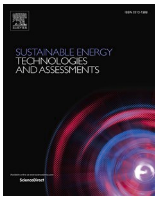
Sustainable Energy Technologies and Assessments 61 (2024) 103574

Contents lists available at ScienceDirect

Sustainable Energy Technologies and Assessments

journal homepage: www.elsevier.com/locate/seta



A systematic life cycle assessment of the electroconversion of carbon dioxide

Scott J. Blazer [a,b,c], Yudong Wang [b,c,d], Nengneng Xu [a,c], Xiao-Dong Zhou [a,b,c,d,*], Barbara Marchetti [e,*]

[a] *Department of Chemical & Biomolecular Engineering, University of Connecticut, Storrs, CT 06269, United States*
[b] *Department of Chemical Engineering, University of Louisiana at Lafayette, Lafayette, LA 70503, United States*
[c] *Center for Clean Energy Engineering, University of Connecticut, Storrs, CT 06269, United States*
[d] *Department of Mechanical Engineering, University of Connecticut, Storrs, CT 06269, United States*
[e] *Department of Chemistry, University of Louisiana at Lafayette, Lafayette, LA 70504, United States*

ARTICLE INFO

Keywords:
Carbon utilization process
Environmental analysis
Life cycle assessment
Climate Change Mitigation

ABSTRACT

In this study, we report a cradle-to-gate life cycle assessment of assembling and operating an electrochemical cell for conversion of carbon dioxide (CO_2) to formic acid. This process is followed by product separation *via* liquid–liquid extraction. The results suggest that cell operation yields the highest environmental impact *cf.* other processes. Parametric studies were performed to identify conditions that minimize the environmental impact. It was found that:

(i) the stability of cell components, durability of cell performance, and cell operating parameters (e.g., current density or cell voltage) play a pivotal role on the carbon emissions;
(ii) the optimal parameters include stable operation for at least 4,000 h at (ultrahigh) current densities (0.50–1.00 A cm^{-2});
(iii) through use of renewable energy sources zero carbon emissions may be achieved only if high cell performance conditions are met; and.
(iv) the cumulative carbon emissions were predicted during the entire life cycle of the system (4,000 h), while modelling cell aging and corresponding decrease in performance. Here, the use of renewable energy is of outmost importance to achieve climate change mitigation.

Introduction

Over the past few decades, an increasing anthropogenic emission of greenhouse gases (GHGs), especially CO_2, has led to profound consequences for both human well-being and the environment [1–4] (*v.* Section S1 of Supplementary Information). Such negative ramifications have driven much of the current research efforts towards the development of technologies to reduce GHG emissions and mitigate their environmental consequences. Among various approaches, electrocatalytic reduction of CO_2 (CO_2ER) represents a promising route to promote conversion of CO_2 to useful chemicals [5–13], which, coupled with the use of electricity from renewable energy providers, could represent a green approach to utilize previously captured CO_2. Despite this, construction of electrochemical cells for CO_2ER and their operation are complex processes, which may require sophisticated synthetic procedures or use of methods and technologies which may themselves come with a non-zero environmental impact. Clearly, environmental assessment of CO_2 utilization technologies requires further exploration, as only a handful of studies has been reported so far [14–28]. Previous studies have successfully implemented a life cycle assessment (LCA)

Abbreviations: EPA, Environmental Protection Agency; GHG, Greenhouse Gas; CO2ER, Carbon Dioxide Electrochemical Reduction; LCA, Life Cycle Assessment; LCIA, Life Cycle Inventory Analysis; FA, Formic Acid; CCU, Carbon Capture and Utilization; NETL, National Energy Technology Laboratory; GWP100, Global Warming Potential (100 years); Bi-MOF, bismuth-based metal-organic framework; PG, Power Grid.

* Corresponding authors.
E-mail addresses: xiao-dong.zhou@uconn.edu (X.-D. Zhou), barbara.marchetti1@louisiana.edu (B. Marchetti).

https://doi.org/10.1016/j.seta.2023.103574
Received 28 August 2023; Received in revised form 27 October 2023; Accepted 25 November 2023

approach to evaluate the environmental benefits of using CO_2ER technologies to produce formate (or formic acid), or other relevant products [15–26]. Typically, the formation of CO_2ER products is main focus in such studies, however, upstream processes and downstream product separation need accounting as they may require considerable energy consumption [17,29]. A recent study by Nabil *et al.* [17] reports a comparative LCA which analyses the conversion of CO_2 to an array of eight different products, considering both their generation and separation to obtain high purity chemicals. In their assessment, most of the environmental impact was related to the energy requirement, which was majorly attributed to the conversion of CO_2 to such products and their separation processes. Interestingly, the global warming impact was found to depend on the CO_2 conversion product.

To the best of our knowledge, there is still a lack of comprehensive, systematic LCA studies aimed at evaluating the environmental effect of tuning various experimental and operation parameters (*v.* Table S1 in Section S2 for survey of previous reports). The interplay between manufacturing and operation of CO_2ER technologies also requires further exploration. In particular, it is paramount to investigate how optimization of energy-to-product conversion, stability and durability of CO_2ER technologies can aid reducing the net carbon emissions, since these factors are potentially highly impactful under health and environmental perspectives. Previous reports have investigated the production of various useful chemicals from CO_2ER, while also including some sensitivity analysis or the effect of the energy source to some extent [14–28]. However, since different frameworks and methods may yield to substantially different results, thus making comparison between studies rather challenging. Thus, it is paramount to provide a unifying, comprehensive study, in which these different aspects are explored using the same approach and consistent methodology. Furthermore, evaluation of manufacturing procedures should be as comprehensive as possible.

