

Conversion of Reactive Carbon Solutions into CO at Low Voltage and High Carbon Efficiency

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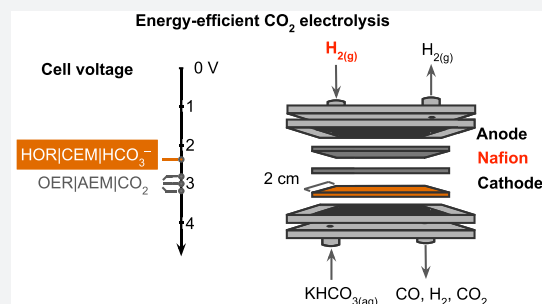
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ABSTRACT: Electrolyzers are now capable of reducing carbon dioxide (CO₂) into products at high reaction rates but are often characterized by low energy efficiencies and low CO₂ utilization efficiencies. We report here an electrolyzer that reduces 3.0 M KHCO₃(aq) into CO(g) at a high rate (partial current density for CO of 220 mA cm⁻²) and a CO₂ utilization efficiency of 40%, at a voltage of merely 2.3 V. These results were made possible by using: (i) a reactive carbon solution enriched in KHCO₃ as the feedstock instead of gaseous CO₂; (ii) a cation exchange membrane instead of an anion exchange membrane, which is common to the field; and (iii) the hydrogen oxidation reaction (HOR) at the anode instead of the oxygen evolution reaction (OER). The voltage reported here is the lowest reported for any CO₂ to CO electrolyzer that operates at high current densities (i.e., a partial current density for CO greater than 200 mA cm⁻²) with a CO₂ utilization efficiency of greater than 20%. This study highlights how the choice of feedstock, membrane, and anode chemistries affects the rate and efficiency at which CO₂ is converted into products.



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The CO₂ reduction reaction (CO₂RR) is a means of using electricity to convert CO₂ into fuels and chemicals.^{1–4} A commercial CO₂RR electrolyzer will likely need to operate at current densities (*j*) greater than 200 mA cm⁻² and cell voltages (*V*_{cell}) below 3 V.^{5–9} The only laboratory-scale CO₂RR electrolyzers that meet these criteria use anion exchange membranes (AEMs) to separate the cathode and anode compartments.^{9–12} The AEM creates an alkaline environment at the cathode and anode electrodes.¹³ The alkalinity at the anode reduces the applied potential required to drive the oxygen evolution reaction (OER) and enables the use of inexpensive nickel electrodes.¹⁴ However, the alkalinity at the cathode also converts the CO₂ reactant into (bi)carbonates (eq 1).^{15,16} This situation is problematic because (bi)carbonates are electrochemically inert and can migrate across the AEM to react with H⁺ in the anode chamber to regenerate CO₂.^{9,14} Consequently, the maximum possible CO₂ utilization efficiency (eq 2) for CO production is 50% for a CO₂RR electrolyzer that does not neutralize the OH⁻ byproduct (Figure 1); that is, one molecule of CO is produced for every two molecules of CO₂ entering the electrolyzer at steady state.¹⁷ In practice, CO₂ utilization efficiencies of <20% are generally observed for CO₂RR electrolyzers that contain an AEM to separate the OER and CO₂RR in the anode and cathode chambers, respectively (denoted “OER|AEM|CO₂” to indicate the anode|membrane|cathode configuration).¹⁸ Moreover, the O₂ produced at the anode of OER|AEM|CO₂ electrolyzers has little economic value and is therefore not utilized.¹⁴

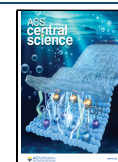


$$\begin{aligned} \text{CO}_2 \text{ utilization efficiency} \\ = \frac{\text{moles of CO produced}}{\text{moles of CO produced} + \text{moles of unreacted CO}_2} \\ \times 100\% \quad (2) \end{aligned}$$

CO₂RR electrolyzers that use bipolar membranes (BPMs) instead of AEMs convert a larger fraction of the CO₂ feedstock into CO (Figure 1).^{20–22} BPMs dissociate water at the interface of an AEM and a CEM to deliver OH⁻ and H⁺ to the anode and cathode, respectively (eq 3). The H⁺ supplied to the cathode reacts with bicarbonate to form CO₂ *in situ* (eq 4). The H⁺ also neutralizes the OH⁻ product formed during CO₂RR electrolysis (eq 5), which negates the alkalinity problem encountered with AEMs.²³ However, at high current densities a significant potential is required to dissociate water into H⁺ and OH⁻ using a BPM.²⁴ This potential can be reduced using a water dissociation catalyst.²⁵ However, CO₂RR electrolyzers that use BPMs currently require much

