ANALYSIS

https://doi.org/10.1038/s41893-021-00739-x



Techno-economic assessment of low-temperature carbon dioxide electrolysis

Haeun Shin ^{1,2}, Kentaro U. Hansen ^{1,2} and Feng Jiao ¹

□ ^{1,2}

Low-temperature CO₂ electrolysis represents a potential enabling process in the production of renewable chemicals and fuels, notably carbon monoxide, formic acid, ethylene and ethanol. Because this technology has progressed rapidly in recent years, a systematic techno-economic assessment has become necessary to evaluate its feasibility as a CO₂ utilization approach. Here this work provides a comprehensive techno-economic assessment of four major products and prioritizes the technological development with systematic guidelines to facilitate the market deployment of low-temperature CO₂ electrolysis. First, we survey state-of-the-art electrolyser performance and parameterize figures of merit. The analysis shows that production costs of carbon monoxide and formic acid (C₁ products) are approaching US\$0.44 and 0.59 kg⁻¹, respectively, competitive with conventional processes. In comparison, the production of ethylene and ethanol (C₂ products) is not immediately feasible due to their substantially higher costs of US\$2.50 and 2.06 kg⁻¹, respectively. We then provide a detailed roadmap to making C₂ product production economically viable: an improvement in energetic efficiency to ~50% and a reduction in electricity price to US\$0.01 kWh⁻¹. We also propose industrially relevant benchmarks: 5-year stability of electrolyser components and the single-pass conversion of 30 and 15% for C₁ and C₂ products, respectively. Finally we discuss the economic aspects of two potential strategies to address electrolyte neutralization utilizing either an anion exchange membrane or bipolar membrane.

he massive quantities of fossil fuels used by our society have led to unprecedented atmospheric CO2 levels, with widespread climate impacts^{1,2}. Carbon capture, utilization and storage (CCUS) technologies are being developed to mitigate CO₂ emission issues^{3,4}. Large-scale CO₂ capture and sequestration facilities, such as Petra Nova⁵, have been built to store thousands of tons of CO₂ per day. However, the typical capital investment required for centralized CCUS facilities is at a billion-dollar scale^{6,7}, making it challenging to finance. Sequestering captured CO₂ in geological repositories often requires additional investment in CO₂ pipelines and infrastructure⁸, which further increases the financial challenge to the rapid deployment of highly centralized facilities. More importantly, the carbon capture and sequestration process itself is not profitable without subsidies or a carbon tax8. As a critical component of CCUS, carbon utilization holds the key to generation of revenues that can offset capture cost. It enables the conversion of captured CO₂ into valuable materials such as concrete, building materials and platform molecules for fuel and chemical production³.

Utilizing electrical energy, CO_2 electrolysis reduces CO_2 at the cathode while oxidizing water at the anode, generating oxygen for venting to the atmosphere. In the past decade, notable progress has been made in CO_2 electrolysis technologies, including high-temperature routes based on solid-oxide electrolytes and low-temperature routes based on polymer membrane electrolytes⁹⁻¹³. Uniquely, low-temperature, membrane-based CO_2 electrolysers can directly produce multi-carbon (C_{2+}) products, such as ethylene (C_2H_4) and ethanol (EtOH), from CO_2 and water^{14–16}. At a laboratory-bench scale, the research focus for low-temperature CO_2 electrolysis has been directed towards the production of carbon monoxide (CO), formic acid (CO), ethylene and ethanol at industrially relevant current densities, with each product having unique market conditions^{17–23} (Fig. 1a). Compared to C_1 products, C_2 products have a larger global market by order of magnitude while

market prices, when normalized to the number of carbon molecules, are less by more than a factor of two. Simultaneously, for each CO_2 molecule that is electrochemically reduced, C_2 products would require triple the amount of charge passed, thus utilizing more electricity and incurring substantial production costs.

Fortunately, the decreasing cost of renewable electricity sources makes electrochemical CO_2 utilization technologies increasingly viable for commercial applications. Recent studies on renewable electricity generation show a decreasing trend for photovoltaic electricity prices over time, with a projected price as low as US\$0.03 kWh⁻¹ in the near future^{24,25}. A similar trend also holds for wind energy, with wind electricity price already being ~US\$0.02 kWh⁻¹ (ref. ²⁶). Moreover, recent developments in modular and distributed manufacturing have introduced a new angle to the economy of scale²⁷. Therefore, when scaled within the range 0.1–100 MW per unit, the deployment of CO_2 electrolysis presents an exciting opportunity for local linkage of renewable energy sources with CO_2 capture and sequestration facilities.

Initial techno-economic studies $^{28-35}$ have shown that CO_2 electrolysis can be economically feasible, while citing that the cost of electricity and electrolyser energy efficiency are primary cost obstacles. Furthermore, the need to develop practical electrolysers that can operate beyond a threshold current density (> $100\,\mathrm{mA\,cm^{-2}}$), to limit the electrolyser's capital cost, has been identified and met. In the past 5 years, numerous reports have demonstrated electrolyser configurations designed to maximize both CO_2 availability at the cathode and catalytically active surface area for CO_2 reduction electrodes, while also minimizing internal resistance spanning the whole system (Supplementary Note 1). Shown in Fig. 2, in the aggregate, there are now sufficient data available to make better estimates for performance metrics that are economically relevant.

Nevertheless, previous analyses and existing challenges leave us with an incomplete roadmap for low-temperature CO₂ electrolysis.

²These authors contributed equally: Haeun Shin and Kentaro U. Hansen. [™]e-mail: jiao@udel.edu

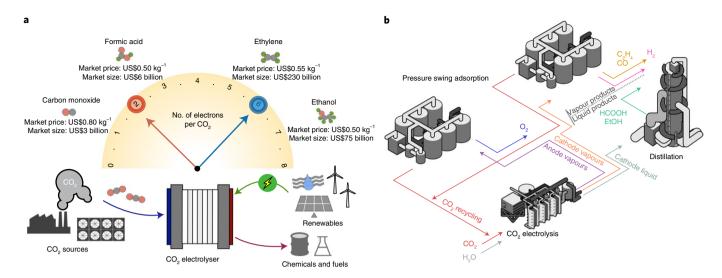


Fig. 1 | Chemical production process via low-temperature CO_2 electrolysis. **a**, Schematic of CO_2 electrolysis and market data on selected products. For visual clarity, only chemicals investigated as potential products are shown. Prices and global market size are taken from various sources¹⁷⁻²³ and should be taken as approximations for process feasibility. **b**, Simplified flow diagram for modelled CO_2 electrolysis plant. CO_2 electrolysis is modelled in series with a pressure swing adsorption process for the separation of vapour species. When the primary product is liquid phase, the distillation process is included in the model for separation of liquid species.