For this purpose, a detailed cradle-to-gate LCA was carried out. Firstly, a comprehensive assessment was used to investigate the impact of CO_2ER technologies using a baseline model. We focused on evaluating resource and energy requirements for CO_2ER and all related energy-intensive upstream and downstream processes. As such, the aim is to evaluate the environmental impact of the entire electrochemical cell lifetime, including manufacturing or synthesis processes, cell operation and separation procedures to obtain a commercial grade product, here taken as formic acid (FA). The choice of the product is justified by various motivations, including its numerous applications in many industrial processes and potential of being used as hydrogen carrier and for fuel cell applications. Furthermore, compared to other CO_2ER products, FA has a high molar mass per electron transfer, which is energetically preferable for production by CO_2ER [17]. Secondly, an important objective of this LCA is to complement the existing reports by carefully evaluating various factors, *i.e.*, operational variables, which may affect most heavily the outcome of the analysis. Therefore, a parametric analysis was performed on the model; the overarching goal is to identify the set of conditions which offer a good match and/or compromise between product output, energy requirements, and minimal environmental impact. Finally, we performed an overall evaluation of the cumulative global warming impact over the whole lifespan of the cell, by factoring the decline in energy efficiency as the electrochemical device ages. The results of this LCA study are expected to provide valuable guidance in the development of CO_2ER technologies, especially regarding ways in which, not only their performance, but also the environmental benefits of their implementation can be maximized. This will provide insights to favour the transition to more sustainable, carbon neutral technology.

Methodology and framework

The methodology, mathematical relationships, and life cycle inventory for this study are described in detail in Sections S3 and S4 of the Supplementary Information, alongside the goal and scope of the study. Briefly, the LCA was performed using the openLCA software in conjunction with the freely available databases. [30,31] The Tool for the Reduction and Assessment of Chemical and other environmental Impacts, [32–34] (TRACI 2.1) was selected as Life Cycle Inventory Analysis (LCIA) method (*v.* Section S5.1). Results of the analysis are tabulated in Section S6.

The goal of the study is to evaluate the environmental impact of manufacturing and operating an electrochemical reactor for CO_2ER and identify the optimal circumstances under which the impact can be minimized. To this end, we consider the interplay between upstream manufacturing processes and the operation of the CO_2ER technology. We include an extremely extensive inventory to ensure that the description of manufacturing processes is as accurate as possible. The upstream processes will then be linked to the main process of interest for the model system, e.g., operation of the reactor. An additional downstream process is added for product purification, to obtain commercial grade FA, e.g., at least 85 % purity of the product in its acidic form, rather than FA or its conjugate base (formate) in dilute aqueous solution. Details on the operation and purification unit processes are reported in Section S4.2 Operation processes of the Supporting Information.

Results and discussion

In the following sections, the LCIA results and their interpretation will be discussed. We will focus our attention on the GWP100 since provides a good tool for estimating the GHG emissions and is therefore particularly relevant to this study. For simplicity, we will often refer to the GWP100 as GHG emissions in the text. All results will be referenced to the functional unit of 1 kg of FA produced upon CO_2ER. A schematic version of the model system is illustrated in Fig. 1, which shows the most relevant upstream processes linked to the process of interest, (operation of the reactor). The product purification ('formic acid separation') follows it. Additional schematics are shown in Figures S1 and S2.

The power grid (PG) (v. Section S3) is displayed explicitly as connected to a subset of unit processes. This PG has a carbon intensity of about 0.467 kg of CO_2e per kWh of electricity produced. This value may be relatively large compared to the carbon emissions of electricity produced in many US states or European countries. It can however be

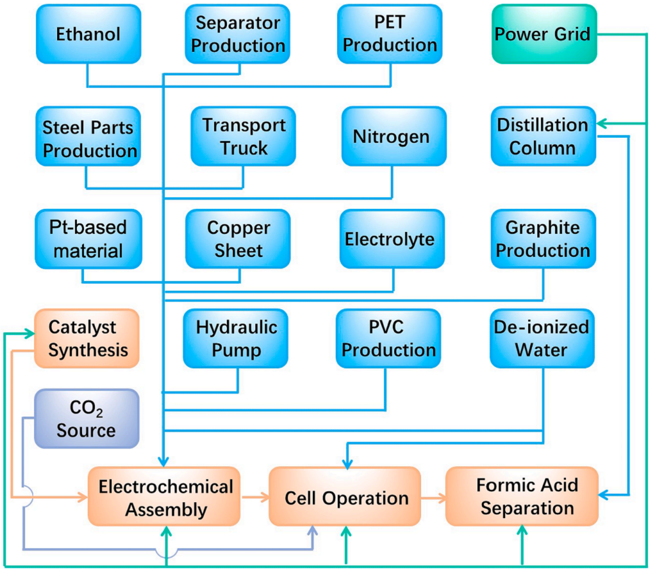


Fig. 1. Collapsed model graph of the model system for CO_2ER. The Federal Commons LCA data repository [33] and the National Energy Technology Laboratory database for carbon capture and utilization [34] were used to support the implementation of the model.

considered more representative of the average carbon emissions from electricity worldwide, as of 2019. [35] In Section 4.5, we provide a Country-specific scenario of the global warming impact associated with the technology. Other processes in Fig. 1 also require energy providers but, for conciseness and clarity, these are not explicitly displayed.

Baseline model

For the first assessment, the results were computed by setting operational parameters as explained in Section S4.2, *i.e.*, 4,000 h cell operation time at 1.00 A cm^{-2} (at 5.48 V), [36] and 255 g/hour recovery of commercial grade FA upon liquid–liquid extraction. [37] A general analysis of the environmental impact for various categories is reported in Section S5.2.

GHG Emissions. The analysis of GWP100 predicts ~ 5.5 kg of CO_2e for 1 kg of FA produced (0.861 kg of CO_2 utilized). Hence, positive, net emissions of GHGs are predicted when a generic electricity provider is used. More detailed impact results are reported in Table S15 and Fig. 2, which illustrate the absolute and percentage contributions to the GWP100 of the various processes.

The energy consumption for cell operation and product separation account for most of the GHG emissions. Manufacturing of the electrochemical and extraction/distillation assemblies accounts collectively for 5.09 % of the overall GHG emissions, with manufacturing the reactor only accounting for 1.55 %. These findings are in qualitative agreement with previous reports of CO_2ER and other energy-related technologies [15–17,38–40]. However, this study predicts higher GHG emissions *cf.* previous literature, likely due to the higher carbon intensity of the electricity and/or more extensive inventories of materials. Using a more comprehensive inventory of materials has the purpose of providing the most realistic as possible description of the manufacturing processes and thus the associated environmental impact, even if such processes are predicted to be less impactful *cf.* cell operation. When considering the electricity grid, this study uses a medium–high carbon intensity to provide a modern representation of the GHG emissions at the present time. For instance, Nabil *et al.* report an LCA for a similar technology using an electricity emission intensity of ~ 0.17 kg of CO_2e/kWh (average carbon intensity in Canada) [17], *i.e.*, nearly 3-fold lower than the one used in the present study (0.467 kg of CO_2e/kWh). We envisage that the selection of a more carbon intense power grid helps assessing the global warming impact of the technology even in non-optimal cases, *i.e.*, when the electricity used is not reliant on large shares of clean renewable energy sources.

