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Maximizing single-pass conversion does not result in practical readiness for CO_2 reduction electrolyzers

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For many chemical processes, high single-pass conversion of reactants into products reduces the need to separate products downstream. However, low-temperature carbon dioxide electrolyzers that maximize single-pass conversion suffer from low product concentration. Maximizing product concentration is therefore a more meaningful target for CO_2 electrolyzers than maximizing single-pass conversion.

To integrate electrochemical carbon dioxide reduction (CO_2R) into the chemicals industry at scale, the outlet streams from CO_2 electrolyzers must be product rich. Single-pass conversion is becoming increasingly common as a performance benchmark for CO_2 electrolyzers because it suggests concentrated products and reduced separation energy for many catalytic processes. However, our analysis shows that CO_2R reactor configurations that maximize single-pass conversion currently suffer from low product concentration in the outlet stream. This is because they restrict CO_2 flow or operate in acidic cathode environments, which promote considerable hydrogen evolution as a side reaction. For any gas products besides syngas, high single-pass conversion does not signify that separation energy losses have been eliminated, or that product streams are directly suitable as feedstocks for downstream processes. We therefore recommend that researchers targeting CO_2R scaleup report product concentrations rather than relying on single-pass conversion as an indicator of overall performance. To commercialize CO_2 electrolysis, maximizing product concentration is a more relevant goal than maximizing single-pass conversion.

Outlet streams from CO_2 electrolyzers need gas separations

Electrochemical CO_2 reduction is a potential low-carbon pathway for producing chemical feedstocks and fuels from renewable electricity, water, and CO_2[1,2]. CO_2 electrolyzers can produce value-added chemicals whose industrial demand is in the hundreds of megatons annually[3,4], including carbon monoxide[5], formic acid[6], and ethylene[7]. CO can in turn be electrochemically reduced in a two-stage cascade[8,9].

State-of-the-art CO_2 electrolyzers have limited reactant utilization because of the incomplete reaction of CO_2, as well as CO_2 crossover due to carbonate acid-base equilibrium. Selectivity is also limited by competition from water reduction via the hydrogen evolution reaction (HER), in which CO_2 is uninvolved. Gas separations and recycle streams are needed to recover and convert unreacted CO_2, and to purify products.

Typical CO_2 electrolyzers produce two gas-phase outlet streams, one each at the cathode and anode, which can both contain residual CO_2 (Fig. 1a). The cathode gas contains gas-phase CO_2R products, unreacted CO_2, and hydrogen as a byproduct from the HER. CO_2 electrolyzers can produce carbon monoxide with 100% molar selectivity[10,11], so a hydrogen-free tail gas can be produced under optimal conditions. For multicarbon products, state-of-the-art electrolyzers have a molar selectivity of 50% to C_{2+} products, with the co-evolution of 20% H_2 and 30% C_1 products at the cathode[12,13]. The scope of this discussion is limited to gas-phase separations, which are necessary for the production of CO or of ethylene, the dominant C_{2+} product. For liquid products like formic acid, ethanol, and acetate, both gas and liquid product streams are formed, which requires an independent analysis of separations.

At the anode, oxygen is produced from water oxidation via the oxygen evolution reaction (OER). The anode outlet also contains CO_2 that crosses over the membrane via the homogeneous reactions shown in Fig. 1a[14,15]. This crossover arises from acid-base equilibria—hydroxide generated by the cathodic reaction converts dissolved CO_2 into (bi)carbonate ions in neutral electrolytes, like $KHCO_3$ or Cs_2CO_3, or alkaline electrolytes such as KOH. (Bi)carbonate anions migrate towards the anode, where they buffer protons generated by OER, regenerating CO_2 gas. This buffering reaction limits single-pass conversion at most pH[16]. In neutral electrolytes, carbonate ion crossover stoichiometrically consumes 0.5 mol CO_2/mol e^-, which results in an anode tail gas consisting of 67 mol% CO_2 + 33 mol% O_2. Therefore, both the cathode and anode outlet gases can contain unreacted CO_2 that must be captured and recycled to the cathode (Fig. 1b, c).