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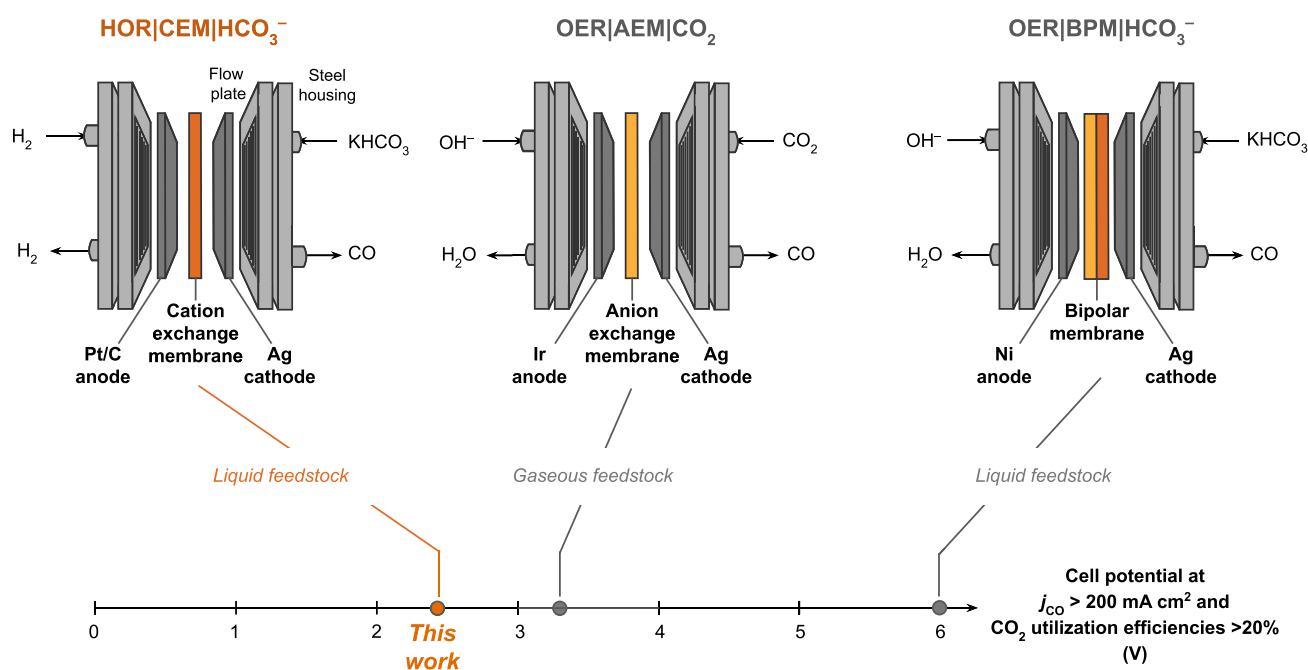
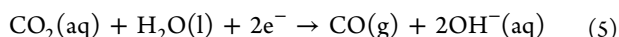
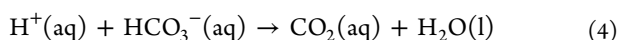
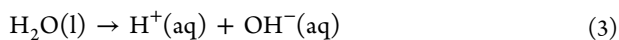


Figure 1. Schematics and nomenclature of prototypical CO₂ to CO electrolyzer configurations. The electrolyzer reported in this work is HOR|CEM|HCO₃[−], which uses a reactive carbon solution feedstock, a cation exchange membrane (CEM), and hydrogen oxidation at the anode to achieve low voltages and high CO₂ utilization efficiencies. The OER|AEM|CO₂ electrolyzer is a widely used architecture that uses a gaseous CO₂ feedstock. The OER|BPM|HCO₃[−] electrolyzer also uses a reactive carbon solution feedstock, but the BPM needs to be optimized to achieve lower applied cell voltages. The lowest cell voltages reported in the literature for each electrolyzer architecture are indicated, but only the electrolyzers that produce $j_{\text{CO}} > 200 \text{ mA cm}^{-2}$ and CO₂ utilization efficiency $> 20\%$ are considered.^{9,19} The nomenclature follows “anode|membrane|cathode”.