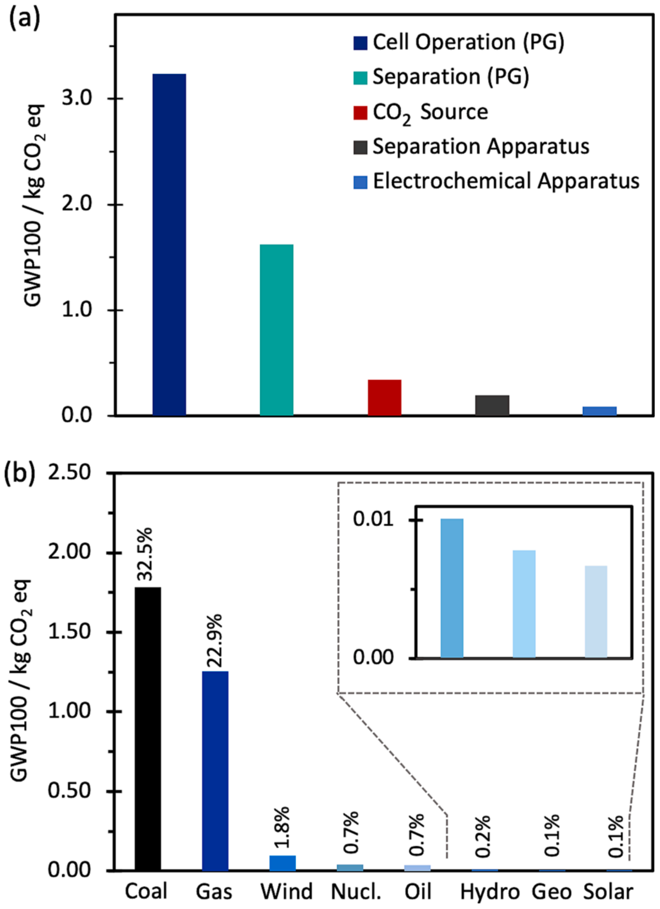


Fig. 2. GWP100 impact results. (a) contribution of various processes, (b) Contribution of various power generation sources. The inset shows an enlarged view of the lowest contributions.

If we break down the impact of cell operation (PG) in various energy sources (Fig. 2(b)), the highest emissions arise from coal-based power generation (32.5 %) although it accounts only for 22 % of the total electricity produced. Clearly, decreasing the fraction of coal-based power, at least for cell operation, can help reduce GHG emissions by up to a third of the overall value. This is a clear-cut illustration of the importance of using clean electricity at least for operation, as a first step towards lower carbon emitting technologies.

The following sections will present exploratory studies aimed at identifying ways in which these emissions can be minimized. This will be done by scanning selected operation parameters to ensure minimal environmental impact of implementing the CO_2ER technology.

To identify the parameters of highest environmental impact, a parametric study was performed on the model system. Details of the analysis are included in Section S5.2 of the Supporting Information, which also reports a general description of the impact for various categories. In the text below, we will focus on how varying different parameters affects GHG emissions.

Parametric analysis

Stability and durability

Overview. The stability and durability of the electrochemical cell represent important factors which can affect both its economic and environmental feasibility. [16,41] Thus, in this study, we explore the implications that varying the cell lifetime has on the environment and global climate impact. To this end, the overall operation time of the cell was set to discrete values between 24 and 40,000 h; subsequently, assessment of environmental impact was performed at the various points. To first approximation, all other parameters are kept constant: in the last section of this study, we will assess how decline of cell performance throughout its lifetime can affect environmental impact.

GHG emissions. The overall GWP100 as a function of the operation time is shown in Fig. 3(a). The GHG emissions are predicted to decrease sharply with the increase of cell lifetime. These results are broken down into different contributions, arising from various processes in Fig. 3(a) and 3(b). Manufacturing the two assemblies accounts for over 80 % of the overall GHG emissions at limited cell lifetimes. Particularly, assembling the extraction/distillation apparatus is associated by far with the highest impact (Fig. 3(b)). As our quantitative reference is the production of 1 kg of FA, when the operation time increases, contributions of manufacturing the assemblies decreases at the same time, until becoming negligible at 40,000 h of operation. Consequently, the impact associated with cell operation and product separation (energy consumption) becomes dominant at longer lifetime (>250 h). This is true since the faradaic efficiency, current density and other parameters are not changed. The GHG emissions approximately plateau around 4,000 h; further extension of the cell lifetime is not expected to greatly affect the impact.