Recycling unreacted CO_2 requires a gas separation that could be a capital- and energy-intensive process. An electrolyzer with incomplete conversion, imperfect selectivity, and reactant crossover needs at least three pairwise separations: one to separate the target product from byproducts, and two to recycle CO_2 from the cathode and anode outlets (Fig. 1d). Separation units are typically modeled as pressure-swing adsorption using electrical utilities[3], but cryogenic distillation or amine scrubbing could be preferable depending on process scale, stream compositions, and available utilities[17]. If extensive downstream separations are required to purify products and recycle CO_2, the overall energy efficiency of CO_2R has been argued to suffer unacceptably[15]. To account for separation demands, studies on CO_2 electrolyzers increasingly report single-pass conversion (SPC):

$$SPC = \frac{CO_2 \text{ moles converted to products}}{CO_2 \text{ moles fed to reactor}}$$

In other catalytic processes, high SPC generally suggests improved reactant utilization and hence lower energy to recycle the unreacted
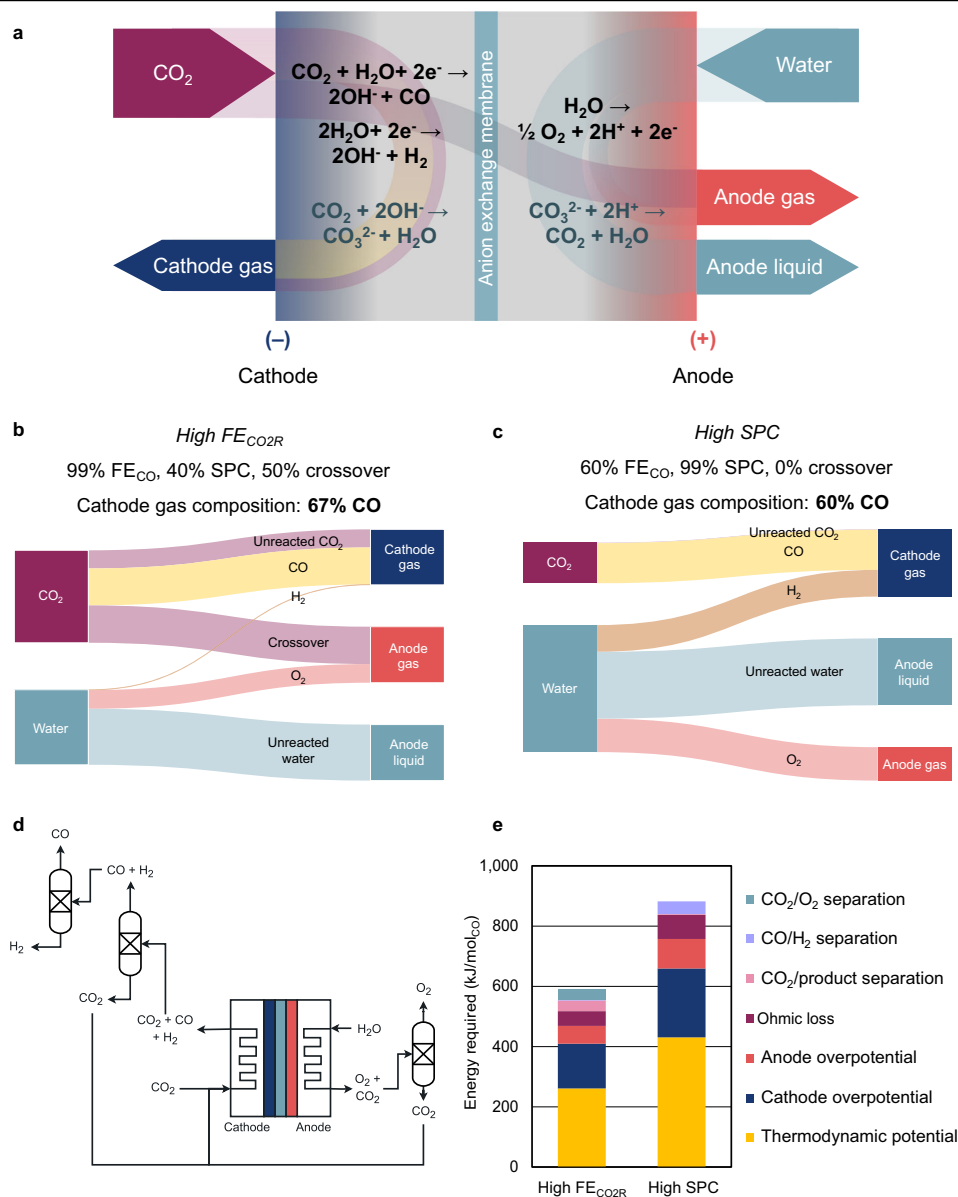
Comment

