geq 90% capture.^{36,43,44} Despite the substantial impact of these two output gasses on the minimum work of separation, it is common in the electrochemical literature to estimate energy demands without reporting lean or product gas concentrations.^{45–50} In rare cases, studies conducted with unrealistic feed, lean, and product gas characteristics are corrected to explicitly determined gas concentrations using thermodynamic models.^{30,51}

Beyond their impact on the minimum work of separation, these two output gasses have notable impacts on experimental measurements themselves. For instance, it is customary for electrochemical capture experiments to quantify the amount of CO₂ captured by using an inert carrier gas or vacuum pressure over the desorption unit to carry the CO₂ to a detector.^{47,52–54} This is consistent with industrial-scale temperature-swing capture, which uses steam as both a source of heat and a condensable sweep gas to increase the desorption efficiency,^{55,56} but can pose a problem when comparing among bench-scale studies. Two systems with identical electrochemical cells, solution compositions, and operating conditions can have different energy demands if one has a more efficient desorber, such as via a greater sweep gas flow rate, but the cause of that difference would not be known unless desorber conditions were reported.

Similarly, it is not uncommon to allow bench-scale CO₂ absorption to proceed until near-equilibrium with the feed gas.^{30,46,51,54,57,58} While typical absorber units use countercurrent flow to maximize absorption rates, equilibrium is rarely achieved in practice. For instance, CO₂ absorption from coal emissions into 30wt % MEA often exits the absorber at 0.4–0.5 mol C/mol N,⁴³ compared to 0.5–0.6 mol C/mol N at equilibrium.⁵⁹ Like with increased desorption from a sweep gas, assuming equilibration would translate to a greater amount of CO₂ being captured per volume of solution, lowering apparent energy demands at the cost of process rate. While equilibrium can be useful for determining the limits of these systems,^{31,51,60} e.g., asking if 90% removal is possible,³⁰ equilibrium-based energy demands should be clearly discussed as a limiting case. This is particularly true given the increased pumping work required to reach equilibrium, as pumping is rarely considered in electrochemical studies. While pumping work is frequently ignored in temperature-swing capture, when the sorbent regeneration energy demand decreases, pumping work makes up a more substantial fraction of the total energy demand. Ultimately, like with the product gas stream, the issue is not methodological inconsistency but a lack of reporting of critical experimental details despite their impact on the system performance.

Energy demand guidelines

The emphasis on the energy demand of electrochemical carbon capture largely emerged from its emphasis and use in the temperature-swing literature. However, many experimental

conditions for electrochemical capture are not reported, and these conditions have not yet been sufficiently standardized to be confidently assumed, nor, as we note with the changing energy landscape, should those conditions remain static. Without unambiguous and transparent reporting, there will always be the potential to minimize the system energy demand by obfuscating,^{9,11,16} for instance, how little CO₂ was removed rather than scientific or engineering developments. Similarly, the energy demand benchmarks and targets for electrochemical capture depend on the type of power plant being considered. While most carbon capture research focuses on applications for coal, the expected phase-out of coal in favor of natural gas and biomass feedstocks^{1,40,41} means the energy demand targets should change correspondingly. The same electrical energy consumption may therefore be more promising in different socio-techno-environmental contexts,^{10,14,24} so it will be important to be clear what the assumed future contexts, e.g., type of power plant and, consequently, plant efficiency and feed gas contents, are when discussing measurements and conclusions. This contextual transparency is not only important from an isolated monetary cost perspective but also from a broader social perspective, for the addition of an energy-intensive capture unit or facility may have energy justice and electrical grid resilience implications.

Given the importance of these experimental variables in contextualizing the energy demand measurements, we suggest reporting the second law efficiency as a key energy metric. This efficiency compares the experimental and minimum work of separation and consequently incorporates the inlet and outlet CO₂ concentrations. It therefore forces researchers to validate – and ideally report – important CO₂ concentrations that impact the measured energy demand. This is in contrast to the multitude of energy efficiency measures in the electrochemical carbon capture literature frequently based on electron usage stoichiometry^{6,27,29,60–65} or the minimum (Nernstian) electrochemical work.^{30,66} Although these existing efficiency measures are useful for understanding and optimizing electrochemical parameters such as current or voltage, they are incomplete for assessing overall separation performance.

