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Learning from the past: Limitations of techno-economic assessments for low-temperature CO_2 electrolysis

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ABSTRACT

Reaching climate goals requires a rapid scale-up of clean energy technologies, which, in many cases, are still under development. Low-temperature CO_2 electrolysis (LT CO_2E) is a promising pre-commercial technology (TRL 3 to 6) that can produce CO_2-based fuels and chemicals using electricity. To understand the future competitiveness of such novel technologies, techno-economic assessments (TEAs) are conducted using the best available knowledge at the time, ensuring that the highest-quality TEA information supports decision-making regarding future investments. As LT CO_2E advances, its techno-economic research must evolve toward more in-depth process designs, integrating the latest knowledge regarding the technology's development and any aspects essential to commercial implementation. To do so, it is important to understand the robustness and limitations of existing LT CO_2E TEAs to identify areas for further improvement; for example, electricity and CO_2 cost assumptions vary significantly between TEAs for syngas, accounting for 18–81% and up to 28% of the total operational expenditure, respectively. This review assessed the origins and justifications behind common assumptions used in TEAs of LT CO_2E with three main findings: 1) the methodological justifications seem stuck in the past, relying on three key studies and mature electrolysis technologies from previous decades; 2) the latest advancements in electrolyzer modeling underscore the need to update existing LT CO_2E performance benchmarks, and 3) future LT CO_2E TEAs need to include pre-treatment of CO_2 and water, product separation steps, as well as heat integration, recycling, and waste valorization, to progress beyond the preliminary conceptual design phase.

1. Introduction

Achieving our global climate goal of net-zero emissions by 2050 requires the rapid scaling up of existing, new, and emerging clean energy technologies [1]. For example, renewable power generation technologies, water electrolysis, biofuel technologies, and carbon capture and utilization (CCU) are currently at different technology readiness levels (TRLs) [2]. TRLs are used to indicate how far a certain technology is from commercial implementation, ranging from 1 (i.e., basic principles observed) to 9 (i.e., proven in an operational environment) [3,4]. However, knowing the TRL of a technology is insufficient to predict or make investment decisions to further develop it to a commercial status.

An ex-ante techno-economic assessment (TEA) is a method that can be used to evaluate and steer research and investment in emerging (low TRL) technologies. An ex-ante technology assessment implies that the TEA is based on observed performance at a pre-commercial scale and, therefore, is inherently subject to high uncertainty. The TEA entails an integrated analysis of the technical and economic performance of a technology [5] that uses the best available data at the moment of the assessment (e.g., first-order TEA estimations for low TRL technologies [6]). Its main aim is to uncover potential cost-performance tradeoffs to support research and development and to identify bottlenecks for industrial implementation so they can be resolved as early as possible [7]. Using the information obtained from TEAs, different investment options can be compared side by side in terms of process design requirements as well as overall economic feasibility.

Carbon dioxide electrolysis (CO_2E) is a pre-commercial CCU technology that facilitates the production of CO_2-based chemicals, using CO_2 as a raw material coupled with electricity from renewable sources. Besides the potential to reduce CO_2 emissions, CO_2E is considered a key technology for enabling the transition away from fossil-based feedstocks

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Nomenclature

Abbreviations

0-D	Zero-dimensional
1-D	One-dimensional
2-D	Two-dimensional
AC	Alternating current
AE	Alkaline electrolyzer
AEM	Anion exchange membrane
BPM	Bipolar membrane
CAPEX	Capital expenditure
CCU	Carbon capture and utilization
CO	Carbon monoxide
CO_2	Carbon dioxide
CO_2E	Carbon dioxide electrolysis
DAC	Direct air capture
DC	Direct current
DOE H2A	United States Department of Energy Hydrogen Analysis model
EE	Energy efficiency
ETS	Emission Trade System
EUR	Euro
FE	Faradaic efficiency
GDEs	Gas diffusion electrodes
H_2	Hydrogen
HT	High-temperature
ISPT	Dutch Institute for Sustainable Process Technology
LT	Low-temperature
MEA	Monoethanolamine
MCE	Molten carbonate electrolyzer
NASEM	National Academies of Sciences, Engineering, and Medicine
NPV	Net present value
OPEX	Operational expenditure
P	Pressure
PEM	Proton exchange membrane
PFD	Process flow diagram
PSA	Pressure-swing adsorption
PV	Photovoltaic
SOE	Solid-oxide electrolyzer
Syngas	Synthesis gas
T	Temperature
TRL	Technology readiness level
TEA	Techno-economic assessment
USD	United States dollar