Fig. 1 | Typical CO₂ electrolyzer outlet gases. a Reaction pathways for a typical CO_2 electrolyzer reducing CO_2 to CO in a neutral electrolyte with an anion exchange membrane. Black text indicates heterogeneous redox reactions, while blue indicates homogeneous reactions. **b**, **c** Molar flow rates in a single pass of CO_2 reduction to CO for example scenarios with (**b**) high Faradaic efficiency towards CO₂R with realistic parameters and (**c**) high single-pass conversion with optimistic parameters. The total current is the same in both cases. Despite optimistically high single-pass conversion in (**c**), CO concentration in the cathode outlet is decreased. **d** Sample gas separation scheme required for a CO_2 electrolyzer. **e** Energy required per mole of product for reaction (modeled as overpotential) versus separation (modeled thermodynamically) shows that the high-SPC scenario wastes reactor energy on hydrogen evolution. Process parameters correspond to (**b**) and (**c**).

feed. Also, downstream applications, like ethylene polymerization, typically require high-purity feedstocks. Since high SPC implies that the products are not diluted in leftover reactant, it is associated with marketable product streams.

High SPC does not eliminate cathode gas separations

Molar stream composition is a critical consideration for downstream applications. For instance, thermal reactors are sensitive to reactant partial pressures, so CO₂R products must often be concentrated for

downstream processing. To assess the relationship between SPC and reactor outlet composition, we analyzed data from literature reports targeting state-of-the-art CO_2 electrolyzer performance (for details, see Supplementary Information, Section 3). These relationships are demonstrated in Fig. 2, with additional representations in Fig. S2 and pH dependence in Fig. S3.