Therefore, we reckon that 4,000 h (or longer) of operation at the set

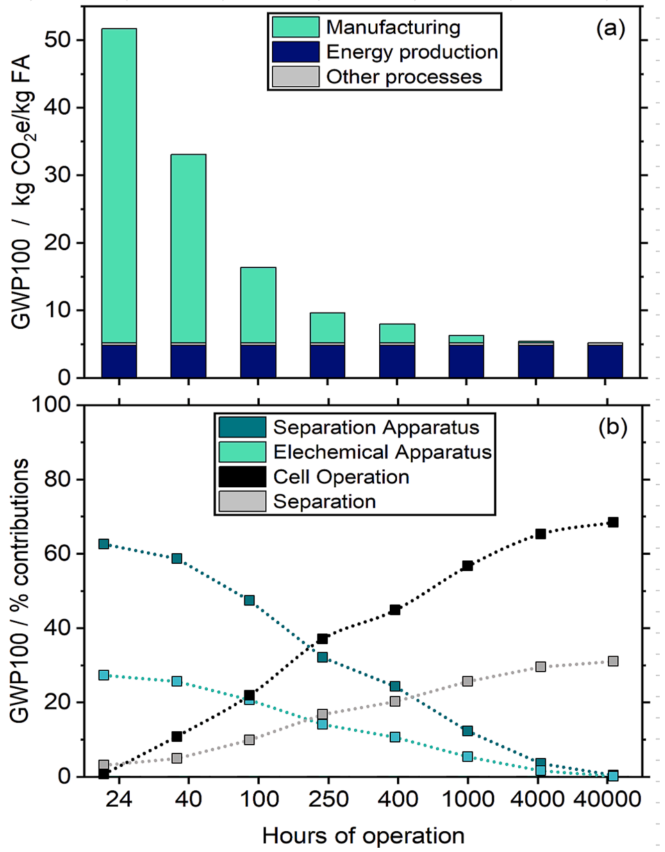


Fig. 3. (a) Carbon emissions at different operation time; the contributions of various processes are shown, (b) percentage contributions of various processes.

conditions could be considered an appropriate time to amortize the environmental cost of manufacturing and will be set as the ‘target’ durability for the following investigations. This would provide lower, although not negligible global warming impact. This finding is in only qualitative agreement with a previous study by Rumayor et al. [16], wherein the authors predicted a desired lifetime of cathode materials of 210 h for amortizing the environmental effect of cathode fabrication. The discrepancy with the current results may arise from the use of a more extensive inventory, consisting of all major components to construct both the reactor and separation apparatus.

In general, durability of a technology is always an important factor to limit the impact of manufacturing processes. However, the dependence is process-specific and, for CO$_2$ER technologies is likely product-specific. For products of CO$_2$ER with lower molar mass per electronic transfer (e.g., ethylene), energy requirements for CO$_2$ER may dominate the impact even at short lifetimes; manufacturing processes would then be comparatively less urgent issues, *c.f.* optimization of operation parameters (e.g., cell voltage/resistance, faradaic and energy efficiency, etc.).

Current density and cell voltage

The effect of varying the current density j (and associated voltage V) is explored by setting this variable to discrete values between 0.25 and 1.50 A cm^{-2}, while maintaining the other parameters unvaried. The energy consumption required to perform CO$_2$ER at various j values (and V) was calculated in accordance with previous studies on analogous electrochemical devices [36] (see Section S4.2). The energy for ancillary equipment was also included but was set to exclusively depend on the operation time (4,000 h). The product is assumed to be collected and separated by liquid–liquid extraction. The rate of recovery of the final product was not changed. The following analysis will assume that the faradaic efficiency does not change in this (wide) range of current densities.

GHG emissions. The GWP100 results as a function of j are shown in Fig. 4. An initial, modest decrease in the GHG emissions is observed with increasing j (up to 0.75 A cm^{-2}), followed by a small, but steady increase. The initial decrease is attributed primarily to lower contributions arising from manufacturing the assemblies, as seen in both Fig. 4(a) and 4(b). However, this effect is outrun by the larger demand in energy for CO$_2$ER at higher current densities, wherein operation of the cell becomes dominant. By contrast, since the separation of the product is assumed to occur at the same rate, the associated (absolute) contribution stays constant. Under the environmental perspective, it is therefore inferred that current densities of 0.50–1.00 A/cm^2 may be favourable and sufficient to (i) render manufacturing processes negligeable and (ii) limit the environmental impact of energy consumption. Aiming for the higher end of this range could represent a good trade-off between lowering GHG emissions and obtaining good product output. The latter is necessary for larger-scale applications, and techno-economic analysis also suggests that current densities exceeding 1.00 A cm^{-2} (lifetimes > 30,000 h) are required for industrial deployment of electrolysis technologies. [42] When generic PG is considered, very high j values do not seem compatible with efforts aimed at minimizing the environmental impact. Furthermore, operation at high j may have an impact on the stability/durability of the cell materials. In this context, lower j may be advisable for decreasing material degradation, thus extending the cell

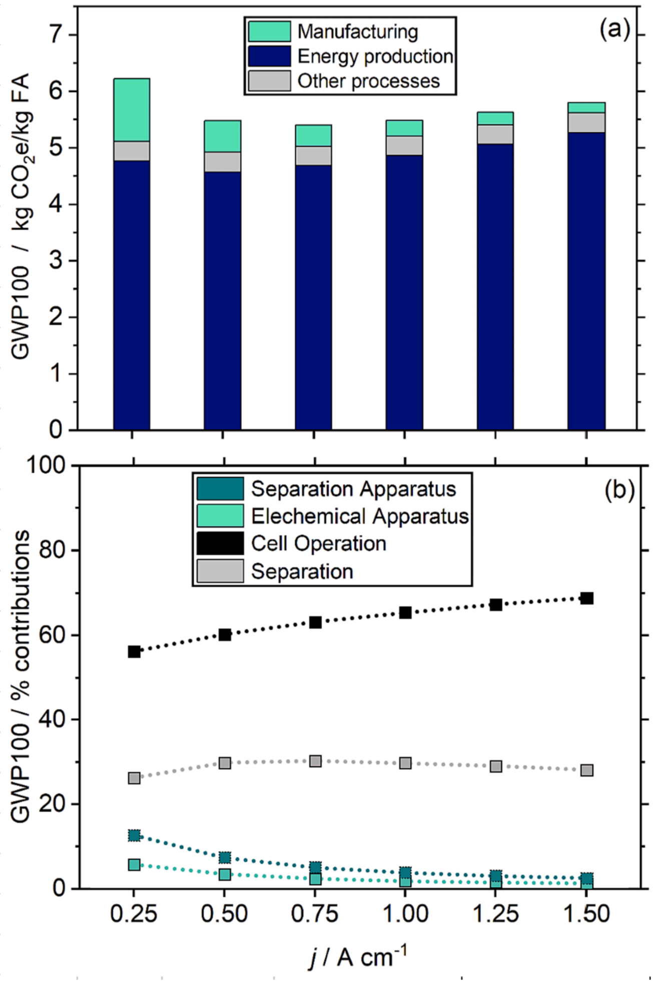


Fig. 4. Carbon emissions at different current densities; the contributions of various processes are shown, (b) percentage contributions of different processes. Recent experiments were used to model how the energy requirements for CO$_2$ER change upon varying j [36].

lifetime.