Symbols

j	Current density
€	Euro (EUR)
$	United States dollar (USD)

Units

GW	Gigawatt
K	Kelvin
kmol	Kilomoles
kPa	Kilopascal
kWh	Kilowatt hours
MW	Megawatt
mA/cm^2	Milliampere per square centimeter
m^2	Square meter
m^3	Cubic meter
pH	Potential of hydrogen
V	Voltage

in the chemical and energy sectors [2,8]. CO_2E belongs to the family of electrolysis-based technologies (e.g., batteries, fuel cells) that can also be used as energy storage solutions to deal with fluctuations in electricity supply and demand due to the increased use of intermittent renewable electricity sources (i.e., wind and solar) [9,10]. However, the focus of CO_2E development has been progressively moving away from being mainly an electricity storage solution towards being considered as an alternative to fossil-based processes due to the need for alternative carbon sources for synthesizing hydrocarbon molecules for materials or fuels, such as air and heavy-duty transport applications [11].

1.1. Electrolysis-based technologies

Electrolysis-based technologies exist at various TRLs. Mature technologies include aluminum production via the Hall-Héroult process (i.e., an electrolytic process for smelting alumina (Al_2O_3) dissolved in a molten fluoride solvent (cryolite) to produce pure aluminum metal [12]), chlorine production via the chlor-alkali process, and hydrogen production via alkaline water electrolyzers powered by continuous electricity (e.g., hydropower), all commercially deployed since the 19th century [13,14]. Technologies such as water electrolysis for hydrogen production powered by a renewable intermittent electricity supply (e.g., wind or solar) are currently at demonstration scale (TRL 8), and are being scaled up, for instance, to meet the 1 Gigawatt target by 2030 [2,15,16]. Nevertheless, challenges remain to make green hydrogen cost-competitive, such as the process safety of large-scale green hydrogen plants (e.g., hazards related to hydrogen and oxygen mixing inside the electrolyzer unit), as well as considerable reductions in capital costs via standardizing the equipment, increasing durability to extend equipment lifetime, and determining the optimal mode(s) of operation (e.g., load-following in accordance with the intermittent electricity supply, or in combination with mass and/or energy storage means) [15,17,18].

Between 2000 and 2023, CO_2 electrolysis (and co-electrolysis of CO_2 and steam) received increasing attention in academic research due to its potential to speed up the energy and industrial transition away from fossil resources [19–26]. Inspired by water electrolyzers [17] and fuel cells, various CO_2 electrolyzer designs have been proposed, namely alkaline electrolyzers (AE), proton exchange membrane electrolyzers (PEM), anion exchange membrane electrolyzers (AEM), bipolar membrane electrolyzers (BPM), molten carbonate electrolyzers (MCE), and solid-oxide electrolyzers (SOE) [2,15,17,19,21]. Major differences among designs include the electrolyte phase (liquid vs. solid) and required operating temperatures (low: 40–90°C vs. high: 700–900°C), which in turn, affect the electrolyzer's capital and operational expenditures due to differences in materials and components for the electrolyzer equipment as well as process design requirements (e.g., electrolyte recycling or heat integration possibilities).

