

1 **Maximizing single-pass conversion does not result in practical readiness for**
2 **CO₂ reduction electrolyzers**

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6
7 **Abstract**

8 Although single-pass conversion is a metric that suggests a lower need to separate products in
9 many chemical processes, low-temperature carbon dioxide electrolyzers that maximize single-
10 pass conversion suffer from low product concentration. Maximizing product concentration is
11 therefore a more meaningful target for CO₂ electrolyzers than maximizing single-pass
12 conversion.

13 To integrate electrochemical CO₂ reduction into the chemicals industry at scale, the outlet
14 streams from CO₂R electrolyzers must be product rich. Single-pass conversion is becoming
15 increasingly common as a performance benchmark for CO₂R electrolyzers because it indicates
16 concentrated products and reduced separation energy for many catalytic processes. However, our
17 analysis shows that CO₂R reactor configurations that maximize single-pass conversion currently
18 suffer from low product concentration in the outlet stream. This is because they restrict CO₂ flow
19 or operate in acidic cathode environments, which promote considerable hydrogen evolution as a
20 side reaction. For any gas products besides syngas, high single-pass conversion does not signify
21 that separation energy losses have been eliminated, or that product streams are suitable
22 feedstocks for downstream processes. We therefore recommend that researchers targeting CO₂R
23 scaleup report product concentrations rather than relying on single-pass conversion as an
24 indicator of overall performance. Maximizing product concentration is a more meaningful target
25 than maximizing single-pass conversion.

26

27 **CO₂R electrolyzers produce impure streams that require gas separations**

28 Electrochemical carbon dioxide reduction (CO₂R) has the potential to be a low-carbon
29 pathway for producing chemical feedstocks and fuels from renewable electricity, water, and
30 CO₂.^{1,2} State-of-the-art CO₂R electrolyzers have limited reactant utilization because of the
31 incomplete reaction of CO₂, as well as CO₂ crossover due to carbonate acid-base equilibrium.
32 Selectivity is also limited by competition from water reduction via the hydrogen evolution
33 reaction (HER), in which CO₂ is uninvolved. Gas separations and recycle streams are needed to
34 recover and convert unreacted CO₂, and to purify products.

35 Typical CO₂R electrolyzers produce two gas-phase outlet streams, one each at the cathode
36 and anode, which can both contain residual CO₂ (Figure 1A). The cathode gas contains gas-
37 phase CO₂R products, unreacted CO₂, and hydrogen as a byproduct from the HER. CO₂R can
38 produce carbon monoxide with 100% molar selectivity,^{3,4} so a hydrogen-free tail gas can be
39 produced under optimal conditions. For multicarbon products, state-of-the-art electrolyzers have
40 a molar selectivity of 50% to C₂₊ products, with the co-evolution of 20% H₂ and 30% C₁
41 products at the cathode.^{5,6} The scope of this discussion is limited to gas-phase separations, which
42 are necessary for the production of CO or ethylene, the dominant C₂₊ product.