First, guidance for realistic single-pass conversions and active component stability remains arbitrary and ambiguous. Moreover, the severity of electrolyte carbonation for electrolysers operating with a locally alkaline cathode surface has been identified, with potentially crippling economic ramifications³⁴. One strategy is to utilize a fully carbonated system with an anion exchange membrane (AEM) that leads to consumption of CO₂ at the cathode and re-emission of CO₂ with oxygen at a locally acidic anode. While an additional separation step is required for anode vapours to recover CO₂, the system is fully carbonated, eliminating the need to alkalize the electrolyte perpetually. Another strategy is to utilize a bipolar membrane (BPM) that curtails this phenomenon altogether. However, current commercial BPMs result in electrolysis with substantially higher internal resistance. The economics for these strategies aimed at addressing this issue have not been incorporated into previous analyses.

Here, we conducted a comprehensive techno-economic assessment (TEA) on low-temperature CO2 electrolysis processes based on a polymer membrane electrolyte for four major products: carbon monoxide, formic acid, ethylene and ethanol. We first built a techno-economic model to conduct a product-by-product analysis. The modelled system consists of pressure swing adsorption units for gas separation and distillation for liquid separation in tandem with the electrolyser, as shown in Fig. 1b. Beyond using parameters that reflect recent progress in the field, we implemented a voltammetric model³⁶ (relating electrolyser utilization to inefficiency), considered anode vapour separations when necessary and explored the two aforementioned strategies to address electrolyte carbonation issues34 and thus improve the rigour of TEA. We then identified the remaining electrolyser improvements from our TEA and, more specifically, industrially relevant single-pass conversions and electrolyser stabilities, to target for accelerated testing and pilot-scale demonstrations for all products.

Results

Cost analysis for carbon monoxide and formic acid (both C_1). We performed a single-variable sensitivity analysis to answer the following: (1) what are the estimated production costs for state-of-the-art (SOA) CO_2 electrolysis processes? (2) What are the main parameters contributing to the determination of each product's

production cost? Notably, the sensitivity analysis baseline shown in Fig. 3 was chosen based on a recent literature survey and, hence, the base production cost of each product captures the near-term economic viability of the current technology. The baseline parameters are tabulated in Supplementary Table 6, and each optimistic and pessimistic parameter is tabulated in Supplementary Table 8. It is worth mentioning that different cell configurations were chosen for different products—the AEM configuration for carbon monoxide and BPM configuration for formic acid-based on the membrane cost analysis described in Supplementary Note 7. According to the cost analysis, the base production costs were set at US\$0.44 and 0.59 kg⁻¹ for carbon monoxide and formic acid, respectively. Although the market prices for those products vary between different regions and production methods, the base production costs fall within the range of market prices (Fig. 1a)17,19,21, indicating the cost competitiveness of CO₂ electrolysis for C₁ products compared to traditional production processes.

Interestingly, the change in production cost in the sensitivity analysis is nonlinear for several parameter changes, illustrated by skews in the plotted bar plots. Thus, improvement in several parameters (including single-pass conversion for carbon monoxide and current density for formic acid) would result in diminished benefits for cost reduction. It must be noted that the same voltammetric model was used for the sensitivity analysis, leaving an opportunity for further cost reduction when cell performance improves. Among all parameters, reduction in electricity price and stack cost has the greatest potential for cost reduction (>10% of the overall production cost for each improvement) for all C₁ products. The optimization of single-pass conversion and current density for carbon monoxide, current density and membrane electrode assembly (MEA) replacement interval for formic acid would be the next priorities. In detail, 7 and 9% cost reduction for carbon monoxide and formic acid, respectively, is expected when the stack cost reaches US\$250 kW⁻¹. Alternatively, operating at lower current density with larger electrolysers is possible at the same production cost. (that is, operating at roughly 50% lower current density with US\$250 kW⁻¹ stack cost; Supplementary Fig. 7). A stack cost of US\$230 kW⁻¹ is attainable when iridium-based anode catalysts, the well-known oxygen evolution reaction (OER) catalyst, are replaced with non-precious metals

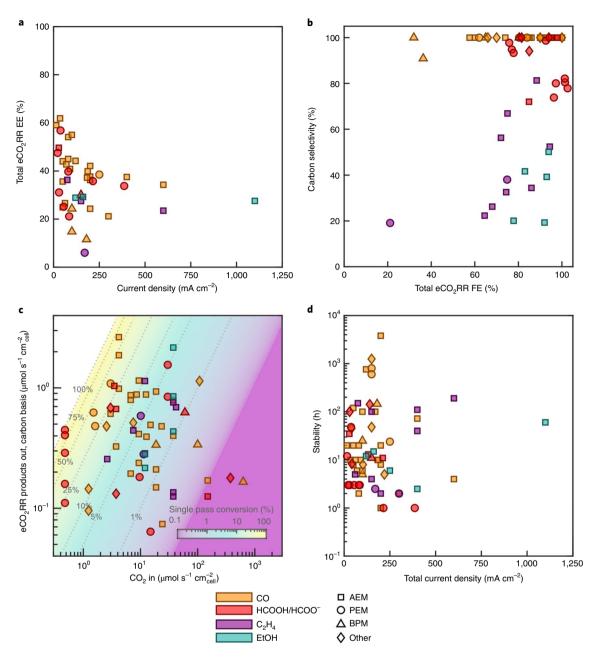


Fig. 2 | Laboratory-bench-scale CO₂ electrolysis performance. a-d, The surveyed literature was selected with the criterion that >1h of stable performance is reported with at least 10 mA cm⁻² current density towards eCO_2RR for the production of carbon monoxide, formic acid, ethylene and ethanol. Only data from stability tests are included. Marker shape denotes the type of membrane used to facilitate ion transport: AEM, proton exchange membrane (PEM) or reverse-bias BPM. **a**, Full-cell EE (for reports with a full-cell configuration) versus total cell current density. **b**, Carbon selectivity versus FE towards eCO_2RR . Carbon selectivity considers only the carbon ratio of production rates for the main product versus the total electrochemical conversion rate of CO_2 . **c**, Single-pass conversion of CO_2 represented as a heatmap over the log-log plot of the molar flow rate of carbon fed in (as CO_2) and out (as eCO_2RR products). **d**, Duration of stability test for each report. HCOO⁻, formate. A detailed set of criteria for the survey is available in the Methods.