more than 3 V to realize $j_{\text{CO}} > 200 \text{ mA cm}^{-2}$ (Figure 1).¹⁴ It is for these reasons that neither a BPM or an AEM electrolyzer has simultaneously demonstrated high CO₂ utilizations and low voltages (Figure 1).^{20,26–29}



Another factor to consider is where the gaseous CO₂ feedstock is derived from. This feedstock requires the isolation, purification, and compression of CO₂ before it enters a CO₂RR electrolyzer.^{30,31} The isolation of CO₂ from air capture streams requires 23 MJ to convert 100 mol of K₂CO₃ into CO₂,³⁰ and CO₂ compression requires 2 MJ per 100 mol of CO₂.³² These steps are capital intensive, and the energy penalties are significant relative to electrolysis (12 MJ per 100 mol CO₂; Table S1). Moreover, the low CO₂ utilization efficiencies obtained with OER|AEM|CO₂ electrolyzers increase the cost of separating unreacted CO₂ from the product stream.³³

We report here a “bicarbonate electrolyzer” that solves the aforementioned problems by (i) reducing “reactive carbon solutions” (instead of gaseous CO₂) into CO at the cathode, (ii) replacing the AEM with a CEM, and (iii) replacing the OER with the HOR at the anode. Reactive carbon solutions are defined herein as the aqueous eluent from carbon capture units that use hydroxide to capture CO₂. These bicarbonate-enriched solutions enable captured carbon to be delivered to the electrolyzer as a liquid (e.g., KHCO₃(aq)) rather than as gaseous CO₂. This liquid feed not only simplifies the design of the electrolyzer, but also helps to increase CO₂ utilization

efficiency and bypasses the need to thermally generate pure CO₂ upstream of the electrolyzer.²⁶ The CEM also lowers Ohmic resistances relative to an AEM by transporting highly mobile H⁺ (ionic mobility $36 \times 10^{-8} \text{ m}^2 \text{ s}^{-1} \text{ V}^{-1}$) instead of OH[−] (ionic mobility $21 \times 10^{-8} \text{ m}^2 \text{ s}^{-1} \text{ V}^{-1}$; note that CO₃^{2−} is actually the dominant charge carrier in current AEM CO₂RR electrolyzers and is characterized by an even lower ionic mobility of $7.5 \times 10^{-8} \text{ m}^2 \text{ s}^{-1} \text{ V}^{-1}$).^{13,15,34} Moreover, CEMs avoid the high water dissociation overpotential associated with BPMs. The transport of H⁺ by the CEM into the cathode compartment enables the conversion of bicarbonate into electrochemically active CO₂ at the cathode (eq 4). The oxidation of hydrogen gas instead of water at the anode also serves to lower the applied potential required to drive electrolysis. These collective features of our electrolyzer (denoted “HOR|CEM|HCO₃[−]” to reflect the configuration) enabled a partial current density for CO of 220 mA cm^{−2} at a cell voltage of $2.3 \pm 0.1 \text{ V}$ and a CO₂ utilization efficiency of $40 \pm 2\%$. The lowest previously reported voltage for a CO₂RR electrolyzer that operates at a partial current density for CO $> 200 \text{ mA cm}^{-2}$ is 2.8 V and 16% CO₂ utilization.³⁵

RESULTS AND DISCUSSION

The HOR|CEM|HCO₃[−] electrolyzer reported here presses the anode and cathode tightly against opposite faces of Nafion, a CEM (Figure 1). Flowplates with serpentine channels were used to deliver humidified H₂ gas and 3 M KHCO₃ to the anode and cathode, respectively. The gas diffusion electrode (GDE) in the anode chamber consisted of platinum on carbon black, while a silver-foam electrode was used in the cathode chamber. This electrolyzer was used to perform electrolysis experiments at applied current densities over a 100–1000 mA cm^{−2} range. Product formation rates of CO and H₂ from the

cathode compartment along with V_{cell} values (the cell voltage measured across the anode and cathode) were recorded over the course of the electrolysis experiments. Measurements of the unreacted H_2 from the anode showed H_2 utilization values of up to 28% (Figure S2). The electrolysis experiments were performed under different pressures using a custom-made pressurized electrolyzer test station (Figure S3).¹⁹

Electrolysis experiments using this reactor architecture at an applied current density of 100 mA cm^{-2} and ambient conditions yielded a V_{cell} value of $1.7 \pm 0.1 \text{ V}$ (Figure 2).