High-temperature solid-oxide electrolyzers (HT CO_2E) can be used to generate carbon monoxide, methane, and syngas (i.e., a targeted mixture of H_2 and CO, where the H_2:CO ratio can be controlled by adjusting the feed gas composition, temperature, and cell voltage to suit downstream applications, such as 1:1 for the production of aldehydes via hydroformylation or 2:1 for Fischer-Tropsch fuel synthesis [27]) with high CO_2 conversion and electricity efficiencies, and have the potential for coupling with waste heat streams. HT CO_2E powered by continuous electricity sources has already been demonstrated in an operational environment (TRL 6–8) [19–21,28–31]. Companies producing demo units include Haldor Topsøe and SUNFIRE [32,33]. Initial applications focus on incorporating HT CO_2E into existing processes, such as in methanol synthesis (where carbon monoxide is used on a large scale) to improve environmental performance [31,34]. To reach large-scale commercial applications, some explorative TEAs envision producing liquid products via an indirect route (i.e., HT CO_2E to syngas combined with Fischer-Tropsch fuel synthesis), whereas others focus on evaluating the economic viability of the products directly formed via HT CO_2E (i.e., carbon monoxide/syngas) [35–39].

Low-temperature liquid-phase electrolyzers (LT CO_2E) are at the pre-commercial stage (TRL 3–6), with initial commercialization efforts by startups such as Twelve and Dioxycle [2,20,40]. So far, advancements in understanding the fundamental reaction mechanisms, catalyst innovations, and novel cell architectures (i.e., the evolution from H-cells to flow-cell reactors) [19,21,23,41–43] have enabled the synthesis of carbon monoxide, formate, as well as a wide range of multi-carbon products at the lab-scale. In experimental LT CO_2E studies, the most used figures of merit are current density (j), faradaic efficiency (FE), energy efficiency (EE), equipment size, and durability [22]. Advancements in these figures of merit vary depending on the chemicals produced and are not directly comparable across products [44]. This review primarily highlights advancements in carbon monoxide/syngas and ethylene, serving as representative C1 and C2 chemicals at different TRLs. For carbon monoxide and syngas, FE, EE, and current densities have significantly increased [19,22,44,45] alongside validation in preliminary process development projects [46], placing their TRL at 5–6 [40]. For direct LT CO_2E to ethylene, low selectivity (i.e., FE) and the challenge of reducing electrolyzer potential below 3 V currently constrain its TRL to 3–4 [40,47]. Additional technological challenges, such as CO_2 cross-over issues related to carbonate formation in AEs, energy losses, as well as limited electrochemical cell durabilities, continue to hinder the overall development of LT CO_2E toward commercial applications [20,44,47].

1.2. Techno-economic research for low-temperature CO_2 electrolysis

In the last five years (2018–2023), research exploring the pathways toward industrial implementation of LT CO_2E has gained increasing attention [22]. First-order TEAs by e.g., de Luna et al., Bushyev et al., and Jouny et al. [45,48,49] have sketched the contours of economic feasibility for LT CO_2E using lab-scale performance data in combination with process systems engineering methods to estimate and compare future industrial-scale production costs for LT CO_2E products [48–51]. The results focus on the performance benchmarks that LT CO_2E would need to achieve to be economically feasible for various C1, C2, and even C3 products (e.g., syngas, olefins, alcohols) [48–51]. Particular attention in this review is given to carbon monoxide/syngas and ethylene. These two chemicals are not only well-studied products in the CO_2E field [22,43,44], but also strategically position LT CO_2E as a potential technology to complement or replace petrochemical processes by enabling the production of a broad range of chemicals from CO_2 in both the short- and long-term future (e.g., production of liquid fuels via Fischer-Tropsch synthesis, or ethanol via syngas fermentation, as well as manufacturing of plastics, detergents, and agricultural products) [8]. However, despite this potential, research has also begun to discuss scale-up bottlenecks for LT CO_2E, such as CO_2 availability (i.e., location, quantity, and required purity), energy requirement, and land use, as well as direct, indirect, and tandem set-ups [11,52,53].