PROCESS RATE

Presently used metrics and benchmarks

Unlike the energy demand, the rate of electrochemical carbon capture has no consistent reporting measure. While the electrochemical capture literature consistently reports electrochemical kinetics measurements, primarily the electric current or current density, our survey of 120 articles from a SCOPUS search found a small fraction mentioning CO₂ mass transport processes (17%). Among articles reporting CO₂ mass transport rates, the units vary, with the most notable being the mass of CO₂ captured per time,^{28,37,45,66} volume of CO₂ captured per electrode area per time,⁴⁷ and volume of CO₂ captured per volume of solution in a batch operation mode.⁵⁴ For most of these measurements, the absorption rate is conducted in a well-mixed sparging setup, which overestimates mass transport compared to industrial absorber towers. Only recently have CO₂ absorption rates have been of interest in the electrochemical capture litera-

ture.^{62,67} More commonly, electrochemical rates (current densities) are reported as the defining rate metric (e.g.,^{5,29,32,48,51,62,64,68}). The fact that these metrics are often dependent on the same variables as those of energy demand^{20,21} means these rate discussions must also always occur in tandem with energy demand.

The inconsistency in rate reporting is further complicated by the incomparability of these measures with temperature-swing capture, which tends to report either the second order (bimolecular) rate constant of the sorbent-CO₂ reaction (e.g.,^{69–71}) or the CO₂ flux across the vapor-liquid interface in an absorber (e.g.,^{71–73}). While none of these measurements have a widely agreed upon target, the results from temperature-swing capture provide valuable benchmarks to assess electrochemical capture's viability. Given that temperature-swing capture inherently lacks electrochemical rates, it is important to consider: what new rates become relevant because of the electrochemical reactions, and what old rates remain important? In the absence of a standardized experimental setup, our analysis focused on what makes particular rate metrics more or less translatable across studies and across scales. Our goal, again, is not to dictate how to conduct a measurement (e.g., dictating the exact absorber to use in testing) but to evaluate different potential metrics (e.g., fluxes vs. rate constants) as optimization targets.

Relation to metric assumptions

Our primary question when evaluating rate metrics in electrochemical capture is what relations are assumed by the measurement.^{9,14,17,24} For instance, the focus on electrochemical rates over mass transport rates implies that the electrochemical rate substantially impacts system performance. However, under typical experimental conditions (current density > 10 mA/cm²^{27,29,54,68,74}), the rate-limiting step will be CO₂ mass transport across the vapor-liquid interface during absorption.^{20,21} As a result, while charge and mass transfer processes at the electrode surface impact the cell overpotential and energy efficiency, improvements to electrochemical kinetics may not have a substantial impact on the overall process time. We argue electrochemical rate measurements are actually indirect energy measurements and provide limited utility when assessing process rate.

Among mass transfer rates, such as all metrics,^{11,13,16} the selection of one metric over another also has assumed relations built into the decision. For instance, the mass or volume of CO₂ per time assumes future scale-up scenarios use the same reactors in parallel; otherwise, there is a risk of continuing to capture the same amount of CO₂ per time despite reactor size increases. It is this reason why the CO₂ flux is more frequently used in the temperature-swing literature: it makes explicit the design variable with which absorption rate scales. However, comparing temperature swing and electrochemical CO₂ absorption fluxes assume similarity in the absorber configuration, e.g., countercurrent or cross-flow and become less useful when comparing different chemistries under different configurations. Similarly, comparisons among second order rate constants assume that molecular kinetics are predictive of reactor-scale kinetics, and thus ignore the relationships with, for instance, diffusion coefficients and viscosity.

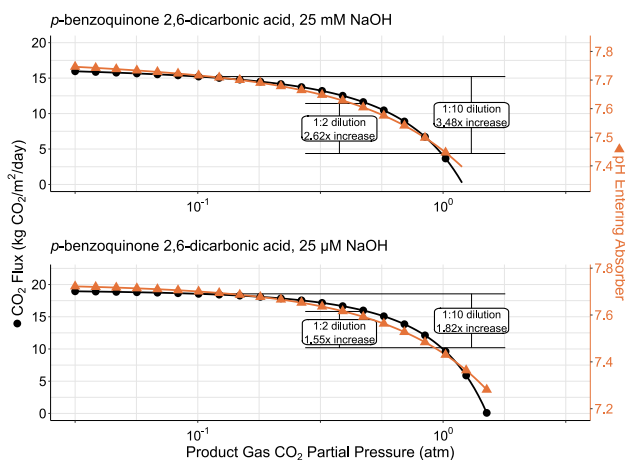


Figure 3. CO₂ dilution in the desorber affects absorption rate

Predicted mean absorber CO₂ flux (left, black circles) and solution pH entering the absorber (right, orange triangles) as the outlet CO₂ partial pressure changes (50 mM quinone, pK_a = 6.01 and 7.75, 0.15 atm CO₂ feed gas). Calculated using the electrochemical pH-swing model developed for.²⁰ Points are a subset of the data for visualization only. Dilution effect is the proportional change in flux relative to a 1 atm CO₂ product.