such as nickel-based catalysts, assuming the same catalytic performance. Recent studies have shown improved OER activities, with nickel-based anodes demonstrating several hundred-hours-long operation with <400 mV overpotentials in neutral media ^{37,38}. Operating in neutral media can also alleviate the stability issue of AEMs under high pH, which has recently been raised ³⁹. Lowering of iridium loading can also substantially lower the stack cost, since it alone accounts for >50% of the stack cost with 2 mg cm⁻² loading (Supplementary Note 2). Another potential lies in different anode reactions. For instance, one recent study manifested the feasibility of an alternative anode reaction with glycerol oxidation replacing

OER, which requires less energy input and creates the potential of valuable chemical production other than oxygen, such as epoxidation chemistry and inexpensive catalyst discovery³¹. However, for broad implementation, the ubiquity, cost of water and ability to vent oxygen to the atmosphere still make OER compelling. For example, while the selective electro-oxidation of glycerol to formic acid could be appealing when coupled with a cathode also producing formic acid⁴⁰, the cost of glycerol would be of great importance. For instance, based on a spot price ranging US\$0.25–1.00 kg⁻¹ (ref. ⁴¹), using a refined glycerol feedstock would cost US\$0.17–0.67 kg_{HCOOH}⁻¹ (for formic acid generated anodically), which could undermine the

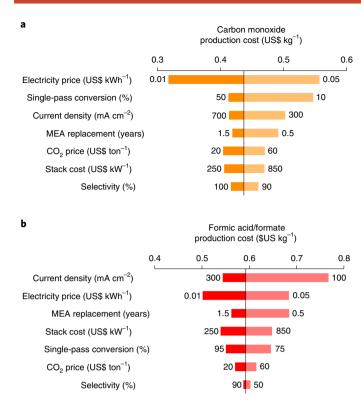


Fig. 3 | Production cost changes for various parameters. a,b, Single-variable sensitivity analysis for the production cost of carbon monoxide (US\$0.44 kg⁻¹; **a**) and formic acid (US\$0.59 kg⁻¹; **b**). The baseline parameters tabulated in Supplementary Table 6 were chosen based on the current SOA. The values shown in the figures indicate each optimistic and pessimistic parameter. Detailed cost breakdowns for all products, and sensitivity analysis for ethylene and ethanol, are provided in Supplementary Figs. 5 and 6.

benefits realized from improvement in electrolyser performance. Nonetheless, breakthroughs in the development of promising anode catalysts and reactions can provide exciting opportunities for linkage of renewable electricity and electrolysis to other industries.

Sensitivity analysis also shows that the production cost is most susceptible to variation in electricity price cost, from US\$0.01 to 0.05 kWh⁻¹. Nonetheless, reasonable production costs with an electricity price of US\$0.03 kWh⁻¹, and US\$0.44 and 0.59 kg⁻¹ for carbon monoxide and formic acid, respectively, indicate that it should be profitable in the short term to produce these products via CO₂ electrolysis^{25,42}. However, unlike the production cost of carbon monoxide, that of formic acid begins to deviate from the reference cost at >US\$0.03 kWh⁻¹, indicating a higher sensitivity to change in electricity price mainly due to the voltage penalty derived from a BPM configuration. Variation in voltage penalties emphasizes that optimal operating conditions depend on the configuration: designing the process to operate at low, rather than high, current density can be more economically sensible because the cost of electricity shifts the cost-benefit of operating at higher current densities (Supplementary Fig. 8).

Key technical gaps regarding ethylene and ethanol products (both C_2). Considering the large market size for C_2 products (Fig. 1a), the impact of shifting their production method to a renewable route will be considerable. However, the original production cost estimate for C_2 products is untenable with existing market conditions of US\$2.48 and $2.06 \, \mathrm{kg}^{-1}$ for ethylene and ethanol, respectively (Figs. 1 and 4). Here, we show a potential roadmap for achieving a

more competitive production cost in the form of a waterfall analysis to understand the cumulative result of several process optimizations (Fig. 4). From a single-variable sensitivity analysis (Supplementary Fig. 6), high electricity consumption and low selectivity must be reconciled for feasibility. Hence, we consider a mix of improvement in electrolyser efficiency and lowered electricity cost as the first steps in the roadmap. More than 30% of the production cost is eliminated for both C, products by improvement in faradaic efficiency (FE) to >80% and rescaling the voltammetric model to match 50% of energetic efficiency (EE) at baseline current density (Supplementary Fig. 2). One pathway to achieving the rescaled voltammetric model involves improvement in both membrane and cathode (Supplementary Note 5 and Supplementary Fig. 4). However, one issue with this path is that excellent efficiency is often achieved with an alkaline electrolyte43,44 that is inherently prone to gradual carbonation⁴⁵. The regeneration cost of a carbonated electrolyte could introduce a cost of approximately US\$0.2-0.3 kg⁻¹ (ref. ³⁴), or even more for non-zero-gap configurations with >75% of fed CO₂ loss in alkaline solution⁴⁶. Also, high FE for C₂ products at >500 mA cm⁻² needs to be replicated under fully carbonated conditions. Moreover, inefficiencies other than the cathode could be minimized for the membrane and anode⁴⁷. However, more detailed recommendations are precluded by the lack of studies experimentally identifying the energetic losses throughout practical electrolyser configurations (that is, at the anode and membrane).