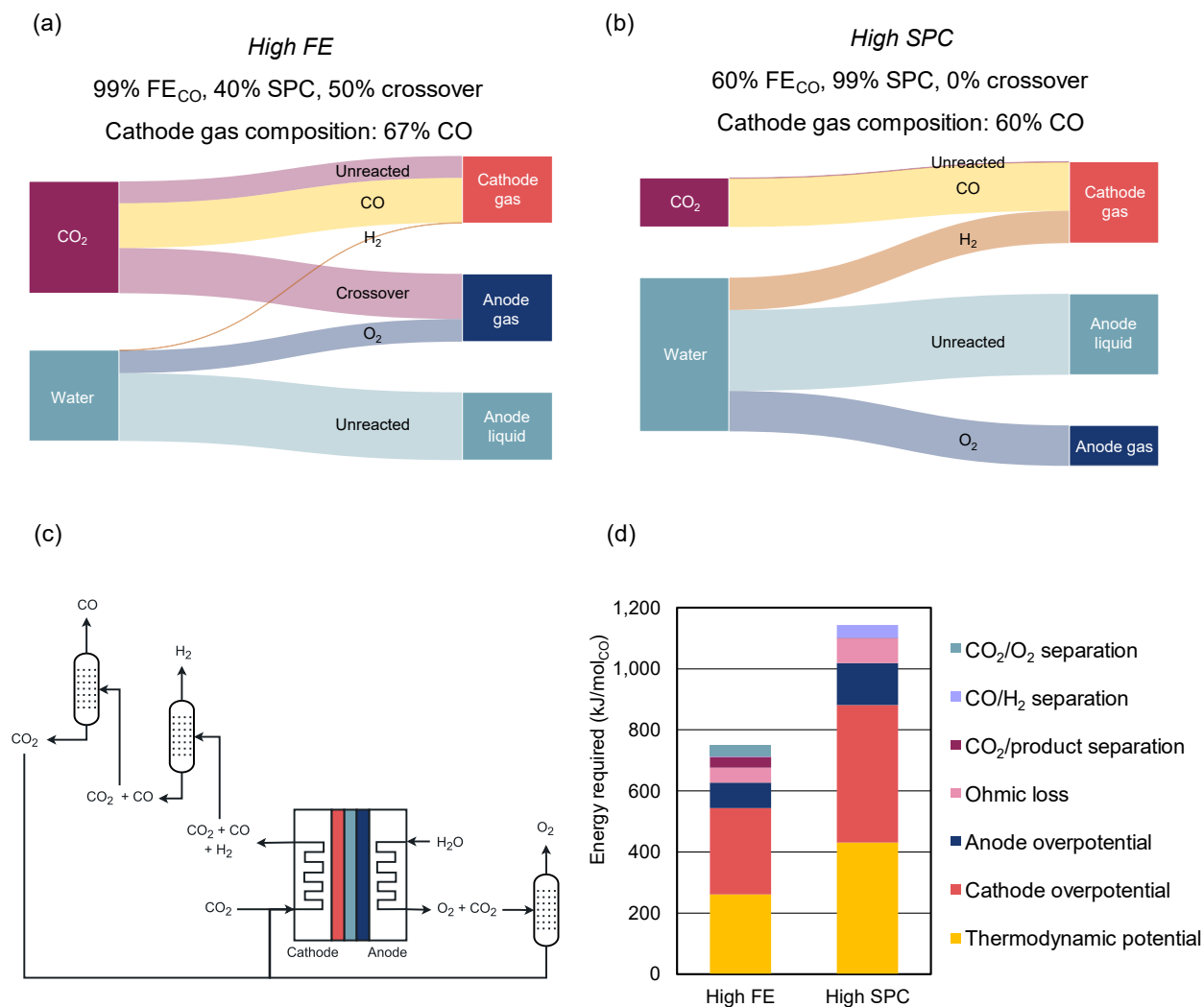
43 At the anode, oxygen is produced from water oxidation via the oxygen evolution reaction
44 (OER). The anode outlet also contains CO₂ that crosses over the membrane.^{7,8} This crossover
45 arises from homogeneous acid-base equilibria – in neutral or alkaline electrolytes, hydroxide
46 generated by the cathodic reaction converts dissolved CO₂ into carbonate ions. Carbonate anions
47 migrate towards the anode, where they buffer protons generated by OER, regenerating CO₂ gas.
48 This buffering reaction limits single-pass conversion at most pH.⁹ In neutral electrolytes,
49 carbonate ion crossover stoichiometrically consumes 0.5 mol CO₂/ mol e⁻. Carbonate crossover
50 results in an anode tail gas consisting of 67 mol% CO₂ + 33 mol% O₂. Therefore, both the
51 cathode and anode outlet gases can contain unreacted CO₂ that must be captured and recycled to
52 the cathode (Figures 1A, B).

53 Recycling unreacted CO₂ requires a gas separation that could be a capital- and energy-
54 intensive process. An electrolyzer with incomplete conversion, imperfect selectivity, and reactant
55 crossover needs at least three pairwise separations: one to separate the target product from
56 byproducts, and two to recycle CO₂ from the cathode and anode outlets (Figure 1C). Separation
57 units are typically modeled as pressure-swing adsorption using electrical utilities,¹⁰ but cryogenic

58 distillation or amine scrubbing could be preferable depending on process scale and available
59 utilities.¹¹ If extensive downstream separations are required to purify products and recycle CO₂,
60 the overall energy efficiency of CO₂R has been argued to suffer unacceptably.⁸ To account for
61 separation demands, studies on CO₂R electrolyzers increasingly report single-pass conversion:

62
$$SPC = \frac{CO_2 \text{ moles converted to products}}{CO_2 \text{ moles fed to reactor}}$$

63 In other catalytic processes, high SPC generally suggests improved reactant utilization and hence
64 lower energy to recycle the unreacted feed. Also, downstream applications, like ethylene
65 polymerization, typically require high-purity feedstocks. Since high SPC implies that the
66 products are not diluted in leftover reactant, it is associated with marketable product streams.