The ambiguity of what a future absorber will look like at pilot- and industrial scales means there is no singular answer for what metric to pick, but it does not mean all metrics are equally good or bad. While the choice of metric can seem benign, a metric's built-in assumptions can substantially impact design and thus cost.²⁴ We previously used computational models of electrochemical capture via pH swings in two studies, one considering CO₂ flux and another considering minimum requisite residence time.^{20,31} Despite the residence time estimate being based on the same flux model, the two studies found different optimal chemical conditions for CO₂ absorption. Consequently, it is insufficient to study any arbitrary CO₂ absorption rate: one must be deliberate in choosing which rate is most relevant toward the research goals. No measurement is without built-in assumptions,^{10,25} and the choice of metric should be based on which assumptions are acceptable while making those assumptions transparent. If the primary motivation for studying rate is its relation to capital cost, further studies are needed to determine which rate metric most strongly controls capital cost and what that relationship is.

Relation to experimental setups

As with energy demand, experimental flux measurements are dependent on experimental conditions that are not often reported with sufficient detail. For example, it is well understood that CO₂ absorption rates in temperature-swing capture depend on the flow geometry and both the liquid and gas phase velocities (e.g.,^{38,43}), but electrochemical capture rarely reports these variables, let alone examine their impact on performance. Similarly, while the electrolyte composition of electrochemical capture is generally reported, studies tend to ascribe performance differences to molecular species differences,^{64,75,76} despite differences in concentration and starting pH having a substantial impact on absorption kinetics.^{20,31} These concentration differ-

ences may also be obscured by other experimental conditions: more effective desorption conditions, such as by applying a partial vacuum or dilution by a sweep gas such that the outlet CO₂ partial pressure is less than 1 atm, leading to a greater pH at the end of desorption, enhancing absorption rates (Figure 3). Conversely, pressurizing the outlet, which has been proposed to minimize downstream compression energy demands,⁵¹ will decrease both the pH and CO₂ flux. However, the impact of the desorption pressure is dependent on the precise solution chemistry, making it difficult to take into account when relating bench-scale study data to pilot-scale design.

These inconsistencies become more salient when relating bench-scale setups to industrial-scale practices. Electrochemical capture often uses bench-scale sparging setups,^{30,54,57,58} while industrial-scale absorption uses single-pass packed bed towers.^{38,43,77} Consequently, bench-scale mass flow rates are unlikely to reflect real-world scenarios. Similarly, industrial-scale capture typically operates at 40°C, whereas electrochemical capture tends to operate at room temperature (20–25°C); the few electrochemical capture studies examining temperature typically do so with the aim of thermoelectric codrive, i.e., applying both temperature and oxidation state changes to enhance absorption and desorption effectiveness beyond either change alone.^{45,78} While one of the benefits of electrochemical capture is that it can operate under isothermal conditions, the flue gas must be cooled to 40°C for temperature-swing capture, meaning the temperature of the flue gas is not predetermined. A different temperature may maximize the absorption rate or minimize energy demands. Collectively, these experimental differences make it unclear how absorption kinetics measurements from electrochemical capture compare to each other or temperature-swing capture under realistic scenarios, even if they use the same units.

Process rate guidelines

Present measurement practices for electrochemical carbon capture make it hard to understand the process's kinetics. Not only are CO₂ mass transfer rate measurements of electrochemical capture inconsistent with the rest of the carbon capture literature, but they are also hard to compare to each other because metrics such as the mass of CO₂ captured per time depend on infrequently reported flow and reactor geometry conditions. When these experimental conditions are reported, it is often unclear how well the reactor would perform at industrial scales. The fact that absorption rate metrics are only reported in 17% of studies from our literature review despite being the rate-limiting step worsens the problem.