Other sources of cost reduction in their summation are as crucial as improvement in electrolyser performance. Cheap electricity is critical to achieving reasonable C2 costs, for example, with each US\$0.01 kWh⁻¹ corresponding to a price change of US\$0.30 kg⁻¹ for ethylene. However, cheap renewable electricity will probably be intermittent if produced from renewable sources and, whether the levelled cost of renewables falls <US\$0.03 kWh⁻¹ is still an open question. Operating at a higher current for shorter periods would address the issue of intermittent availability. However, a deep understanding of the implications of intermittent stress on the electrolyser on durability/stability is still lacking. In this analysis, we scaled the process based on 96% production capacity; notwithstanding daily fluctuations, total wind electricity generation can deviate by >50% monthly⁴⁸. Therefore, the roadmap indicates that economic C, production needs a fundamentally different approach based on intermittent cheap electricity. The use of intermittent production modelling, with simulated solar or wind profiles, as has been performed for high-temperature CO₂ electrolysis⁴⁹, will be vital to determine the required variable range of operating current for a given electrolyser size to leverage the majority of intermittent electricity sources. Reducing or eliminating iridium-based anodes to reduce stack cost could also decrease total production cost, as discussed above. Reducing replacement costs by increasing electrolyser stability to 5 years, in reference to a 5-year MEA replacement interval, could also help, but the development of a set of stability test protocols is necessary for rigorous study of MEA lifespan, which is still nascent and requires more work. Figure 4 illustrates how economic factors, such as a carbon tax credit (based on US Federal 45Q tax credits⁵⁰) and selling of hydrogen (at US\$2 kg⁻¹)⁵¹ could lower total production but are marginal at 10% of the initial cost. However, this might be negated by coproducing other electrochemical CO₂ reduction (eCO₂RR) products, leading to diseconomies of scope from other products' market conditions or necessitating additional downstream processes. Fortunately, progress is being made to make a copper catalyst more selective to either of the C_2 products 16,44,52 .

In conclusion, the roadmap for C_2 products diverges from the immediately feasible production of C_1 products. The intrinsically high energy demand, requiring six electrons rather than two per CO_2 , exacerbates electrolyser inefficiencies and electricity costs. For C_2 products, using a BPM without appreciable improvement⁵³ is out of the question even if it mitigates anode separation and improves

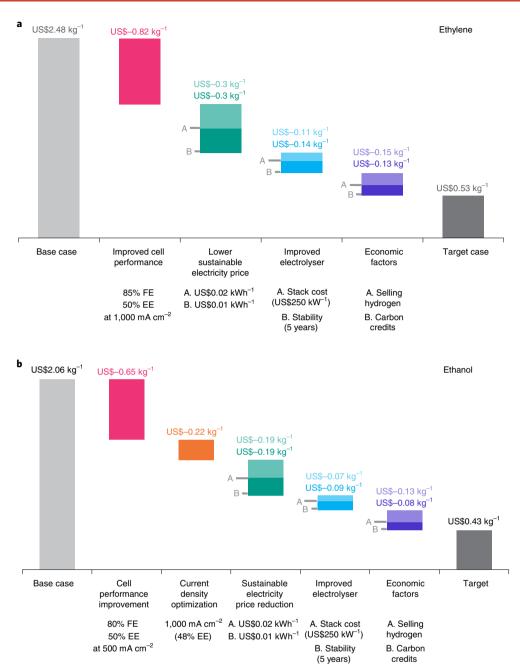


Fig. 4 | Roadmap to reducing base case production cost by successive changes to cost-relevant parameters. a,b, Production of ethylene (a) and ethanol (b).

single-pass conversion (Supplementary Fig. 11). However, we have shown that it is possible to achieve reasonable production costs and identify the key steps to realize its market feasibility. The immediate steps will be to elucidate losses in electrolysers in situ rigorously⁵⁴, such as with other electrochemical systems^{55–57}, and to evaluate what peak current densities are necessary to enable economically feasible intermittent operation.

CO₂ electrolyser stability and conversion benchmarks. The feasibility of prototype electrolysers must be validated by operation at practical current density, stability and single-pass conversion. Here we provide feasibility-driven benchmarks for such tests by evaluating the conditions necessary to keep production costs constant. Figure 5 shows the economic trade-off between current density and either single-pass conversion or stability. The objective here is to provide a basis for economic trade-offs to determine the design

space for CO_2 electrolysis components. While only one cost curve is shown for visual simplicity for each product, the trends shown here can still serve as a basis for rational test designs in the development of new materials for CO_2 electrolysis.

The active part of the electrolyser, MEA, consists of the polymer electrolyte, catalyst support and the catalyst material and is assumed to be consumable for the process requiring routine replacement. Figure 5a puts into context the current density–MEA stability relationship for each product, with recent experimental reports. Higher current density reduces the electrolyser's capital cost, and improved stability mitigates consumable costs. For simplicity we define stability as the time required for the MEA to be rendered inoperable and assume perfect operation otherwise. Holding production costs constant, we identify asymptotic limits for current density and MEA stability that provide the most lenient limits for stability testing. For all products, increasing stability beyond 5 years provides only

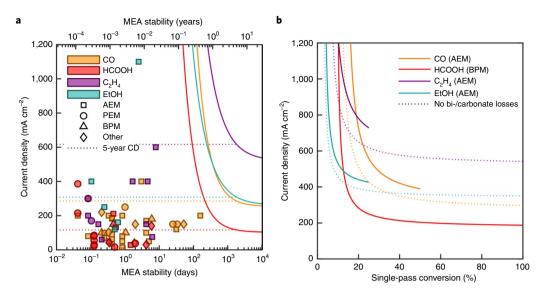


Fig. 5 | Electrolyser current density required to maintain constant production cost. a,b, Production cost is set at US\$0.44, 0.59, 1.68 and $1.41 \, \mathrm{kg^{-1}}$ for carbon monoxide, formic acid, ethylene and ethanol, respectively, with either MEA stability (modelled as a replacement interval; **a**) or single-pass conversion of CO_2 (**b**). CD, current density. Ethylene and ethanol were modelled with improved voltammetric performance. The stability plot is overlaid with experimental stability test durations as listed in Supplementary Table 1. For the conversion plot, carbonate crossover mitigation was considered to create curves extending out to 100% conversion.

marginal improvements to production costs. Therefore, the current densities for 5-year MEA replacement intervals (approximately 250, 100, 300 and $600\,\mathrm{mA\,cm^{-2}}$ for carbon monoxide, formic acid, ethylene and ethanol, respectively) provide a reasonable baseline, or the minimum target current density, for a practical stability test. The asymptotic behaviour of the curves in Fig. 5a also indicates that, at a minimum, stability to at least $100\,\mathrm{days}$ is a good first target for a prototype electrolyser.