Finally, the dependence of the GHG emissions on *j* is related to the cell performance and energy efficiency. Here, we refer to a recent publication [36] to model how the energy requirements for CO_2ER change upon varying *j*, e.g., this part was evaluated based on available experimental data. The details in the trend and associated GHG emissions may vary to some extent depending on the experimental data used for the model. However, it is reasonable to assume that, at higher current densities, energy efficiency tends to decrease (*v.* Section 8). It can then be inferred that, despite higher products output, the global climate impact is bound to increase (*v.* Fig. 4 and Figure S3). It is our understanding that a dependence on the product of CO_2ER may also be observed due to possibly different molar masses per electron transfer. In these cases, the interplay between product output and energy requirements may be different.

Role of renewable energies sources

The results presented in the previous sections show that the energy usage, especially for CO_2ER, is the dominant factor affecting the environmental feasibility of the CO_2ER technologies. We investigated the role that renewable energy sources may play in favouring transition to lower carbon-emitting technologies. Thus, we selected five different electricity providers within the NETL CO2U Database version 2.0 [31], e.g., (adjusted) PG and Nuclear electricity as non-renewable providers, Solar Thermal, Hydrothermal and Wind as renewable sources. The operation time and current densities were varied in turns and the results computed at each point.

GHG emissions. Fig. 5(a) shows a comparison between the GWP100 estimated for the five electricity sources at three current densities (0.25, 0.50 and 1.00 A cm^{-2}) and 4,000 h of operation. The dashed horizontal line represents the mass of CO_2 consumed per production 1 kg of FA (0.861 kg), that is, the threshold at which zero carbon emissions are achieved. The use of PG results in 2–5 times higher GHG emissions *cf.* the other energy sources, some which return (close to) zero GHG emissions at 1.00 A cm^{-2}.

Fig. 5(b) shows the estimated GHG emissions as a function of varying both *j* and durability of the cell for a renewable provider. The emissions consistently decrease with both higher *j* and longer cell lifetime. Our predictions suggests that very long lifetimes, exceeding 15,000 h, are required to achieve zero or negative GHGs emissions at current densities equal or lower than 0.25 A cm^{-2}, while 3,000–4,000 h are requiring at higher *j* values. Thus. the renewable source shows a different trend of GHG emissions with *j* (*c.f.* the generic PG), because the impact of manufacturing decreases higher *j* values, and, at the same time, higher energy consumption is counterbalanced by higher yields of FA. Thus, operation at ultrahigh *j* yields higher environmental benefits. This implies that clean energy favours minimum environmental impact at conditions which are more compatible with those invoked by technoeconomic analysis [42]. For their applicability, this also means that stability and durability of the cell materials becomes a less environmentally compelling issues when compared to the generic PG.

In conclusions, the results in Fig. 5 confirm that use of clean energy sources [15,43], could represent a viable route to favour eco-compatibility of electrolysis and/or carbon utilization technologies, while also maximizing the use of intermittent renewable energy sources [15,23,43,44].

Cumulative life cycle emissions

The primary aim of this section is to explore how the life cycle GWP100 varies with the decrease efficiency as a function of the operation time. A higher operation voltage, owing to the degraded electrochemical cell, leads to the lower energy efficiency for constant current or constant FA production. In this framework, we set the output of FA to be constant, while the energy consumption is increasing with increasing lifetime. Details about the model and background are reported in Section S.6 and S.8. Briefly, we perform the assessment at 100-hour increments (4,000 h in total). The functional unit was not restricted to 1 kg of FA, rather, the total mass of FA at each increment was used; consumption of CO_2 and GHG emissions were compared.

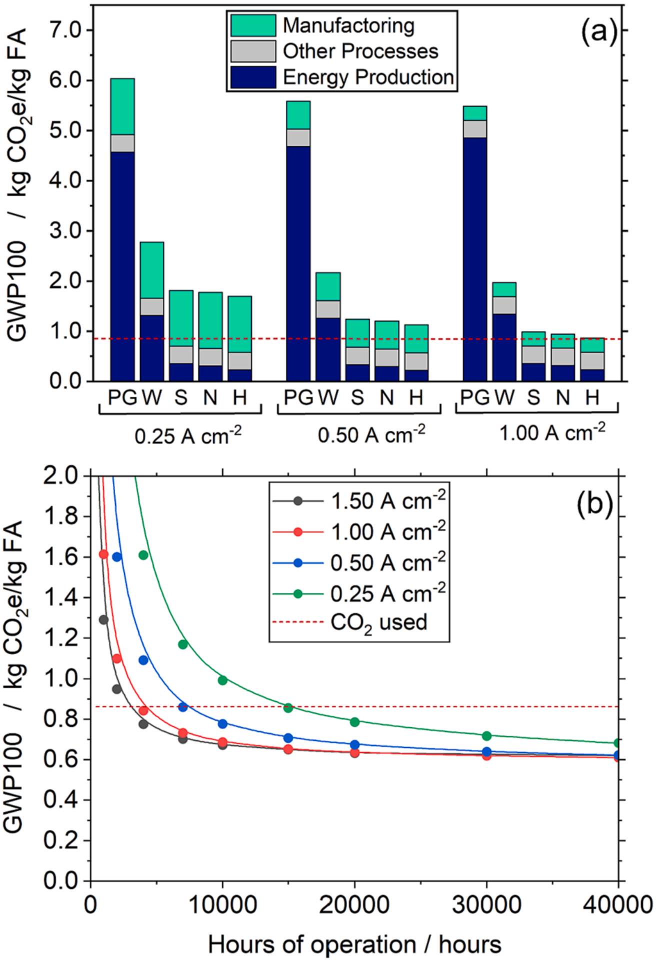


Fig. 5. GHG missions for various energy sources at three different current densities. (b) GHG emissions as a function of current density and lifetime when the hydroelectric energy provider is used for cell operation and product separation. Horizontal dashed line represents the net 0 emission threshold.