67
 68 **Figure 1. Typical CO_2R electrolyzer outlet gases.** (A, B) Molar flow rates in a single pass of CO_2R to
 69 CO for example scenarios with (A) high FE_{CO_2R} with realistic parameters and (B) high single-pass
 70 conversion with optimistic parameters. The total current is the same in both cases. Despite optimistically
 71 high single-pass conversion in (B), CO concentration in the cathode outlet is decreased. (C) Sample gas
 72 separation scheme required for a CO_2R electrolyzer. (D) Energy required per mole of product for reaction
 73 (modeled as overpotential) vs separation (modeled as thermodynamics) show that the high-SPC scenario
 74 wastes reactor energy on hydrogen evolution. Process parameters correspond to (A) and (B).

75

76 **High single-pass conversion does not eliminate cathode gas separations**

77 Molar stream composition is a critical consideration for downstream applications. For
78 instance, thermal reactors are sensitive to reactant partial pressures, so CO₂R products must often
79 be concentrated for downstream processing. To assess the relationship between SPC and reactor
80 outlet composition, we analyzed data from literature reports targeting state-of-the-art CO₂R
81 electrolyzer performance (for details, see SI).

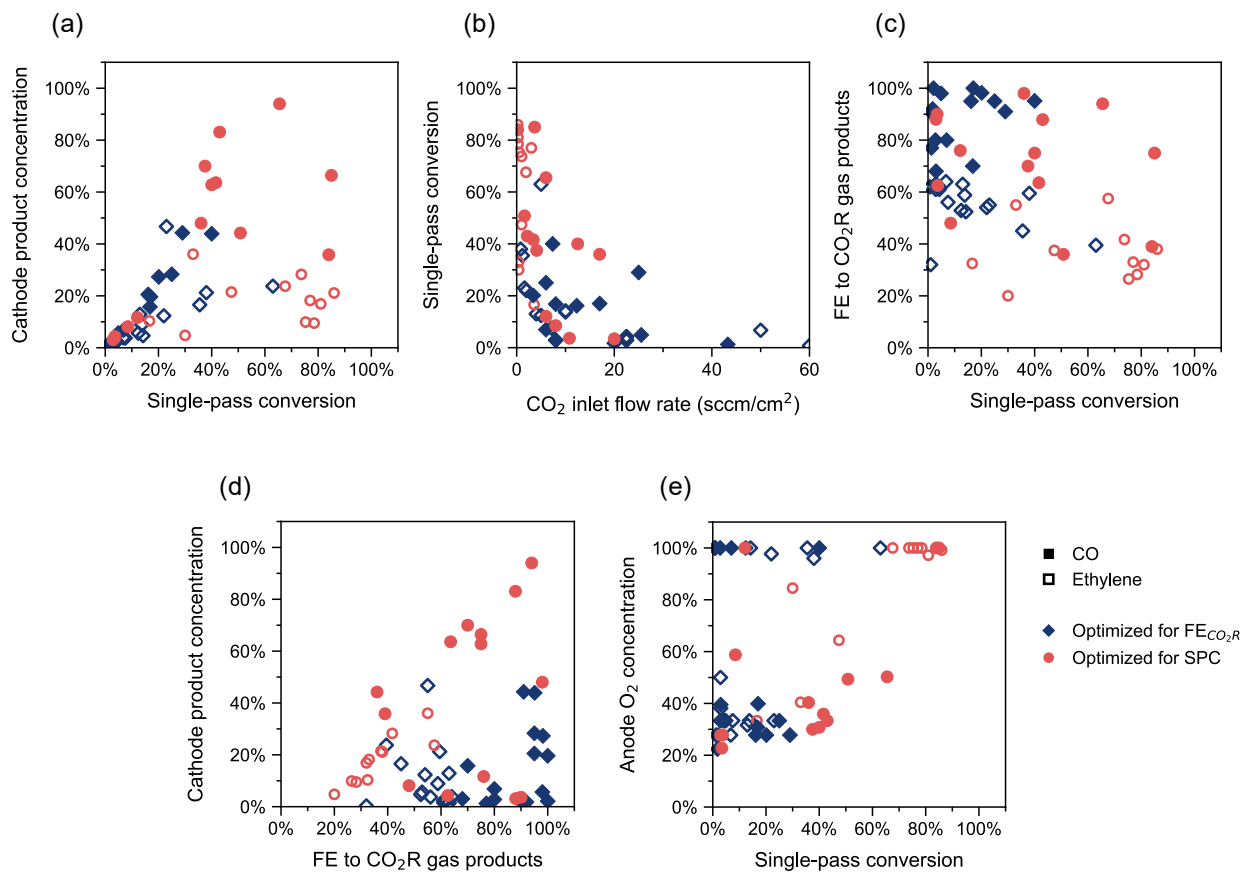
82 As shown in Figure 2A, we find that experimental conditions that maximize single-pass
83 conversion do not correspond to concentrated product streams suitable for further reaction. To
84 maximize SPC, studies are often conducted at low inlet CO₂ flow rates (Figure 2B). Under these
85 conditions, the partial pressure of CO₂ drops steeply across the reactor as it is consumed by
86 reaction, resulting in a loss in CO₂R selectivity in favor of HER.^{12–14} In some cases, SPC has also
87 been increased using an acidic electrolyte or reverse-biased bipolar membrane to eliminate
88 carbonate formation and crossover. Unfortunately, the high availability of protons in such
89 configurations steers selectivity towards HER over CO₂R, which is not captured in SPC since
90 CO₂ plays no direct role in the HER reaction. This tradeoff between FE_{CO₂R} and SPC has been
91 demonstrated for various electrolyzers previously,^{15–17} and is reflected across the dataset we
92 analyzed (Figure 2C). Hence strategies to reduce the need to recycle CO₂ still require separations
93 to remove H₂ and concentrate the product for downstream processes. Additionally, electrolyzer
94 energy is wasted on making H₂ in these scenarios. A comparison of the energy required for
95 reaction and separation (Figure 1D) and a recent in-depth analysis¹⁸ suggest that reactor energy
96 requirements significantly exceed the energetic costs of separations. Hence the energy demand
97 for CO₂R is dramatically increased at high SPC/ low FE_{CO₂R} conditions, even if gas separations