Nonetheless, Fig. 5a depicts the clear gap between the literature and practical stability target for industrial application. The major causes of stability loss are often found in poor water management. Local dry out can limit the ion conductivity and local flooding can inhibit CO₂ permeation, both critical properties necessary for maintaining the triple-phase boundary where the reaction occurs. Once the catalyst layer fails to maintain its hydrophobicity, penetration of electrolyte, flooding, hinders CO2 access to the catalyst surface, leading to an unwanted hydrogen evolution reaction. Moreover, salt precipitation severely damages the integrity of MEA⁵⁴. To estimate the economically feasible region of current density and replacement rates considering stability losses, accelerated stability tests are necessary. However, definition of the performance reduction at which MEA should be replaced (for example, 10% reduction in EE) will remain an open question until key stability/durability relationships are experimentally established via accelerated testing protocols. Addressing the lack of these methods, Kenis and coworkers reviewed a series of postmortem characterization techniques, condition cycling strategies and different electrolyte concentrations to rapidly develop such a test⁵⁸. In situ techniques to break down cell voltage into constitutive components (for example, transmembrane potential drop), as demonstrated by Berlinguette and coworkers, are also of interest in identification of individual component deterioration rates⁵⁴. Furthermore, the development of routines for recovery of performance, efficient material recovery and streamlined catalyst production can substantially reduce the cost of replacement as complementary approaches⁵⁹.

We also evaluated to what extent the single-pass conversion of CO_2 feed will impact product costs. As shown in Fig. 5b, similar asymptotic limits exist with inflection points near 30 and 15% single-pass conversion for C_1 and C_2 products, respectively.

However, there is a break in the curves. In the current model we account for the crossover of bi/carbonate anions (when necessary), which increases separation costs and parasitically consumes CO₂ fed to the cathode, limiting single-pass conversion. The modelling and in-depth analysis addressing this issue are discussed in Supplementary Note 7. Overall, most of the benefits of increasing single-pass conversion are captured before the limiting point, which is mostly in agreement with a previous TEA study showing that 22% of single-pass conversion is required for optimization of the cost of carbon monoxide production at 80 mA cm⁻² (ref. ⁶⁰). Regardless, there is still a cost reduction from assuming no crossover, represented by the shift in required current density shown as the dotted curves in Fig. 5b. Coupling the analysis with the current SOA shown in Fig. 2, sufficient single-pass conversions (>30% for C₁ products) and insufficient single-pass conversions (<5% for C₂ products) have been generally demonstrated with >1 h of performance data.

Discussion

Low-temperature CO₂ electrolysis is a potential means for offsetting carbon capture costs while providing valuable products from captured CO₂. In this work, a comprehensive TEA outlines a path for development of CO₂ electrolysis at scale and economically for both C_1 and C_2 products, summarized in Table 1. We also provide specific system design guidelines for single-pass conversion and stability and explore the best opportunities for reduction in each product's cost. We consider two short-term solutions for addressing the bi/carbonate issue: operating with either an AEM using a fully carbonated electrolyte or a BPM that mitigates the bi/carbonate issue at the cost of an energetic penalty. Comparing the two methods side by side (Supplementary Note 7), we determined that a BPM makes sense only for formic acid production. The results here indicate that C₁ products are economically feasible based on the current literature, while C2 products are currently not, the main stumbling block being EE. Waterfall analysis shows specific implementation plans for profitable manufacture of C₂ products: intermittent operation at high-current densities is a primary mode of operation when electricity is in overabundance. The main technical challenge with direct CO₂ electrolysis to C₂ products is improving operation under nonalkaline conditions where the bi/carbonate

Estimated production cost	Carbon monoxide (US\$0.44 kg ⁻¹)	Formic acid (US\$0.59 kg ⁻¹)	Ethylene (US\$2.48 kg ⁻¹) (US\$1.68 kg ⁻¹) ^a	Ethanol (US\$2.06 kg ⁻¹) (US\$1.41 kg ⁻¹) ^a
TEA analysis	SOA electrolyser performance is sufficient for economic feasibility. The following optimizations should be considered: stack cost reduction (for example, via the development of non-precious anodes) and separation cost reduction (for example, via the use of processes with lower energy demand and capital cost).		SOA electrolyser performance is insufficient for economic feasibility, but opportunities and a roadmap to reducing costs exist. Improvement in EE and product selectivity is the most important technical challenge. The impact of expanding the economy of scope to byproducts (for example, hydrogen) and other eCO ₂ RR products should be explored.	
	Access to inexpensive renewable electricity sources is a crucial component to profitability. The impact of intermittent availability of cheap renewable electricity on operating capacity and production schedules is not considered herein.			
Electrolyser optimization	Membrane: mitigation of bi/carbonate crossover is generally desirable but with the following conditions: limited accumulation of carbonate salts at the cathode and limited increase in energetic losses to water splitting and high contact resistances.			
	CO_2 conversion: production can be cost competitive within the theoretical limits posed by using an AEM C_1 product. SOA performances are close to the theoretical limit of conversion (50%), but those of C_2 products are generally further from the theoretical limit (25%).			
	Durability: the target time is 5 years of operation lifespan, or shorter lifespans with methods that restore performance routinely and limit replacement costs. The development of robust MEA with better water and salt management is desirable.			

Production cost using an improved voltammetric performance model. Costs are calculated with US\$0.03 kWh⁻¹ for the electricity price with current densities of 500, 200, 1,000 and 500 mA cm⁻² for carbon monoxide, formic acid, ethylene and ethanol, respectively. Production costs using different electricity prices and current densities are provided in Supplementary Fig. 8.

issue (with <75% CO₂ losses to the electrolyte) is not economically egregious. However, the scope of the challenge in achieving 50% full-cell EE and >80% CO₂ reduction FE at >500 mA cm⁻² requires a holistic analysis of all electrolyser components to achieve this goal. Alternatively, the scope of improvement warrants exploration and demonstration of whether other routes—for example, tandem CO₂ electrolysis^{32,34}—can achieve a net equivalence for efficiencies and capital cost. Regardless, there is a pressing need to demonstrate process stability and durability in the order of 5 years at relevant current densities for all products. Accelerated testing protocols could also inform whether intermittent, high-current-density operation is feasible⁵⁸ and compatible with renewable electricity sources.