GHG Emissions. Fig. 6 illustrates the incremental and cumulative GHG emissions when the PG (Fig. 6(a) and 6(b)), and hydrothermal provider (Fig. 6(c) and 6(d)) are used for operation and product separation. Contributions of preparation of the set-ups are accounted once and are distributed throughout the entire lifetime of the cell.

In Fig. 6(a) and 6(b), the GHG emissions undergo a steep increase after 3,000 h of operation, due to sharp increase in energy consumption. Here, the GHG emissions overwhelmingly outweigh the mass of CO_2 utilized, thus retuning net positive emissions at all times. When using hydropower (Fig. 6(c)), the GHG emissions are much lower than the CO_2 consumption (e.g., 7.4 kg of CO_2e emitted *vs.* 8.3 CO_2 consumed in the first 100 h of operation), due to the selection of the clean, renewable energy source for cell operation and product separation. Only at lifetimes longer than 3,000 h the environmental benefits are largely

Fig. 6. (a) GWP100 results and contributions of different processes at 100-hour time intervals up to 4,000 h, (b) cumulative GWP100, and mass of CO_2 converted a at the above conditions. The electricity provider is a generic power grid representative of the US modern grid.

outweighed by the energy consumption. However, the cumulative GHG emissions (Fig. 6(d)) always remain negative or close to zero. After 4,000 h, the cumulative GWP100 is calculated to be of ~ 334 kg of CO_2e being emitted, per ~ 330 kg of CO_2 converted, suggesting, again, that renewable energy for CO_2ER, coupled to CO_2 capture, can help reducing the global warming impact by a considerable amount, while at the same time obtaining a valuable product.

Further considerations

As a final consideration, we show three possible case scenarios for our proposed model system. Case 1 is modelled in accord with a recent study exploring the stability and durability of electrolysers for CO_2-to-FA conversion [36], which we will take as a plausible scenario for state-of-the-art electrolysers developed in laboratory settings. While no additional process was added to the model, the product yield was here rescaled to match Faradaic Efficiency of ~75 % across 1,000 h operation at 0.25 A cm^{-2}. The results represent a lower limit for the GHG emissions, as in complete model, separation of by-products might also require accounting. Case 2 represents a desirable scenario under the environmental perspective, while case 3 is analysed by setting parameters to optimistic values for large scale deployment according to recent techno-economic assessments (highest performance, stability, and durability) [42]. The parameters are specified in Fig. 7(a), alongside the associated results. Estimating uncertainties for our results is challenging as it requires accounting of possible uncertainty arising from a vast number of measurements as well as possible deviations due to assumptions and approximations made in the model. However, we may consider the results in Fig. 7(a) as upper and lower limits for GHG emissions in three possible case scenarios; in this instance, the associated GHG emissions show a deviation of about 1.1 kg of CO_2e/kg of FA considering the three values (v. Section S5.1) for the PG, or 0.7 kg of CO_2e/kg of FA for hydropower. Interestingly, when using the generic PG lower, the desired conditions for industrial deployment estimated upon techno-economic analysis do not match the optimal conditions for least GHG emitting scenario. This occurs only when a clean energy source (hydropower) is selected for operation of the cell and separation of the final product. In this model, uncertainties related to GHG emissions from energy production/usage may probably come with the largest uncertainty when compared to other processes.

Furthermore, due to the heavy contribution of power generation, the carbon intensity of the electricity used for operation deserves consideration, since, for set operation parameters, it can affect the GHG emissions to colossal extent. In this context, for instance, the carbon intensity of electricity in European countries as of 2021 ranged from 946 to 9 kg of CO_2e per kWh [45]. As such, when Case 2 is considered, the associated distribution of GHG emissions would span a range between 11.2 and 0.6 kg of CO_2e/kg of FA (Fig. 7(b)), depending on the geographical location (as well as the time) which is considered for the analysis. In Fig. 7(b), we can see that the model system returns net negative emissions in Sweden (below red dashed line), where electricity comes primarily from renewable sources. The average US grid will place emissions in the US in between Ireland and Romania, still below the GHG emissions modelled in this study (grey dashed line).

We did not model any alternative pathways for production of FA, however, LCA of thermochemical methods for FA production have been reported in previous studies [46,47], which primarily focus on energetic requirements for the process. The light blue shaded areas in Fig. 7 show the approximate ranges of GHG emissions evaluated for thermochemical methods (without subtracting for CO_2 consumption) according to such previous studies. From this comparison, we may conclude that production of FA by CO_2ER may be a comparably clean process when low carbon emitting electricity is used for cell operation and product separation.

Fig. 7. (a) comparison between the GWP100 for Case 1–3 (from left to right), (b) predicted GWP100 for Case 2 in various European Countries.

Conclusions and outlook

We investigated the life-cycle environmental impact of manufacturing and operating an electrochemical cell for reduction of previously captured CO_2 to FA. Purification of the product by liquid–liquid extraction was considered. In many instances, cell operation and product separation were associated with the highest GHG emissions *cf.* other processes. This is attributed due to the high energy consumption required to perform these processes. Parametric studies were performed to evaluate the conditions and parameters which mostly affect the impact results. Furthermore, with the addition of the production of the cell to the life cycle analysis, the optimum minimum operation time can be evaluated. Accordingly, some considerations on the implementation of CO_2ER technologies should be made, as it follows:

(1) The use of clean energy sources is truly essential for cutting down on the emissions of GHGs. However, contrarily to the common conception, clean energy sources cannot be considered *a priori* the solution to obtain net zero or negative emissions. Our studies show that high cell performance, durability and high product yields are still crucial for achieving low GHG emissions, even when clean energy is used for operation processes.

(2) Durability and stability of the electrochemical cell should be a concerning issue when considering the GHG emissions, especially if non-renewable electricity is used. Stability and longevity of the cell assure that environmental impact of manufacturing processes is minimized. In such cases, performing routine operation (CO_2ER and product separation) can be considered the primary cause of environmental impact and optimization of performance can then promote meaningful reduction of GHG emissions. In our study, minimal impact is predicted at cell lifetime equal or longer than 4,000 h.