As shown in Fig. 2a, we find that experimental conditions that maximize single-pass conversion do not correspond to concentrated product streams suitable for further reaction. To maximize SPC,
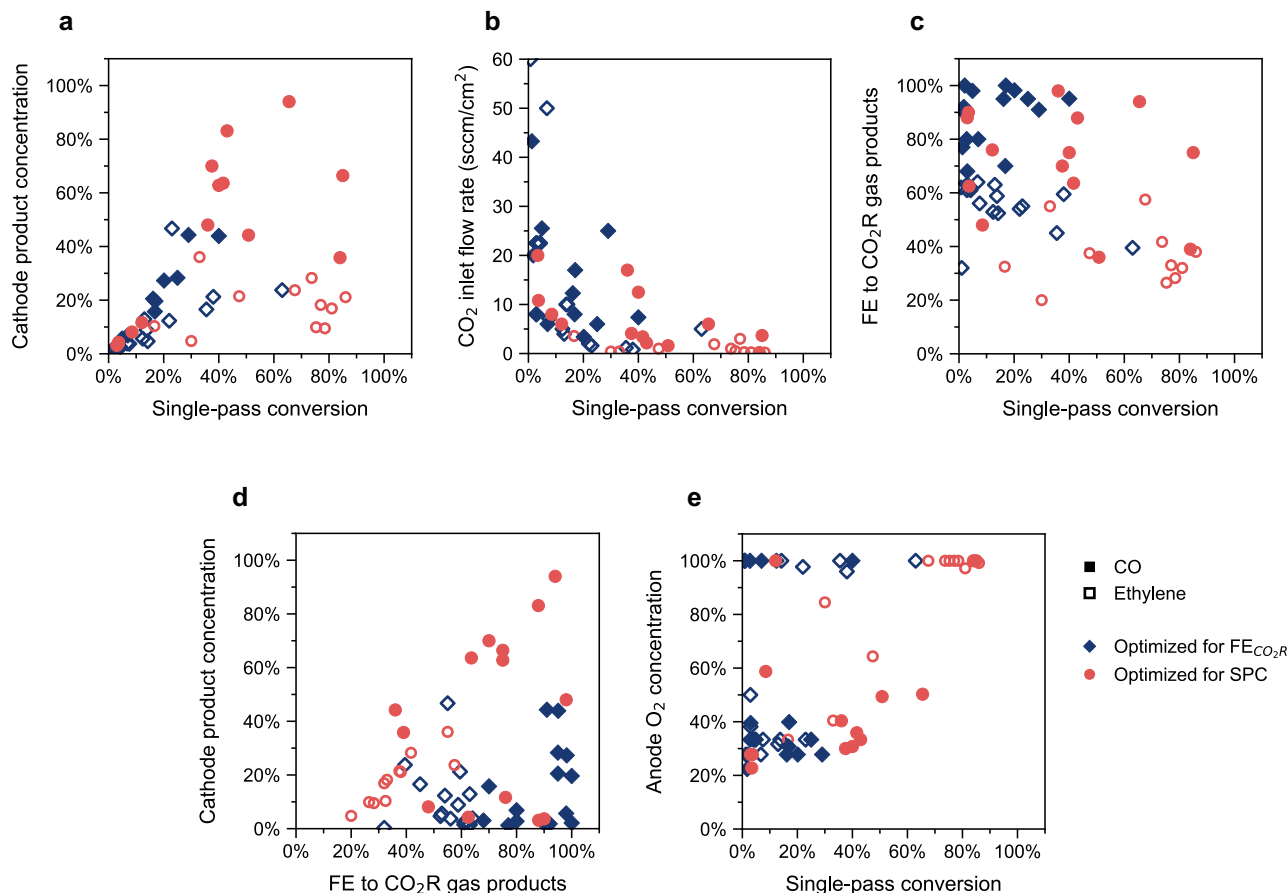
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Fig. 2 | Correlations between single-pass conversion and CO₂R performance in literature. a High single-pass conversion does not result in highly concentrated products that are suitable for downstream applications. **b** High single-pass conversion has been achieved at low feed flow rates, at which hydrogen evolution is the dominant reaction. **c** Faradaic efficiency to CO₂R trades off with single-pass conversion across various reaction conditions. **d** Concentrated cathode streams are produced by maximizing Faradaic efficiency, which is extremely challenging at high single-pass conversions. **e** Anode gas separations can be minimized across a range of single-pass conversion. Hollow symbols (□) correspond to ethylene production and filled symbols (■) represent CO generation. Red circles (●) correspond to the highest single-pass conversion in a report, and blue diamonds (◆) correspond to the highest FE_{CO_2R} in a report; their operating conditions are usually different since selectivity and SPC trade off in current electrolyzer configurations[10,16,18–20,22,27,29–49].

studies are often conducted at low inlet CO₂ flow rates (Fig. 2b). Under these conditions, the partial pressure of CO₂ drops steeply across the reactor as it is consumed by reaction, resulting in a loss in CO₂R selectivity in favor of HER[18–20]. In some cases, SPC has also been increased using an acidic electrolyte (e.g. $H_2SO_4 + K_2SO_4$), or reverse-biased bipolar membrane to eliminate carbonate formation and crossover. Unfortunately, the high availability of protons in such configurations steers selectivity towards HER over CO₂R, which is not captured in SPC since CO₂ plays no direct role in the HER reaction. This tradeoff between Faradaic efficiency (FE_{CO_2R}) and SPC has been demonstrated for various electrolyzers previously[21–23], and is reflected across the dataset we analyzed (Fig. 2c). Hence strategies that reduce the need to recycle CO₂ still require separations to remove H₂ and concentrate the product for downstream processes. Additionally, electrolyzer energy is wasted on making H₂ in these scenarios. A comparison of the energy required for reaction and separation (Fig. 1e, Fig. S1) and a recent in-depth analysis[24] suggest that reactor energy requirements significantly exceed the energetic costs of separations.