Additionally, future work has many interdisciplinary points that must be addressed to position low-temperature CO_2 electrolysis as a potential economic on-ramp for carbon capture, utilization and storage. Accounting for fluctuations in operating schedules and price (variable abundance of cheap renewable electricity) is inevitable and is challenging to account for in economic models. The alternative scenarios for gas separation where the primary species is CO_2 (>50%) and for distillation for liquid separation with lower energy intensity are of interest. Overall, these concepts would tie in well with the broader context of the CO_2 utilization technique presented in Supplementary Fig. 13.

Methods

Literature survey. To identify initial estimates for electrolyser performance parameters, a literature survey of works published from 2016 to 2020 was conducted using ScienceDirect. Several cutoff criteria were imposed to limit the survey to practical electrolyser configurations, namely: (1) at least 1 h of stability test data with either half- or full-cell voltage reported; (2) at least 10 mA cm $^{-2}$ of net eCO $_2$ RR current density demonstrated; (3) test data report product FEs or selectivity; and (4) the electrolyser configuration operates in flow-cell mode. Details of the survey are given in Supplementary Note 1 and surveyed works are tabulated in Supplementary Tables 1–4.

Techno-economic model. The techno-economic model used in this analysis is based on our previous model³³. Several parameters were updated to capture the current technology cost. All parameters changed from the previous model are marked with an asterisk in Supplementary Table 7, but are otherwise the same. The production cost was numerically evaluated by solving the product selling

cost, resulting in a net present value of zero at the end of the process life. The modelled system consists of pressure swing adsorption units for gas separation and distillation for liquid separation in tandem with the electrolyser, as shown in Fig. 1b. For liquid products, formic acid and ethanol, 10% product concentration is assumed before entering a distillation process. The outstanding technical challenges for electrolysis at high liquid product concentrations are elaborated in Supplementary Note 2. The additional protonation process of formate is excluded in this study. For the electrolyser stack cost and MEA replacement cost, a detailed derivation for the selected values is provided in Supplementary Note 2. A calculation example set is provided in Supplementary Note 3. The initial scale of production was set at 50,000 kg d⁻¹ of product. This corresponds to an electrolyser size of approximately 10 MW for C₁ products, 100 MW for C₂ products and 50 MW for C₂ products, assuming an optimistic voltammetric model. A sensitivity analysis looking at changing the process scale is described in Supplementary Note 6 and is available as Supplementary Fig. 9. For single-variable sensitivity analysis there are product-dependent (that is, membrane configuration, current density, FE and single-pass conversion) and product-independent parameters (MEA replacement interval, stack cost, CO, price and electricity price). Product-dependent parameters were selected based on their SOA, and hence their base scenario captures current technology production cost. For single-pass conversion, the optimistic scenario was based on the maximum single-pass conversion possible. These parameters are provided in Supplementary Table 8. Additionally a cost breakdown, assuming all optimistic parameters simultaneously, and sensitivity analysis results for C2 are provided as Supplementary Figs. 5 and 6, respectively. The cost breakdown for all products is presented as a fraction of the total production cost (US\$ kg⁻¹) by normalization of both capital and operating costs assuming a 20-year lifespan. In the current model, we also consider the loss of feedstock CO2 to the electrolyte in the form of carbonate species 45,46,61, ultimately resulting in the additional separation of CO₂ from the anode by assuming a fully carbonated electrolyte in the case of AEM-based cell configuration (Supplementary Fig. 10). For simplicity, when an AEM is used we assume that CO₂ consumption to carbonation is proportional to the electrochemical conversion rate, based on the stoichiometric ratio of electrons per CO₂. Assuming only CO₃²⁻ transport, C₁ single-pass conversion is limited to 50% with a 1:1 conversion/consumption ratio, and C2 single-pass conversion is limited to 25% with a 1:3 conversion/consumption ratio (Supplementary Note 7). A separate pressure swing adsorption unit for the separation of oxygen from CO₂ at the anode is assumed.

Electrolyser configuration and voltammetric model. For the electrolyser unit we assume the use of a MEA configuration to minimize internal resistances, a carbon selectivity of unity for all carbon-based products and a FE of unity with hydrogen evolution as the only side reaction (that is, non-CO₂ reduction reaction) at the cathode. Moreover, we implement a voltammetric model similar to that of Orella et al. 36 to investigate the relationship between parameters for a given production cost while approximating best-in-class electrolyser performance. Notably, several

terms accounting for concentration gradient arising from high single-pass conversion have been added, and are elaborated in Supplementary Note 4. An overview of relevant parameters and the resulting voltammetric model is discussed in Supplementary Note 5, and the specifics of techno-economic parameters are provided in Supplementary Note 2. To assess the impact of assumptions regarding stack cost and electricity price, as both terms greatly impact production cost, we conducted a separate analysis of these parameters, presented in Supplementary Figs. 7 and 8 and discussed in Supplementary Note 6.

Data availability

The spreadsheet used for cost analyses is available in Supplementary Data 1 (ref. ³³). It includes analyses for two different cell configurations—AEM and BPM—with different voltammetric models.

Code availability

The MATLAB codes for voltammetric profiles are given in Supplementary Data 2.