The bottlenecks and electrolyzer performance benchmarks identified via current techno-economic research are intended to guide the short-term and long-term research and development objectives for LT CO_2E. However, further examination of the TEA studies reveals that the economic viability of LT CO_2E products often hinges on optimistic assumptions of potential future conditions (e.g. a continuous renewable electricity flow at a price of ~0.02 USD/kWh, a carbon tax of ~100 USD/tonne CO_2), and on electrolyzer performance benchmarks not yet proven at the lab-scale (e.g. EE $>70\%$, j $>$ 200 mA/cm^2, 0.5V overpotential at FE $>90\%$ [22,43]). Alongside this, a comparison of nine LT CO_2E TEAs [19,48,50,51,54–58] revealed order-of-magnitude differences in projected production costs between studies for formic acid, methanol, ethanol, and ethylene [22]. Uncertain and heterogeneous input assumptions, as well as differences in ex-ante TEA outcomes (i.e. within one order of magnitude [5]), are to be expected, especially in the preliminary research phase [5]. However, order of magnitude differences between TEAs may jeopardize future investments and targeted research for LT CO_2E, echoing past challenges seen in the development of technologies like solar photovoltaics (PV) [59] and fuel cells [60] (e.g., fragmented research, over-reliance on outdated data, and imposition of unrealistic assumptions).

Improved LT CO_2E techno-economic research based on the best information available is needed to reestablish research targets and provide clarity about the investments required to reach higher TRLs at the speed needed to fulfill our climate goals. To provide such refined results, the underlying modeling and input assumptions used in the current LT CO_2E TEAs need to be further investigated to pinpoint improvement areas. Thus far, the reviews by Ruiz-López et al. [43] and Lin et al. [24] focused on comparing CO_2E products, economic parameters, and performance targets between TEAs [19,48,49,53,61] and highlighted differences in performance benchmarks (e.g., FE$>$70% [48] vs. FE$>$90% [49] for the same products: ethanol, ethylene, and propanol) as well as differences in the most promising LT CO_2E products to target (e.g., carbon monoxide and formic acid [61] vs. ethanol, ethylene, and propanol [49]). Other reviews by Somoza-Tornos et al. [22], Gawel et al. [21], Park et al. [26], Greenblatt et al. [62], and Hung et al. [23] analyzed the process modeling assumptions related to CO_2 capture, product separation, as well as capital and operating costs of the electrolyzer. These studies [21–23,26] conclude that the variety in electricity cost assumptions (0.02–0.10 USD/kWh) and CO_2 electrolyzer metrics (e.g. FE, voltage, current density, lifetime) are potential causes for the significant production cost differences between studies. However, the justifications underlying the process modeling and input assumptions have not yet been investigated in detail [21,22,24,26,43].

This study assessed the origins and justifications behind existing LT CO_2E TEA modeling and input assumptions via a systematic literature review to pinpoint which areas need further improvement and attention in the future, thereby providing a basis for advancing with the best available knowledge and identifying key aspects for commercial implementation. The review focused on uncovering the roots of LT CO_2E TEAs via a family tree analysis (Section 3.1), the modeling and input assumptions related to the CO_2 electrolyzer (Section 3.2), and the plant design and economic assumptions related to the pre-treatment of CO_2 and water, downstream processing, and electricity (Section 3.3).

2. Methodology

The methodology consisted of a systematic literature review based on four steps: 1) review of the ex-ante TEA methodology, 2) identification of relevant literature, 3) investigation of the origins and justifications via a family tree analysis, and 4) compilation and comparison of the modeling and input assumptions.