Hence the energy demand for CO₂R is dramatically increased at high SPC/ low FE_{CO_2R} conditions, even if gas separations are reduced (Fig. 1e). Therefore, high SPC neither eliminates the need for cathode gas separations, nor indicates that products are formed at an improved energy efficiency.

Neither SPC nor FE_{CO_2R} directly reflect outlet composition, as indicated by the lack of a clear trend in Fig. 2a, d. While SPC fails to account for HER, FE_{CO_2R} and partial current density do not reflect molar flow rates. For C₂₊ products, this problem is especially acute since multiple electron pairs are transferred for CO₂R, compared to a single pair to make H₂. For example, a 90% FE to CO with 10% to H₂ at 100% SPC translates to a stream composition of 90 mol% CO + 10 mol% H₂. In contrast, 90% FE to ethylene with 10% to H₂ results in 60 mol% ethylene + 40 mol% H₂ because of the 12 e⁻ transferred to produce ethylene.

Although high FE_{CO_2R} does not perfectly scale to high molar flows, Fig. 2d shows that the most concentrated cathode product is obtained at high FE_{CO_2R}, which is extremely challenging to achieve at the low

Comment

flow rates that give high SPC. At high current densities and FE_{CO2R}, high crossover and selective reaction manifest in very concentrated cathode products. On the other extreme, crossover can be minimized to produce 100% O_2 at the anode. However, a pure O_2 anode stream has been reported across a range of SPC (Fig. 2e), so high SPC is not a necessary condition for low crossover. If minimizing HER is important for CO_2R scaleup, then SPC to CO_2R products is a misleading metric that does not truly reflect that a reactor design is practical.

Electrolyzers for scaleup should report outlet compositions

To pursue industrial relevance for selective CO_2R, we recommend a careful selection of metrics and operating conditions for measuring and reporting electrolyzer performance. Our analysis shows that SPC does not accurately reflect downstream compositions at the cathode. A realistic representation of separation demand can be provided by mole fractions and outlet flow rates. The concentration of products in the cathode and anode streams indicates the readiness of electrolyzers for downstream applications, informing separation energy and process feasibility. Therefore, we recommend that researchers report cathode and anode product stream compositions. Section 3 of the Supplementary Information outlines the conversions between more common metrics and stream compositions, which can also be used on existing datasets to analyze the effects of experimental variables on product streams.

We suggest that researchers characterize the composition of the anode gas, as has been recommended by Seger and coworkers[25,26]. At present, very few reports on state-of-the-art CO_2 electrolyzers explicitly quantify the anode gas stream. The combination of complete anode and cathode gas quantification allows the carbon mass balance to be used as an additional experimental validation step. The carbon balance is especially important in alkaline electrolytes that absorb CO_2, where anode gas quantification can be nuanced. CO_2R reports in alkaline conditions rarely assess the extent of carbonate formation[27] or CO_2 regeneration at the anode, and often report a lower crossover than is physically reasonable. This may be due to the anolyte not being purged of CO_2, thus excluding carbonate formation from the carbon balance.