Received: 26 January 2021; Accepted: 31 May 2021; Published online: 12 July 2021

References

- IPCC: Summary for Policymakers. In Special Report on Global Warming of 1.5°C (eds Masson-Delmotte, V. et al.) (IPCC, WMO, 2018); https://www. ipcc.ch/sr15/chapter/spm/
- Chu, S. & Majumdar, A. Opportunities and challenges for a sustainable energy future. *Nature* 488, 294–303 (2012).
- Pales, A. F. et al. Exploring Clean Energy Pathways: The Role of CO₂ Storage (IEA, 2019); https://www.iea.org/reports/the-role-of-co2-storage/
- Roh, K. et al. Early-stage evaluation of emerging CO₂ utilization technologies at low technology readiness levels. Green Chem. 22, 3842–3859 (2020).
- Kennedy, G. W. A. Parish Post-Combustion CO₂ Capture and Sequestration Demonstration Project Final Technical Report Report DOE-PNPH-03311 (US Department of Energy Office of Scientific and Technical Information, 2020); https://doi.org/10.2172/1608572
- Friedmannn, J., Ochu, E. & Brown, J. D. Capturing Investment: Policy Design to Finance CCUS Projects in the US Power Sector (Columbia School of International and Public Affairs, 2020); https://www.energypolicy.columbia. edu/research/report/capturing-investment-policy-design-finance-ccusprojects-us-power-sector
- Ekmann, J., Huston, J. & Indrakanti, P. Carbon Capture, Utilization, and Storage: Technology and Policy Status and Opportunities (National Association of Regulatory Utility Commissioners, 2018); https://pubs.naruc.org/pub/ 09B7EAAA-0189-830A-04AA-A9430F3D1192
- Edwards, R. W. & Celia, M. A. Infrastructure to enable deployment of carbon capture, utilization, and storage in the United States. *Proc. Natl Acad. Sci.* USA 115, E8815–E8824 (2018).
- Jhong, H.-R. M., Ma, S. & Kenis, P. J. A. Electrochemical conversion of CO₂ to useful chemicals: current status, remaining challenges, and future opportunities. *Curr. Opin. Chem. Eng.* 2, 191–199 (2013).
- Kortlever, R., Shen, J., Schouten, K. J. P., Calle-Vallejo, F. & Koper, M. T. M. Catalysts and reaction pathways for the electrochemical reduction of carbon dioxide. J. Phys. Chem. Lett. 6, 4073–4082 (2015).
- Lu, Q. & Jiao, F. Electrochemical CO₂ reduction: electrocatalyst, reaction mechanism, and process engineering. Nano Energy 29, 439–456 (2016).
- Martín, A. J., Larrazábal, G. O. & Pérez-Ramírez, J. Towards sustainable fuels and chemicals through the electrochemical reduction of CO₂: lessons from water electrolysis. *Green Chem.* 17, 5114–5130 (2015).
- 13. Nitopi, S. et al. Progress and perspectives of electrochemical $\rm CO_2$ reduction on copper in aqueous electrolyte. *Chem. Rev.* 119, 7610–7672 (2019).
- 14. Ma, S. et al. One-step electrosynthesis of ethylene and ethanol from CO₂ in an alkaline electrolyzer. J. Power Sources 301, 219–228 (2016).
- Gabardo, C. M. et al. Continuous carbon dioxide electroreduction to concentrated multi-carbon products using a membrane electrode assembly. *Joule* 3, 2777–2791 (2019).
- Wang, X. et al. Efficient electrically powered CO₂-to-ethanol via suppression of deoxygenation. Nat. Energy 5, 478–486 (2020).
- 17. Alibaba product search: formic acid 85%, category: organic acid, min order: 10 metric tons (Alibaba, 2021); https://www.alibaba.com/trade/search? IndexArea=product_en&SearchText=formic_acid_85%25&c=CID80310&f0=y&moqf=MOQF&moqt=MOQT10%20Metric%20Tons
- Ethylene Market Size Worth \$186.5 Billion by 2026 (Polaris, 2020); https://www.polarismarketresearch.com/press-releases/ethylene-market
- Formic Acid Market Anticipated to Reach Market Value of USD 878.7 Million at a CAGR of 4.94% during 2016 to 2027 (MarketResearchFuture, 2017); https://www.globenewswire.com/news-release/2017/09/08/1116865/0/en/ Formic-Acid-Market-Anticipated-to-Reach-Market-Value-of-USD-878-7-Million-at-a-CAGR-of-4-94-during-2016-to-2027.html