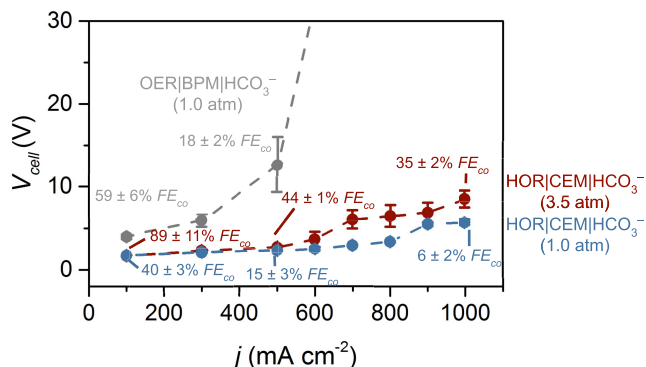


Figure 2. Voltage and current characteristics of an electrolyzer that couples bicarbonate conversion with hydrogen oxidation. V_{cell} values were measured as a function of current density from 100 to 1000 mA cm^{-2} for the OER|BPM|HCO₃⁻ and HOR|CEM|HCO₃⁻ electrolyzers under 1.0 and 3.5 atm of pressure. At 3.5 atm, 50 μm Nafion was used in the HOR|CEM|HCO₃⁻ electrolyzer instead of 25 μm Nafion. Faradaic efficiencies for CO production (FE_{CO}) for each electrolyzer are annotated at discrete points.

This value represents the lowest V_{cell} value ever reported for a liquid-fed CO₂RR electrolyzer. When the current density was held at 500 mA cm^{-2} , the V_{cell} value was measured to be $2.3 \pm 0.1 \text{ V}$, setting a new benchmark for CO₂ electrolysis.

We plotted V_{cell} values over a range of current densities against those obtained with our previously reported bicarbonate electrolyzer OER|BPM|HCO₃⁻ (Figure 2). The OER|BPM|HCO₃⁻ electrolyzer contains a BPM (instead of Nafion) and mediates the OER at a nickel anode (instead of the HOR at a platinum anode). In order to maintain a current density of 100 mA cm^{-2} , OER|BPM|HCO₃⁻ required a V_{cell} value of $4.0 \pm 0.3 \text{ V}$. This value is more than twice as high as that for HOR|CEM|HCO₃⁻, which performs the HOR at the anode. The V_{cell} value of OER|BPM|HCO₃⁻ spiked to $12.7 \pm 3.3 \text{ V}$ at 500 mA cm^{-2} , whereas the V_{cell} value of HOR|CEM|HCO₃⁻ was only $2.3 \pm 0.1 \text{ V}$ at this current density (Figure 2). The low measured voltage of HOR|CEM|HCO₃⁻ relative to OER|BPM|HCO₃⁻ is due not only to the lower thermodynamic voltage limit (achieved by substituting the OER for the HOR), but also the lower overpotentials of the HOR in comparison to the OER and water dissociation reaction (Figure S4).^{5,36} The thinner CEM also reduces Ohmic resistances relative to the BPM (the CEM is 25 or 50 μm , whereas the BPM is 195 μm).^{37,38} The voltage penalties for water dissociation and the OER render the V_{cell} value for the OER|BPM|HCO₃⁻ electrolyzer impractical.

While the V_{cell} values for HOR|CEM|HCO₃⁻ are state of the art, the measured Faradaic efficiencies for CO production (FE_{CO}) were merely $40 \pm 3\%$ at 100 mA cm^{-2} ($15 \pm 3\%$ at 500 mA cm^{-2} ; Figure 2). A supply of H^+ from the membrane

to the cathode is required for *in situ* CO₂ (*i*-CO₂) generation (eq 4); however, this H^+ flux can contribute to the parasitic hydrogen evolution reaction (HER). We therefore added a buffer layer between the cathode and membrane to mitigate the HER by managing H^+ transport to the cathode.³⁹ This buffer layer increased the FE_{CO} value from 40% to 71% at 100 mA cm^{-2} (Figure S5).