At a minimum, a CO₂ absorption rate, not just the electrochemical reaction rate, must be reported. As long as the measurements are internally consistent and internally benchmarked against controls (e.g.,^{75,79}), relative rates can be qualitatively assessed, providing still-useful data for comparison.¹¹ We believe the most useful metric for comparison to temperature-swing capture is the bimolecular rate constant because it is frequently measured,^{69–71} can be measured potentiometrically in some electrochemical systems,⁶⁷ and can predict reactor-scale fluxes using an absorber model.^{20,31,71–73} For context, most temperature-swing sorbents are primary and secondary amines with

bimolecular rate constants $\geq 10^2$ (Ms)⁻¹; sterically hindered sorbents have rate constants between 10^0 and 10^2 (Ms)⁻¹ and are frequently used alongside primary and secondary amines to ensure competitive rates.^{69–71} From the few electrochemical capture studies that follow these guidelines, electrochemical capture using amines and pH-swing has a similar absorption rate to MEA-based capture,^{31,75,79} but electrochemical capture using quinone-carbonate adducts is more similar to using a sterically hindered amine.⁶⁷ We note, though, that the choice of rate metric should be based on the research question at hand: if assessing developments to the absorber itself, rate constants become less useful, and CO₂ fluxes across the vapor-liquid interface may be more appropriate.

Unfortunately, there is no consensus on any CO₂ capture rate metric that would indicate when the process is viable for commercialization. Ideally, the target would be based on the relationship between the absorber cost and the rate metric, but, to our knowledge, no technoeconomic assessment has made this relationship explicit, instead taking absorber size and operation conditions as given (e.g.,^{2,3,80}). This cost-centered perspective also ignores the impact that low rate processes and the subsequently larger reactors will have on supply chains, including the environmental and social ramifications of increased raw material extraction and processing. In the absence of a clear target, one approach is to determine the rate of the 30wt % MEA benchmark under the same test conditions and assumptions and use that as the benchmark (e.g.,^{20,31}). While this does not account for how the cost of a lower absorption rate may be compensated by a lower energy demand, this approach provides a conservative assessment. However, it is entirely likely that the optimal absorber design will depend on sorbent properties and electrochemical cell operation just like energy demand, and thus a holistic and transparent report of conditions and assumptions is still necessary.

CONSIDERATIONS FOR DIRECT AIR CAPTURE

The feasibility of direct air capture (DAC), like that of capture from power plant emissions, is primarily dictated by the monetary cost per unit of CO₂ captured, much of which is attributable to the energy cost and the capital cost of the absorber.^{80–82} However, DAC differs from point source capture not only in its feed gas CO₂ concentration but also in the industrial ecosystem around it. These contextual differences mean our analyses and suggestions for electrochemical DAC differ slightly from those for power plant capture.^{9,10,12,16}

Energy demand

The most obvious energy difference between DAC and capture from power plants is that it is not coupled to an energy-generating combustion process, so there is no readily accessible source of heat. As a result, it would not make sense to convert the thermal energy demand of temperature-swing capture to equivalent electrical energy using the heat-to-electricity energy efficiency of a power plant. Instead, it would be more appropriate to compare the energy demands adjusted by how the cost of fuel compares to the cost of electricity. However, given that these costs vary with locale, a direct comparison of thermal and elec-

trical energy may be appropriate in the absence of a candidate site. Different energy sources will also have different amounts of embedded carbon associated with their production, penalizing DAC's energy demand by decreasing the net CO₂ removal. As electrical grids shift to renewables, electrochemical DAC becomes more appealing because it incurs a smaller CO₂ emissions penalty than fossil-generated heat.

Experimental measurements for the energy demand target for temperature-swing DAC appear to be stable at approximately 350–400 kJ/mol.^{80,83} This \approx a 4-fold increase relative to coal emissions is almost the increase in W_{\min} for coal (15v%) to air (400 ppm) assuming 90% capture and 95% purity. Notably, DAC design is more concerned with total CO₂ removal than the amount per unit of feed gas because air is in excess, whereas combustion flue gas is not. Consequently, 90% removal from a single pass may not be applicable to DAC, so using lower removal efficiencies is a viable option for lowering the energy cost. This may explain why one electrochemical DAC system has been reported to have an energy demand of 65 kJ/mol C, only double the same system's energy demand from 15v% CO₂: the removal efficiencies for this system are unknown.⁵⁷

While lower removal efficiencies may benefit the energy consumption of the sorbent regeneration process, it will lead to an increase in the mechanical work of pumping because more air must be pumped per unit of CO₂ removed. In flue gas capture, pumping work is generally assumed to be negligible compared to the sorbent regeneration energy,³⁸ but the mechanical work of DAC is larger to accommodate lower CO₂ concentrations. Keith et al.⁸⁰ estimate the fan work at approximately 12 kJ/mol C for DAC with 75% removal, \approx 3% of temperature-swing DAC's sorbent regeneration energy, but almost 20% of electrochemical DAC's. The removal efficiency should therefore be considered an important operating parameter for optimizing DAC by balancing the impact on the sorbent regeneration energy against the mechanical work of pumping. There should be an optimal removal efficiency that minimizes the sum of energy contributions, and this may be unique to the absorber configuration and chemistry.