98 are reduced (Figure 1D). Therefore, high SPC neither eliminates the need for cathode gas
99 separations, nor indicates that products are formed at an improved energy efficiency.

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

107 Even though high FE does not perfectly scale to high molar flows, Figure 2D shows that
108 the most concentrated cathode product is obtained at high FE_{CO₂R}, which is extremely
109 challenging at low flow rates that give high SPC. At high current densities and CO₂R FEs, high
110 crossover and selective reaction manifest in very concentrated cathode products. On the other
111 extreme, crossover can be minimized to produce 100% O₂ at the anode. However, crossover can
112 be minimized across a range of SPC (Figure 2E), so high SPC is not a necessary condition for
113 low crossover. If minimizing HER is important for CO₂R scaleup, then SPC to CO₂R products is
114 a misleading metric that does not truly reflect that a reactor design is suitable for scaleup.

115



116

117 **Figure 2. Correlations between single-pass conversion and CO₂R performance in literature.**

118 (A) High single-pass conversion does not result in highly concentrated products that are suitable for
 119 downstream applications. (B) High SPC is achieved at low feed flow rates, at which hydrogen evolution
 120 is the dominant reaction. (C) Faradaic efficiency to CO₂ reduction trades off with single-pass conversion
 121 across various reaction conditions. (D) Concentrated cathode streams are produced by maximizing
 122 Faradaic efficiency, which is extremely challenging at high single-pass conversions. (E) Anode gas
 123 separations can be minimized across a range of SPC.

124 Red circles correspond to the single-pass conversion reported, and blue diamonds correspond to the
 125 highest FE_{CO₂R} reported; hollow symbols correspond to ethylene production and filled symbols represent
 126 CO generation.^{3,9,12–14,16,19–40}

127 **CO₂R electrolyzers targeting scaleup should report outlet compositions**

128 To pursue industrial relevance, we recommend a careful selection of metrics and
129 operating conditions for measuring and reporting the performance of electrolyzers that aim for
130 selective CO₂R. Our analysis shows that SPC does not accurately reflect downstream
131 compositions at the cathode. A realistic representation of separation demand can be provided by
132 mole fractions and outlet flow rates. The concentration of products in the cathode and anode
133 streams indicates the readiness of electrolyzers for downstream applications, informing
134 separation energy and process feasibility. Therefore, we recommend that researchers report
135 cathode and anode product stream compositions. We also suggest researchers characterize the
136 composition of the anode gas, as has been recommended by Seger and coworkers.^{41,42} The
137 combination of complete anode and cathode gas quantification allows the carbon mass balance to
138 be used as an additional experimental validation step. The carbon balance is especially important
139 in alkaline electrolytes that absorb CO₂, where anode gas quantification can be nuanced. Many
140 reports in alkaline conditions report a higher conversion than is physically reasonable. This may
141 be due to the anolyte not being purged of CO₂, thus excluding carbonate formation from the
142 carbon balance.