We increased the FE_{CO} values even further by pressurizing the bicarbonate feedstock, which increases the CO₂ solubility.¹⁹ The pressurized HOR|CEM|HCO₃⁻ electrolyzer under 3.5 atm of pressure yielded a FE_{CO} value of $89 \pm 11\%$ at 100 mA cm^{-2} and $44 \pm 1\%$ at 500 mA cm^{-2} (Figure 2). The molar composition of the gaseous cathode outlet stream at 500 mA cm^{-2} was 30% CO₂(g), 22% CO(g), and 48% H₂(g). The HOR|CEM|HCO₃⁻ electrolyzer therefore achieved a j_{CO} value of $\sim 220 \text{ mA cm}^{-2}$, the highest value reported for CO₂RR electrolyzers using a liquid feedstock (Figures S6 and S7). Importantly, the CO₂ utilization value was measured to be $40 \pm 2\%$, which is higher than that for any OER|AEM|CO₂ electrolyzer which achieves $j_{\text{CO}} > 200 \text{ mA cm}^{-2}$.¹⁸ The cell voltage was stable for 10 h. The slight decrease in FE_{CO} values could be caused by degradation of the platinum anode catalyst over time (Figure S8).⁴⁰

The HOR|CEM|HCO₃⁻ electrolyzer uses (i) a reactive carbon solution feedstock to bypass CO₂ desorption upstream of the electrolyzer and (ii) the HOR at the anode to eliminate the OER voltage penalty. However, the OER|AEM|CO₂ electrolyzer achieves a higher FE_{CO} in comparison to the HOR|CEM|HCO₃⁻ electrolyzer, and it does not require H₂ gas. We therefore performed a mass and energy balance to compare these two types of electrolyzer architectures. This analysis encompasses the energy required for the capture, regeneration, and electrolysis of 100 mol of CO₂. Importantly, the analysis accounts for the CO₂ utilization efficiency of each electrolyzer. The Sankey diagram in Figure 3 illustrates how a HOR|CEM|HCO₃⁻ electrolyzer with a CO₂ utilization efficiency of 40% yields CO with a higher energy efficiency ($1.4 \text{ MJ mol}^{-1} \text{ CO}$) in comparison to a OER|AEM|CO₂ electrolyzer ($1.9 \text{ MJ mol}^{-1} \text{ CO}$) operating at a CO₂ utilization efficiency of 20%.¹⁷ Another advantage of bicarbonate electrolysis is that it does not require the high temperatures and pressures used for thermochemical CO₂ hydrogenation.⁴¹

While the Sankey diagram shows that the HOR|CEM|HCO₃⁻ consumes less energy than the OER|AEM|CO₂ electrolyzer, it requires H₂ gas to be fed to the anode. This reaction reduces electricity consumption relative to the OER, but the source and cost of H₂ must be considered. Ideally, H₂ would be produced from a low-carbon source such as biomass gasification or water electrolysis.⁴⁴ The cost of generating H₂ by biomass gasification is reported to be as low as $\$0.9/\text{kg}$ ^{45,46} (with net negative emissions of 15–22 kg CO₂/kg H₂ when it is coupled to carbon capture and sequestration^{47,48}), and the target price for clean H₂ determined by the DOE Energy Earthshots Initiative is $\$1$ per kg of H₂.⁴⁹ We therefore used a forward-looking purchase price of $\$1/\text{kg}$ of H₂ as a basis for comparing the economics of HOR|CEM|HCO₃⁻, which consumes H₂, to the OER|BPM|HCO₃⁻ and conventional OER|AEM|CO₂, which consume water at the anode (Table 1 and Figure S9). We adopted widely used assumptions for future market conditions for this analysis (electricity $\$0.03/\text{kWh}$, $\$50/\text{tonne CO}_2$, CO sale price of $\$0.60/\text{kg}$)⁷ and electrolyzer performance specifications (voltages, Faradaic efficiencies, CO₂ utilization efficiencies) for each of the three