(3) The current density and cell voltage govern the interplay between energy consumption and reaction rate. Clearly, low voltages/energy consumption are desired for low environmental impact, while ultrahigh current densities are imperative for large-scale deployment of CO_2ER technologies, according to technoeconomic assessments. For the cell modelled here, we found here that current densities of 0.50–1.00 A cm^{-2} are appropriate for minimizing GHG emissions when the generic PG is used. Using a clean energy source may allow environmental benefits even at higher current densities/energy consumption, such as those desired are useful for industrial application, if durability, and faradaic efficient remain suitable across the whole lifespan of the electrochemical cell.

(4) Recent theoretical studies of CO_2 utilization or comparable energy technologies have employed sophisticated statistical or modelling approaches, some of which rely on artificial intelligence and machine learning, to evaluate performance and optimize large number of operational parameters. Such sophisticated models are largely employed to explore and rationalize the relationships between operation conditions and electrochemical properties of electrolysers or batteries [48–50]. Such methodology has already been applied to LCA for selected applications [51,52]. Implementation and use of such approaches could assist exploration of electrolysers and energy storage technologies, by providing a mean to optimize large number of parameters in time-effective manner and paving the way to more accurate environmental assessment of energy technologies.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

We would like to thank the Department of Energy for the support of this work by the Office of Fossil Energy and Carbon Management under DE-FE0031919 and the Advanced Manufacturing Office of the Office of Energy Efficiency and Renewable Energy under DE-EE0009421, and National Science Foundation under NSF-2119688.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.seta.2023.103574.

References

[1] EPA, *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2020. U.S. Environmental Protection Agency, EPA 430-R-22-003*, 2022.

[2] IPCC, *Summary for Policymakers. In: Climate Change 2007. The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, [S. Solomon, D. Qin, M. Manning, Z. Cheng, M. Marquis, K. B. Averyt, M. Tignor, H. L. Miller, (eds)], Cambridge University PressCambridge, United Kingdom and New York, NY, USA, 996 pp., 2007.

[3] USGCRP 2017, *Climate Science Special Report: Fourth National Climate Assessment, Volume I*, [Wuebbles, D.J., D.W. Fahey, K.A. Hibbard, D.J. Dokken, B.C. Stewart, and T.K. Maycock (eds.)]. U.S. Global Change Research Program, Washington, DC, USA, 470 pp, 2017.

[4] IPCC, *Climate Change 2007: Mitigation. Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, e [B. Metz, O.R. Davidson, P.R. Bosch, R. Dave, L.A. Meyer (eds)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA., 851 pp., 2007.

[5] Tufa RA, Chanda D, Ma M, Aili D, Demissie TB, Vaes J, et al. Towards highly efficient electrochemical CO_2 reduction: Cell designs, membranes and electrocatalysts. Appl Energy 2020;277:115557.

[6] Saravanan A, Senthil Kumar P, Vo D-V-N, Jeevanantham S, Bhuvaneswari V, Anantha Narayanan V, et al. A comprehensive review on different approaches for CO_2 utilization and conversion pathways. Chem Eng Sci 2021;236:116515.

[7] Kamkeng ADN, Wang M, Hu J, Du W, Qian F. Transformation technologies for CO_2 utilisation: Current status, challenges and future prospects. Chem Eng J 2021;409:128138.

[8] Garba MD, Usman M, Khan S, Shehzad F, Galadima A, Ehsan MF, et al. CO_2 towards fuels: A review of catalytic conversion of carbon dioxide to hydrocarbons. J Environ Chem Eng 2021;9:104756.

[9] Zhu DD, Liu JL, Qiao SZ. Recent Advances in Inorganic Heterogeneous Electrocatalysts for Reduction of Carbon Dioxide. Adv Mater 2016;28:3423–52.

[10] Liu A, Gao M, Ren X, Meng F, Yang Y, Gao L, et al. Current progress in electrocatalytic carbon dioxide reduction to fuels on heterogeneous catalysts. J Mater Chem A 2020;8:3541–62.

[11] Jia C, Dastafkan K, Ren W, Yang W, Zhao C. Carbon-based catalysts for electrochemical CO_2 reduction. Sustainable Energy Fuels 2019;3:2890–906.

[12] Zhao K, Quan X. Carbon-Based Materials for Electrochemical Reduction of CO2 to C2+ Oxygenates: Recent Progress and Remaining Challenges. ACS Catal 2021;11:2076–97.

[13] Edwards JP, Xu Y, Gabardo CM, Dinh C-T, Li J, Qi Z, et al. Efficient electrocatalytic conversion of carbon dioxide in a low-resistance pressurized alkaline electrolyzer. Appl Energy 2020;261:114305.

[14] Kim D, Han J. Comprehensive analysis of two catalytic processes to produce formic acid from carbon dioxide. Appl Energy 2020;264:114711.

[15] Dominguez-Ramos A, Singh B, Zhang X, Hertwich EG, Irabien A. Global warming footprint of the electrochemical reduction of carbon dioxide to formate. J Clean Prod 2015;104:148–55.

[16] Rumayor M, Dominguez-Ramos A, Irabien A. Environmental and economic assessment of the formic acid electrochemical manufacture using carbon dioxide: Influence of the electrode lifetime. Sustain Prod Consum 2019;18:72–82.

[17] Kibria Nabil S, McCoy S, Kibria MG. Comparative life cycle assessment of electrochemical upgrading of CO_2 to fuels and feedstocks. Green Chemistry 2021;23:867–80.

[18] Greenblatt JB, Miller DJ, Ager JW, Houle FA, Sharp ID. The Technical and Energetic Challenges of Separating (Photo)Electrochemical Carbon Dioxide Reduction Products. Joule 2018;2:381–420.

[19] Li X, Anderson P, Jhong H-R-M, Paster M, Stubbins JF, Kenis PJA. Greenhouse Gas Emissions, Energy Efficiency, and Cost of Synthetic Fuel Production Using Electrochemical CO2 Conversion and the Fischer-Tropsch Process. Energy Fuel 2016;30:5980–9.

[20] P. De Luna, C. Hahn, D. Higgins, S. A. Jaffer, T. F. Jaramillo and E. H. Sargent, What would it take for renewably powered electrosynthesis to displace petrochemical processes?, *Science (1979)*, 2019, **364**, eaav3506.

[21] Khoo HH, Halim I, Handoko AD. LCA of electrochemical reduction of CO_2 to ethylene. J CO2 Util 2020;41:101229.