143 We recommend that researchers supply CO₂ flow rates that correspond to at least the
144 amount of CO₂ consumed stoichiometrically by CO₂R at the applied current. Many reports feed a
145 lower CO₂ molar flow than the chronopotentiometry current. This artificially inflates SPC at the
146 cost of selectivity, since the CO₂R partial current density is severely reactant limited and HER
147 must compensate for the remaining current. The complete reaction of 1 sccm of CO₂ accounts for
148 143 mA of current through a 2-e⁻ pathway making C₁ products, or 430 mA of current through a
149 12-e⁻ C₂ pathway. Therefore, on a 5 cm² cathode where 1 sccm CO₂ is fed, it is impossible to

150 produce more than 29 mA/cm² of C₁ products, even in the absence of crossover. In a neutral
151 electrolyte where carbonate is the dominant charge carrier, CO₂R current is further constrained
152 by the crossover of 0.5 mol CO₂/ mol e⁻. Under these conditions, the partial current density to 12-
153 e⁻ C₂₊ products from 1 sccm CO₂ on a 5 cm² electrode is at most 22 mA/cm². Most MEA
154 experiments are operated at >50 mA/cm², so low CO₂ flow rates guarantee high HER. To operate
155 at a current density of 100 mA/cm² of CO₂R to CO on a 5 cm² electrode in neutral conditions, at
156 least 7 sccm of CO₂ must be fed to the reactor.

157 From a scaleup perspective, the optimal flowrate depends on complex tradeoffs,
158 including between electrolyzer energy and separator capital cost, or selectivity versus
159 crossover.¹⁸ CO₂R has been shown to be severely limited by CO₂ availability in a variety of
160 system configurations^{3,14,43}. Continuum modeling of current electrolyzer designs suggests that
161 concentration gradients on the cathode surface effectively make it impossible to co-optimize
162 conversion and selectivity at low flow rates.⁴³ In contrast, large CO₂ feeds steer selectivity
163 towards CO₂R but increase separation and compression energies by diluting products and
164 requiring recycle streams. Several reports^{13,14,16} show a parabolic trend in product concentration
165 vs feed flow rate. Further techno-economic analysis is needed to determine whether the process
166 energy and cost are optimal at the peak of this parabola.

167 Lastly, to clarify reports of high SPC, we recommend reporting FE and SPC at the same
168 conditions, rather than the best-case scenarios for each. It has been repeatedly shown that
169 selectivity and SPC trade off in current MEA designs.^{17,28} A single star plot should not include
170 the best performance of an electrolyzer under multiple operating conditions. Given the carbonate
171 equilibrium, we also note that conversions should always be reported as CO₂ in CO₂R products/
172 CO₂ fed, and never CO₂ consumed/ CO₂ fed.

173 **Conclusions**

174 By analyzing stream compositions for state-of-the art CO₂R electrolyzers, we show that
175 single-pass conversion alone does not capture the extent of downstream gas separations required.
176 Although electrolyzers with high SPC do not dilute products in unreacted CO₂, they still produce
177 mixed cathode product streams. In current electrolyzer designs, this tradeoff arises from physical
178 limitations. Low CO₂ feed flow rates and proton-rich environments maximize SPC but increase
179 the side reaction of hydrogen evolution. We recommend that researchers prioritize and report
180 outlet gas compositions, since maximizing product yield and mole fraction in the cathode outlet
181 is more practically important than maximizing single-pass conversion. While reactor designs that
182 decouple the tradeoff between selectivity and SPC could be pursued, electrolyzer energy
183 dominates separation for both low and high SPC operation, so this is not the most pressing
184 challenge facing CO₂R scaleup. Thermocatalytic processes and solid oxide electrolytic cells
185 often operate at low SPC with separation and recycle schemes, optimizing systems for the
186 reaction rather than separation. CO₂R electrolyzers can similarly benefit from prioritizing other
187 goals, including high product yields and cell voltages, over increased single-pass conversion.

188

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193

194 **Data availability**

195 The data included in this study are available from the corresponding author upon reasonable
196 request.

197

198 **Author contributions**

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

201

202 **Competing interests**

203 There are no competing interests to declare.

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