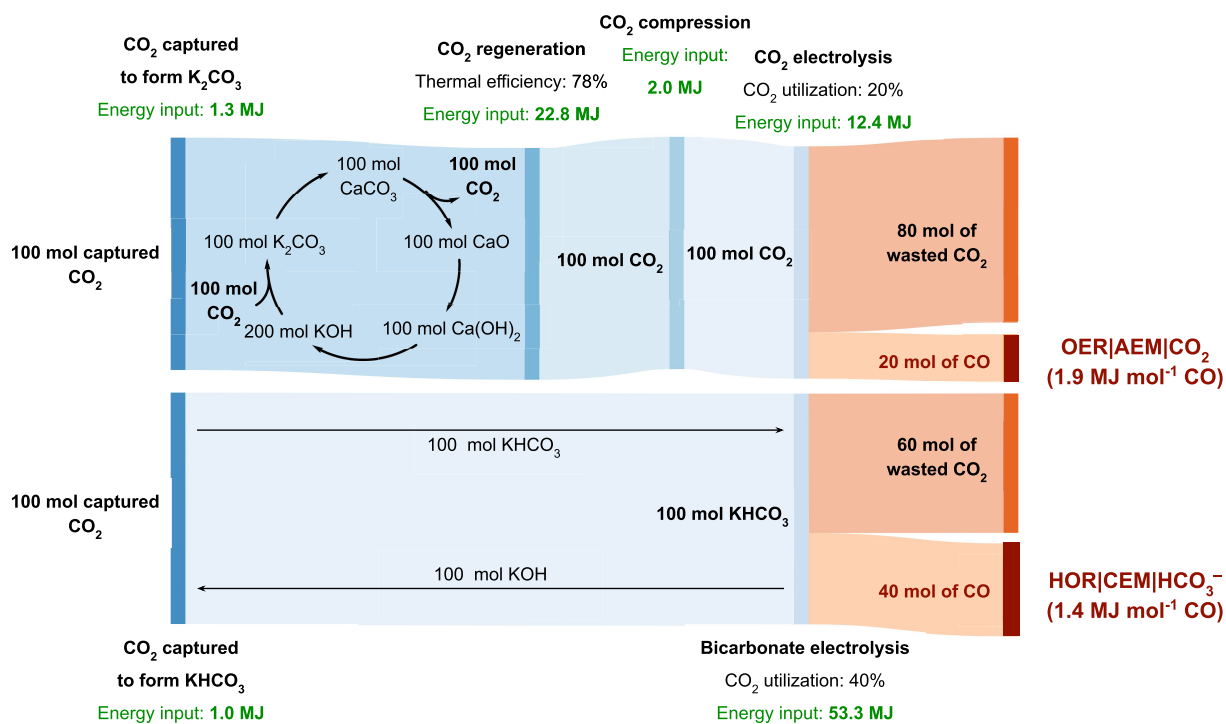


Figure 3. Sankey diagrams illustrating CO₂ mass flows and energy inputs for the capture and conversion of atmospheric CO₂ into CO using an anion exchange membrane (AEM) electrolyzer. The top panel assumes that captured CO₂ is regenerated using a direct air capture process^{30,42} and that the electrolyzer is fed with a compressed CO₂ feed (Faradaic efficiency for CO production, FE_{CO} = 90%; V_{cell} = 3.0 V; CO₂ utilization efficiency 20%; J = 500 mA cm⁻²). The bottom panel relies on the electrolysis of KHCO₃ and bypasses the CO₂ regeneration and compression steps (FE_{CO} = 50%; V_{cell} = 2.5 V; CO₂ utilization efficiency 40%; J = 500 mA cm⁻²). Energy inputs are sourced from refs 30 and 42. The bicarbonate electrolysis pathway analysis accounts for the energy required to generate H₂ from a water electrolyzer (347 kJ mol⁻¹ H₂).⁴³ Details are provided in Table S1.

electrolyzer technologies (Figure S9). We assumed FE_{CO} values of 100% for the OER|AEM|CO₂ and 80% for the HOR|CEM|HCO₃⁻ and OER|BPM|HCO₃⁻ electrolyzers, assumed a current density of 500 mA cm⁻², and expressed the voltages as slightly better than current state of the art metrics for the three electrolyzer technologies (Table 1). To determine the CO₂ capture and separation costs, we used CO₂ utilization efficiencies of 20% for the OER|AEM|CO₂ and 40% for the HOR|CEM|HCO₃⁻ and OER|BPM|HCO₃⁻ electrolyzers.