- 20. Ethanol Market Size Worth Around USD 155.6 Billion by 2030 (PrecedenceResearch, 2021); http://www.globenewswire.com/news-release/2021/01/18/2160198/0/en/Ethanol-Market-Size-Worth-Around-USD-155-6-Billion-by-2030.html#:~:text=The%20global%20ethanol%20market%20size, 5.2%25%20from%202021%20to%202030
- Global Carbon Monoxide Market 2020–2026, with Breakdown Data of Capacity, Sales, Revenue, Price, Cost and Gross Profit (ReportsNMarkets, 2020); https://www.reportsnmarkets.com/report/Global-Carbon-Monoxide-Market-2020-2026-With-Breakdown-Data-of-Capacity-Sales-Revenue-Price-C ost-and-Gross-Profit-60
- Sims, M. US May Ethylene Contracts Settle Up After Six-Month Decline (ICIS, 2020); https://www.icis.com/explore/resources/news/2020/06/02/10514594/us-may-ethylene-contracts-settle-up-after-six-month-decline
- 23. Annual Energy Outlook 2021: Table 12: Petroleum and Other Liquids Prices (EIA, 2020); https://www.eia.gov/outlooks/aeo/data/browser/#/?id=12-AEO20 21&cases=ref2021&sourcekey=0
- 24. Haegel, N. M. et al. Terawatt-scale photovoltaics: trajectories and challenges. *Science* **356**, 141–143 (2017).
- Levelized Cost and Levelized Avoided Cost of New Generation Resources (EIA, 2020); https://www.eia.gov/outlooks/aeo/pdf/electricity_generation.pdf
- Wiser, R. & Bolinger, M. 2016 Wind Technologies Market Report (US Department of Energy, 2016); https://www.energy.gov/sites/default/ files/2017/10/f37/2016_Wind_Technologies_Market_Report_101317.pdf
- Baldea, M., Edgar, T. F., Stanley, B. L. & Kiss, A. A. Modular manufacturing processes: status, challenges, and opportunities. AIChE J. 63, 4262–4272 (2017).
- Verma, S., Kim, B., Jhong, H. R. M., Ma, S. & Kenis, P. J. A. A gross-margin model for defining technoeconomic benchmarks in the electroreduction of CO₂. ChemSusChem 9, 1972–1979 (2016).
- Rumayor, M., Dominguez-Ramos, A., Perez, P. & Irabien, A. A techno-economic evaluation approach to the electrochemical reduction of CO₂ for formic acid manufacture. *J. CO2 Util.* 34, 490–499 (2019).
- Na, J. et al. General technoeconomic analysis for electrochemical coproduction coupling carbon dioxide reduction with organic oxidation. *Nat. Commun.* 10, 5193 (2019).
- 31. Verma, S., Lu, S. & Kenis, P. J. A. Co-electrolysis of $\rm CO_2$ and glycerol as a pathway to carbon chemicals with improved technoeconomics due to low electricity consumption. *Nat. Energy* 4, 466–474 (2019).
- Spurgeon, J. M. & Kumar, B. A comparative technoeconomic analysis of pathways for commercial electrochemical CO₂ reduction to liquid products. *Energy Environ. Sci.* 11, 1536–1551 (2018).
- 33. Jouny, M., Luc, W. & Jiao, F. General techno-economic analysis of $\rm CO_2$ electrolysis systems. *Ind. Eng. Chem. Res.* 57, 2165–2177 (2018).
- Jouny, M., Hutchings, G. S. & Jiao, F. Carbon monoxide electroreduction as an emerging platform for carbon utilization. *Nat. Catal.* 2, 1062–1070 (2019).
- De Luna, P. et al. What would it take for renewably powered electrosynthesis to displace petrochemical processes? Science 364, eaav3506 (2019).
- Orella, M. J., Brown, S. M., Leonard, M. E., Román-Leshkov, Y. & Brushett, F. R. A general technoeconomic model for evaluating emerging electrolytic processes. *Energy Technol.* 8, 1900994 (2019).
- Wang, N. et al. Hydration-effect-promoting Ni–Fe oxyhydroxide catalysts for neutral water oxidation. Adv. Mater. 32, 1906806 (2020).
- Smith, A. M., Trotochaud, L., Burke, M. S. & Boettcher, S. W. Contributions to activity enhancement via Fe incorporation in Ni-(oxy)hydroxide/borate catalysts for near-neutral pH oxygen evolution. *Chem. Commun.* 51, 5261–5263 (2015).
- Salvatore, D. A. et al. Designing anion exchange membranes for CO₂ electrolysers. Nat. Energy 6, 339–348 (2021).
- Houache, M. S. E. et al. Selective electrooxidation of glycerol to formic acid over carbon supported Ni_{1-x}M_x (M = Bi, Pd, and Au) nanocatalysts and coelectrolysis of CO₂. ACS Appl. Energy Mater. 3, 8725–8738 (2020).
- Landress, L. Outlook '19: US Glycerine Markets Mixed Amid Uncertainty (ICIS, 2019); https://www.icis.com/explore/resources/news/2019/01/07/ 10301259/outlook-19-us-glycerine-markets-mixed-amid-uncertainty/
- 42. The SunShot 2030 Goals: 3¢ Per Kilowatt Hour for PV and 5¢ Per Kilowatt Hour for Dispatchable CSP (US Department of Energy, 2017); https://www.energy.gov/sites/prod/files/2020/09/f79/SunShot%202030%20White%20Paper.pdf
- Edwards, J. P. et al. Efficient electrocatalytic conversion of carbon dioxide in a low-resistance pressurized alkaline electrolyzer. Appl. Energy 261, 114305 (2020).
- 44. García de Arquer, F. P. et al. CO₂ electrolysis to multicarbon products at activities greater than 1 A cm⁻². Science 367, 661–666 (2020).
- Rabinowitz, J. A. & Kanan, M. W. The future of low-temperature carbon dioxide electrolysis depends on solving one basic problem. *Nat. Commun.* 11, 5231 (2020).
- 46. Ma, M. et al. Insights into the carbon balance for CO₂ electroreduction on Cu using gas diffusion electrode reactor designs. *Energy Environ. Sci.* 13, 977–985 (2020).
- Endrődi, B. et al. High carbonate ion conductance of a robust PiperION membrane allows industrial current density and conversion in a zero-gap carbon dioxide electrolyzer cell. Energy Environ. Sci. 13, 4098–4105 (2020).

- 48. Electricity Generation in Germany in January 2020 (Energy-Charts, 2020); https://energy-charts.info/charts/power/chart.htm?l=en&c=DE
- Küngas, R. et al. Progress in SOEC development activities at Haldor Topsøe. ECS Trans. 91, 215–223 (2019).
- 50. The Tax Credit for Carbon Sequestration (Section 45Q) (Congressional Research Service, 2020); https://fas.org/sgp/crs/misc/IF11455.pdf
- FY 2018 Progress Report for the DOE Hydrogen and Fuel Cells Program (US Department of Energy, 2019); https://www.nrel.gov/docs/fy19osti/73353.pdf
- Zhong, M. et al. Accelerated discovery of CO₂ electrocatalysts using active machine learning. *Nature* 581, 178–183 (2020).
- 53. Li, T. et al. Electrolytic conversion of bicarbonate into CO in a flow cell. *Joule* 3, 1487–1497 (2019).
- 54. Salvatore, D. & Berlinguette, C. P. Voltage matters when reducing CO₂ in an electrochemical flow cell. *ACS Energy Lett.* **5**, 215–220 (2020).
- Sun, C. N. et al. Probing electrode losses in all-vanadium redox flow batteries with impedance spectroscopy. ECS Electrochem. Lett. 2, 2013–2015 (2013).
- Heinzmann, M., Weber, A. & Ivers-Tiffée, E. Advanced impedance study of polymer electrolyte membrane single cells by means of distribution of relaxation times. *J. Power Sources* 402, 24–33 (2018).
- Xu, Q. et al. Integrated reference electrodes in anion-exchange-membrane electrolyzers: impact of stainless-steel gas-diffusion layers and internal mechanical pressure. ACS Energy Lett. 6, 305–312 (2020).
- Nwabara, U. O. et al. Toward accelerated durability testing protocols for CO₂ electrolysis. *J. Mater. Chem. A* 8, 22557–22571 (2020).
- He, M. et al. Oxygen induced promotion of electrochemical reduction of CO₂ via co-electrolysis. Nat. Commun. 11, 3844 (2020).
- Chae, S. Y. et al. A perspective on practical solar to carbon monoxide production devices with economic evaluation. Sustain. Energy Fuels 4, 199–212 (2020).

61. Larrazábal, G. O. et al. Analysis of mass flows and membrane cross-over in CO₂ reduction at high current densities in an MEA-type electrolyzer. ACS Appl. Mater. Interfaces 11, 41281–41288 (2019).

Acknowledgements

This material is based upon work supported by the US Department of Energy under award number DE-FE0031910. We thank the National Science Foundation for financially supporting H.S. (award no. CBET-1803200). We also acknowledge helpful discussions on developing the methodology of the analysis by M. Jouny, and constructive suggestions by S. Overa and B. H. Ko.

Author contributions

H.S. and K.U.H. contributed equally to this work. H.S., K.U.H. and F.J. performed data analysis and wrote the manuscript. F.J. supervised the whole project.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41893-021-00739-x.

Correspondence and requests for materials should be addressed to E.J.

Peer review information Nature Sustainability thanks Ung Lee and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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