Capture rate

Similar to capture from the power generators, DAC's cost is indirectly impacted by the absorption rate via reactor size. In order to reach end-of-century climate goals, an estimated 10–1000 Gt of CO₂ must be removed from the atmosphere by 2100.¹ While not all of this will use DAC – a commercializable DAC facility is expected to remove on the order of 1 Mt/year – DAC has two major benefits over other negative emissions technologies: scalability and geographic independence.^{81,84} Due to the low concentration of CO₂ in air, 1 Mt/year is typically achieved by multiple absorbers in parallel^{81,82}; slower processes will require more absorber towers to achieve 1 Mt/year.

Given how the rate-limiting step for capture from coal emissions is already CO₂ absorption, the $>$ 100-fold lower CO₂ concentration in air means the CO₂ absorption process will continue to be rate-limiting for electrochemical DAC. However, with the lack of studies establishing best practices for DAC absorber design and operation, flux measurements will be confounded by reactor design improvements. The rate constant of the

absorption reaction in electrochemical DAC continues to be the more appropriate measure, as this is more comparable with the existing literature.^{69–71} An important note, though, is that most kinetic information for CO₂ absorption has been conducted at 40°C due to the focus on power plant capture, whereas DAC will operate under ambient outdoor air temperatures (global average 14°C–15°C). As with the potential for different temperatures in electrochemical capture from power plants, this simply means there may not be as much data to benchmark against.¹¹

CONCLUDING REMARKS

The energy and rate metrics used in electrochemical carbon capture are inconsistent when compared to other electrochemical capture studies or temperature-swing benchmark studies. As a result, it becomes difficult to determine whether true advancements in electrochemical capture have been made. The inconsistency in energy demand reporting is largely due to the lack of consistent experimental conditions and the lack of transparent reporting of key process variables such as inlet and outlet CO₂ concentrations. In contrast, the inconsistency in rate reporting is due to an overemphasis in the literature on electrochemical rate, which, while relevant to energy demand via the overpotential, does not describe the rate-limiting step. Even the few electrochemical carbon capture studies that report CO₂ mass transport measurements do so using units that are not inherently scalable and without adequate description to infer a scalable metric, making it difficult to discern if an improvement in the rate is due to changes in the reactor design, operation, or chemistry. Given the inherent trade-offs between energy demand and CO₂ absorption rates,^{20,31} it is important to report both metrics transparently to show that improvements in energy demand were not solely at the detriment of rate or vice versa. In other words, we, as electrochemical capture researchers, must be accountable for the ways that our experimental choices and assumptions affect measured data.²⁴

Additionally, as noted previously, these relations imply other metrics important to carbon capture, electrochemical or otherwise. There is, for instance, an abundance of work in the non-electrochemical literature on the loss of amine sorbents under CO₂ absorption conditions over the course of weeks to months,^{85,86} an under-studied timescale in the electrochemical literature. The byproducts of temperature-swing capture are hazardous and volatile,^{22,23} placing carbon capture in conversation with public health. What are the long-term byproducts of electrochemical capture, how do they impact health, and how do efforts to minimize health impacts affect the energy and rate? Metrics such as byproduct release rates and toxicity provide additional axes by which electrochemical and temperature-swing capture can be compared, which may further showcase the benefit of electrochemical capture. For instance, lower temperatures in electrochemical capture will reduce volatile losses, reducing both health consequences and ongoing costs of replacing lost sorbent. Beyond the comparative benefit, it will be important for electrochemical carbon capture research to begin to address new non-energy and non-rate questions to avoid falling into the McNamara fallacy,⁸⁷ the mistaken belief that existing measurements were selected because they were

the only metrics of importance and, consequently, the metrics that have not yet been studied are unimportant. Now that electrochemical capture has been proven to work, there are many practical viability questions to answer, and results and experimental conditions must be reported transparently to make useful conclusions.

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AUTHOR CONTRIBUTIONS

J.B. was responsible for conceptualization, methodology, analysis and visualization, writing, and editing. C.A.G. was responsible for supervision, review, and editing. Y.L. was responsible for review and editing.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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