The outcome of this techno-economic analysis is that the HOR|CEM|HCO₃⁻ electrolyzer is the most profitable option of the three electrolyzers (see NPV values in Table 1). The primary drivers for the profitability of a bicarbonate electrolyzer coupled to a H₂ production plant are: (i) the lower electricity costs due to the lower operating voltage; (ii) the lower CO₂ capture and separation costs due to the elimination of CO₂ regeneration; and (iii) the higher CO₂ utilization efficiencies. We note that the recycling of H₂ can further minimize costs. For example, the proof of concept experiments in Figure S10 demonstrate that the amount of virgin H₂ supplied to the system can be reduced by recycling H₂ during the production of formate at the cathode. Formate was targeted for this experiment because CO poisons platinum catalysts used for the HOR and would therefore need to be separated before recycling.⁵⁰

We demonstrate here an electrolyzer that mediates the conversion of reactive carbon solutions enriched with bicarbonate into CO with a partial current density of 220 mA cm⁻² at merely 2.3 V. This high CO formation rate and

low voltage set a benchmark for reactive carbon capture. The CO₂ utilization value of 40% is also state of the art at high current densities (i.e., 500 mA cm⁻²). By sourcing H⁺ from the HOR instead of the OER, the HOR|CEM|HCO₃⁻ electrolyzer requires 2.3 V to drive bicarbonate electrolysis at 500 mA cm⁻² instead of 12.7 V for OER|BPM|HCO₃⁻. Moreover, we show that the FE_{CO} value of the electrolyzer can be increased to 89% at 100 mA cm⁻² and 44% at 500 mA cm⁻² by pressurizing the electrolyzer to increase CO₂ solubility. With these performance parameters, our techno-economic analysis (TEA) shows that coupling the HOR with bicarbonate electrolysis can generate CO more profitably than a traditional OER|AEM|CO₂ electrolyzer. These findings demonstrate a practical method for producing value-added carbon products from reactive carbon capture with low electrical energy input.

METHODS

KHCO₃ (99.5%, Alfa Aesar, USA), 50 wt % platinum on Vulcan XC 72 nanopowder (PK catalyst), and ethylenediaminetetraacetic acid (EDTA; 99%, Sigma-Aldrich, USA) were purchased and used as received. Carbon cloth gas diffusion layers (GDLs; Sigracet 39BB), Fumasep FBM bipolar membranes (BPMs), and Nafion PFSA NR-211 and 212 were purchased from Fuel Cell Store (USA). The BPMs were stored in 1 M NaCl, and the Nafion membranes were stored in 1 M KOH prior to use. Silver foams were obtained from Jiangsu Green Materials Hi-Tech. Co. Ltd. (China). Nafion 117 solutions (5 wt %; in a mixture of lower aliphatic alcohols and water) were obtained from Sigma-Aldrich, USA.

Table 1. Input and Output Parameters from the Technoeconomic Analysis of Three Electrolyzers

	HORICEM HCO ₃ ⁻	OER BPM HCO ₃ ⁻	OER AEM CO ₂
Input Parameters			
voltage (V)	2	4	3
CO ₂ utilization (%)	80	80	40
FE _{CO} (%)	80	80	100
<i>j</i> (mA cm ⁻²)		500	
gross cost of CO ₂ capture (\$/ton)		50	
installed capex cost (\$/kW)		450	
electricity (\$/kWh)		0.03	
H ₂ purchase price (\$/kg)	1	N/A	N/A
Output Parameters			
cost of H ₂ (\$M/yr)	2.61	0.00	0.00
electricity cost (\$M/yr)	5.24	10.48	6.29
CO ₂ capture cost (\$M/yr)	1.06	1.06	2.87
CO ₂ separation cost (\$M/yr)	0.72	0.72	4.30
maintenance and water cost (\$M/yr)	0.25	0.49	0.3
total opex (\$M/yr)	9.88	12.75	13.75
total capex: electrolyzer + balance of plant (\$M)	13.8	27.6	16.56
Revenue from Sale of CO at \$0.60/kg (\$M/yr)		21.90	
profit	11.52	8.78	7.82
NPV 20 years^a	48.96	23.7	28.08

^aThe net present value (NPV) was calculated on the basis of a 20 year plant life, 10% interest rate, 38.9% income tax rate, 2.5% fixed operating cost, and a depreciation schedule based on the modified accelerated cost recovery system that was developed by the Department of Energy for water electrolyzer and hydrogen technologies.