We recommend that researchers supply CO_2 flow rates that correspond to at least the amount of CO_2 consumed stoichiometrically by CO_2R at the applied current. Many reports feed a lower CO_2 molar flow than the chronopotentiometry current. This artificially inflates SPC at the cost of selectivity, since the CO_2R partial current density is severely reactant limited and HER must compensate for the remaining current. The complete reaction of 1 sccm of CO_2 accounts for 143 mA of current through a 2-e^- pathway making C_1 products, or 430 mA of current through a 12-e^- C_2 pathway. Therefore, on a 5 cm^2 cathode where 1 sccm CO_2 is fed, it is impossible to produce more than 29 mA/cm^2 of C_1 products, even in the absence of crossover. In a neutral electrolyte where carbonate is the dominant charge carrier, CO_2R current is further constrained by the crossover of 0.5 mol CO_2/mol e^-. Under these conditions, the partial current density to 12-e^- C_{2+} products from 1 sccm CO_2 on a 5 cm^2 electrode is at most 22 mA/cm^2. Most experiments in membrane electrode assemblies (MEAs) are operated at >50 mA/cm^2, so low CO_2 flow rates guarantee high HER. To operate at a current density of 100 mA/cm^2 of CO_2R to CO on a 5 cm^2 electrode in neutral conditions, at least 7 sccm of CO_2 must be fed to the reactor.

From a scaleup perspective, the optimal flowrate depends on complex tradeoffs, including between electrolyzer energy and

separator capital cost, or selectivity versus crossover[24]. CO_2R has been shown to be severely limited by CO_2 availability in a variety of system configurations[10,20,28]. Continuum modeling of current electrolyzer designs suggests that concentration gradients on the cathode surface effectively make it impossible to co-optimize conversion and selectivity at low flow rates[28]. In contrast, large CO_2 feeds steer selectivity towards CO_2R but increase separation and compression energies by diluting products and requiring recycle streams. Several reports[19,20,22] show a parabolic trend in product concentration versus feed flow rate. Further techno-economic analysis is needed to determine whether the process energy and cost are optimal at the peak of this parabola. We also note that industrial CO_2 feeds differ from most reported experiments. Although our main conclusions are likely transferable, the scale of recycle streams and separation units for a dilute CO_2 feed could change the optimum between separations and reactions.

Lastly, to clarify reports of high SPC, we recommend reporting FE_{CO2R} and SPC at the same conditions, rather than the best-case scenarios for each. It has been repeatedly shown that selectivity and SPC trade off in current MEA designs[23,29]. A singlestar plot should not include the best performance of an electrolyzer under multiple operating conditions. Given the (bi)carbonate equilibrium, we also note that conversions should always be reported as the ratio of CO_2 in CO_2R products/CO_2 fed, and never CO_2 consumed/CO_2 fed.

In summary, we analyzed stream compositions for state-of-the-art CO_2 electrolyzers to show that single-pass conversion alone does not capture the extent of downstream gas separations required. Although electrolyzers with high SPC do not dilute products in unreacted CO_2, they still produce mixed cathode product streams. In current electrolyzer designs, this tradeoff arises from physical limitations. Low CO_2 feed flow rates and proton-rich environments maximize SPC but increase the side reaction of hydrogen evolution. We recommend that researchers prioritize and report outlet gas compositions, since maximizing product yield and mole fraction in the cathode outlet is more practically important than maximizing single-pass conversion. While reactor designs that decouple the tradeoff between selectivity and SPC could be pursued, electrolyzer energy dominates separation for both low and high SPC operation, so this is not the most pressing challenge facing CO_2R scaleup. Thermocatalytic processes and solid oxide electrolytic cells often operate at low SPC with separation and recycle schemes, optimizing systems for the reaction rather than separation. CO_2 electrolyzers can similarly benefit from prioritizing other goals, including high product yields and low cell voltages, over increased single-pass conversion.

Data availability

The source data from this study can be found in the Source Data file (Excel workbook). The workbook includes: literature data, assumptions and calculated metrics used to generate Fig. 2; additional figures comparing metrics; assumptions and analysis for the energy breakdown in Fig. 1e; calculations for limiting CO_2 flow rates and current densities recommended in the main text. Figure 1a, b are discussed in the Supplementary Information, Section 1, and their source data is available from the authors upon reasonable request. The Supplementary Information details the assumptions made, including in Section 5 and Tables S1, S2. Source data are provided with this paper.

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Comment

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Author contributions
S.C.D. developed the concept, analyzed experimental data, and wrote the article. J.R. guided the work. All authors contributed to the discussion, review, and editing of the manuscript.

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