[22] Yue P, Fu Q, Li J, Zhu X, Liao Q. Comparative life cycle and economic assessments of various value-added chemicals' production via electrochemical CO_2 reduction. Green Chem 2022;24:2927–36.

[23] Wyndorps J, Ostovari H, von der Assen N. Is electrochemical CO2 reduction the future technology for power-to-chemicals? An environmental comparison with H2-based pathways. Sustain Energy Fuels 2021;5:5748–61.

[24] Rojas Sánchez D, Khalilpour K, Hoadley AFA. How sustainable is CO2 conversion to ethanol? – A life cycle assessment of a new electrocatalytic carbon utilisation process. Sustain Energy Fuels 2021;5:5866–80.

[25] Yue P, Kang Z, Fu Q, Li J, Zhang L, Zhu X, et al. Life cycle and economic analysis of chemicals production via electrolytic (bi)carbonate and gaseous CO2 conversion. Appl Energy 2021;304:117768.

[26] Somoza-Tornos A, Guerra OJ, Crow AM, Smith WA, Hodge B-M. Process modeling, techno-economic assessment, and life cycle assessment of the electrochemical reduction of CO_2: a review. *iScience* 2021;24:102813.

[27] Thonemann N. Environmental impacts of CO2-based chemical production: A systematic literature review and meta-analysis. Appl Energy 2020;263:114599.

[28] Wang X, Yang M, Zhu X, Zhu L, Wang S. Experimental study and life cycle assessment of CO2 methanation over biochar supported catalysts. Appl Energy 2020;280:115919.

[29] Alerte T, Edwards JP, Gabardo CM, O'Brien CP, Gaona A, Wicks J, et al. Downstream of the CO_2 Electrolyzer: Assessing the Energy Intensity of Product Separation. ACS Energy Lett 2021;6:4405–12.

[30] Federal Commons LCA data repository, https://www.lcacommons.gov/lca-collaboration/.

[31] NETL (2022), NETL CO2U openLCA LCI Database (Version 2.0), Pittsburgh, PA: National Energy Technology Laboratory, U.S. Department of Energy.

[32] Bare JC. Traci. J Ind Ecol 2002;6:49–78.

[33] Bare J. TRACI 2.0: the tool for the reduction and assessment of chemical and other environmental impacts 2.0, *Clean Technol Environ.* Policy 2011;13:687–96.

[34] Bare JC, *Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) TRACI version 2.1 User's Guide*, U.S. EPA Office of Research and Development, Washington, DC, EPA/600/R-12/554, 2014.

[35] International Energy Agency, Global Energy & CO2 Status Report 2019 Emissions, https://www.iea.org/reports/global-energy-co2-status-report-2019/emissions.

[36] Yang H, Kaczur JJ, Sajjad SD, Masel RI. Performance and long-term stability of CO_2 conversion to formic acid using a three-compartment electrolyzer design. J of CO_2 Utilization 2020;42:101349.

[37] US Patent, US4326073A, 1982.

[38] Shemfe M, Gadkari S, Yu E, Rasul S, Scott K, Head IM, et al. Life cycle, techno-economic and dynamic simulation assessment of bioelectrochemical systems: A case of formic acid synthesis. Bioresour Technol 2018;255:39–49.

[39] Jung J, Postels S, Bardow A. Cleaner chlorine production using oxygen depolarized cathodes? A life cycle assessment. *J Clean Prod* 2014;80:46–56.

[40] Sadhukhan J, Joshi N, Shemfe M, Lloyd JR. Life cycle assessment of sustainable raw material acquisition for functional magnetite bionanoparticle production. J Environ Manage 2017;199:116–25.

[41] Martín AJ, Larrazábal GO, Pérez-Ramírez J. Towards sustainable fuels and chemicals through the electrochemical reduction of CO_2: lessons from water electrolysis. Green Chem 2015;17:5114–30.

[42] Malkhandi S, Yeo BS. Electrochemical conversion of carbon dioxide to high value chemicals using gas-diffusion electrodes. Curr Opin Chem Eng 2019;26:112–21.

[43] Rosental M, Fröhlich T, Liebich A. Life Cycle Assessment of Carbon Capture and Utilization for the Production of Large Volume Organic Chemicals. Frontiers in Climate 2020;2:586199.

[44] Sadok R, Benveniste G, Wang L, Clavreul J, Brunot A, Cren J, et al. Life cycle assessment of power-to-gas applications via co-electrolysis of CO_2 and H_2O Life cycle assessment of power-to-gas applications via co-electrolysis of CO_2 and H_2O. J Phys Energy 2020;2:024006.

[45] European Environmental Agency, Greenhouse gas emission intensity of electricity generation in Europe, https://www.eea.europa.eu/ims/greenhouse-gas-emission-intensity-of-1.

[46] Artz J, Müller TE, Thenert K, Kleinekorte J, Meys R, Sternberg A, et al. Sustainable Conversion of Carbon Dioxide: An Integrated Review of Catalysis and Life Cycle Assessment. Chem Rev 2018;118:434–504.

[47] Bennett SJ, Schroeder DJ, McCoy ST. Towards a Framework for Discussing and Assessing CO_2 Utilisation in a Climate Context. Energy Procedia 2014;63:7976–92.

[48] Erol S. A Statistical Design Approach on Electrochemical Impedance Spectroscopy of NMC Li-Ion Battery. J Electrochem Soc 2022;169:100503.

[49] Temiz S, Kurban H, Erol S, Dalkilic MM. Regeneration of Lithium-ion battery impedance using a novel machine learning framework and minimal empirical data. J Energy Storage 2022;52:105022.

[50] Morali U, Erol S. Analysis of electrochemical impedance spectroscopy response for commercial lithium-ion batteries: modeling of equivalent circuit elements. Turk J Chem 2020;44:602–13.

[51] Algren M, Fisher W, A. e.. Landis, in *Data Science Applied to Sustainability Analysis*. Elsevier 2021:167–90.

[52] Ghoroghi A, Rezgui Y, Petri I, Beach T. Advances in application of machine learning to life cycle assessment: a literature review. Int J Life Cycle Assess 2022; 27:433–56.