A CH instrument 660D potentiostat (USA) equipped with an Amp booster, and Keithley Precision Measurement DC Supply were used for electrolysis experiments. An Ag/AgCl (3 M NaCl) reference electrode (BASi) was used for cathode potential measurements. A gas chromatography instrument (GC; PerkinElmer, Clarus 580), equipped with a packed MolSieve 5 Å column and a packed HayeSepD column, was used to detect CO and H₂ using a flame ionization detector (FID) and a thermal conductivity detector (TCD), respectively. The concentrations of the products CO and H₂ (ppm) in the headspace of the catholyte reservoir were quantified by calibrating the signal area for CO and H₂ to known concentrations of the two gases.

Electrode Preparation. The silver foam was cut into the desired dimensions (2 × 2 cm) with a blade and washed with acetone and water. The silver foam was treated with dilute nitric acid solution (30% v/v HNO₃) in a 50 mL beaker for 10 s to remove the oxide layer and increase its electrochemical surface area. The etched silver foam was then washed thoroughly with deionized (DI) water and 3 M KHCO₃ prior to use.

To fabricate the Pt/C gas diffusion electrode (GDE), 8 mg of Pt/C was added to a 2.5 mL mixture of water and isopropanol (IPA) solution (V_{H₂O}:V_{IPA} = 4:1) with 20 wt % Nafion solution (5 wt %). The ink was sonicated in a bath sonicator for 15 min and then drop-casted onto a GDL. The fabricated GDEs were then stored in a fume hood to dry overnight.

Electrolysis. A peristaltic pump was used to deliver 1.0 M KOH to the anode of the control system at a constant flow rate of 40 mL min⁻¹. High-purity H₂ (10–200 sccm, 99.999%) was humidified in a bubbler held at a constant temperature of 60 °C prior to being fed to the anode of HORICEM|HCO₃⁻ (Figure S5). Two 127 μm polytetrafluoroethylene (PTFE) gaskets were placed separately between the anode and anodic flowplate and the cathode and cathodic flowplate to prevent leakage for all of the electrolyzers. The catholyte (3.0 M KHCO₃) was delivered at a constant flow rate of 100 mL min⁻¹ for all electrolyzers. Gaseous products (e.g., H₂ and CO) in the headspace of the cathode electrolyte reservoir were delivered to an in-line gas chromatograph (GC) by a constant rate of N₂ flow at 175 sccm for quantification at 350 s. The actual flow rate of the gas mixture was measured by a flow meter positioned immediately downstream of the GC. Liquid products collected from the catholyte after 1200 s of electrolysis were quantified by ¹H NMR spectroscopy using potassium hydrogen phthalate as the internal standard with a calibration curve.

Safety Statement. No unexpected or unusually high safety hazards were encountered.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acscentsci.2c00329>.

Faradaic efficiency calculation, liquid product detection, CO₂ utilization calculation, pressurized electrolyzer test station, technoeconomic analysis, energy consumption analysis (Sankey diagram), diagram of the experimental setup, the effect of the H₂ flow rate on the cell voltage and FE_{CO}, membrane thickness effect, V_{cell}–*j*_{CO} relationship, stability test, and H₂ recycling data (PDF)

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Author Contributions

[†]Z.Z. and E.W.L. contributed equally to this work.

Author Contributions

C.P.B. supervised the project. Z.Z. and C.P.B. conceived the study, and Z.Z. performed the experiments. E.W.L. designed the pressurized system, performed the technoeconomic analysis, and determined the mass and energy balances. S.R. performed experiments and an analysis and aided in technical editing. B.A.W.M. performed an analysis and aided in writing and technical editing. A.H. contributed to an analysis of experimental data. C.P.B., Z.Z., and E.W.L. wrote the first draft of the manuscript. All authors contributed to the writing of the final manuscript.

Notes

The authors declare the following competing financial interest(s): The authors C.P.B. and Z.Z. have filed a patent related to this work (No. PCT/CA2022/050094; US patent application No. 63/140.167).

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