The ex-ante TEA methodology is centered around the development of an integrated process and cost model [5] for a low-TRL technology to estimate its future industrial-scale performance. For the case of LT CO_2E techno-economic research, the most important element of the model is the main process unit (i.e., the CO_2 electrolyzer) around which a process flow diagram (PFD) can be developed. The PFD depicts the rest of the equipment, mass, and energy flows required to convert the raw materials into the desired products. Typically, process models are created using spreadsheet software (e.g., Microsoft Excel) or process simulators (e.g., Aspen Plus). The capital (CAPEX) and operational expenditure (OPEX) are usually estimated using a factored approach [63], based on the equipment lists, and mass and energy balances from the process model. The overall techno-economic performance is often represented using technical indicators like feedstock conversion efficiency, energy consumption, and economic indicators like levelized cost of production, net present value (NPV), or payback period [63]. The methodological building blocks that comprise the ex-ante TEA methodology are summarized in Fig. 1.

Considering a TEA for a commercial technology (TRL 9), where experience and vendor quotes are available, the classic "bottom-up" engineering-economic approach based on major equipment items has an expected accuracy of ± 25% [5]. The accuracy level is affected by the different levels of uncertainty present in every building block of the TEA. For the ex-ante evaluation of low-TRL technologies such as LT CO_2E, industrial-scale electrolyzer cost-performance data, vendor data, and PFDs are not yet available. From a methodological viewpoint [64,65], this means ex-ante LT CO_2E TEAs have larger uncertainties, which are related to the scale-up of the technology from lab-scale to industrial-scale and the further evolution of the technology's cost and performance over time, as visualized in Fig. 1.

For the identification of literature, we used Scopus and the search query: "electro* AND *reduction AND CO_2 AND economic". In total, 487 articles published in peer-reviewed journals were found (as of December 2021). The search results show that LT CO_2E studies focusing on techno-economics have slowly gained momentum between 2018 and 2021. This review focused on the 324 articles in English published between 2018 and 2021. Articles published in 2022, 2023, and industrial reports were included via snowballing. During the screening, snowballing, and eligibility check, the number of articles was reduced to 29 LT CO_2E TEAs at the process level, which were included for further analysis. Fundamental, supply chain, or (energy) system level studies were considered out of scope. We performed a similar search to include TEAs of LT water electrolysis to hydrogen, power-to-gas, indirect routes for methanol production, and chlor-alkali processes to extract methodological lessons learned from other electrolysis-based technologies. Additional details and a flow diagram describing the literature identification process are provided in Fig. S1 in the Supplementary Information (SI).

Next, we investigated the origins and justifications behind the most common modeling and input assumptions via backward snowballing. Note that we distinguished between assumptions related to the modeling (e.g., the flowsheet, thermodynamics, chemistry, unit operations) and the input parameters (e.g., feed streams, boundary conditions, pure component properties) in the manner described by van der Spek et al. [66]. The focus of our investigation was to trace which electrolysis-based technologies the LT CO_2E TEA assumptions were based on and why, as we noticed a mixture of electrolysis-based technologies in the methodological citations of the LT CO_2E TEAs. We analyzed the connections between the LT CO_2E TEAs and other electrolysis TEAs based on citations. This resulted in 6 additional handbooks and industrial reports. Additionally, we identified the countries and university groups that have performed the research. The results are visualized using a family tree and discussed in Section 3.1.

Finally, we compared the modeling, cost estimation, and input assumptions in the selected literature and the accompanying supplementary materials related to the CO_2 electrolyzer (Section 3.2) and the plant design regarding pre-treatment of CO_2 and water, downstream processing, and electricity (Section 3.3). The comparison tables for the 29 LT CO_2E TEAs (see the SI) were obtained by systematically cataloging the assumptions in the corresponding category. Plots visualizing the correlations between input assumptions (e.g., CO_2 price, electricity price, current density) and TEA outcomes (e.g., CAPEX, OPEX) are shown and discussed in Sections 3.2 and 3.3. Note that the economic figures mentioned and used in the current work are reported in the currency and year used in the original papers.

Possible methodological limitations include 1) the scope of selected articles, constrained by the time period, focus on English-language literature, and number of studies, and 2) the emphasis on specific aspects such as the modeling and input assumptions related to CO_2 electrolyzers (mainly focused on carbon monoxide/syngas and ethylene), pre-treatment of CO_2, downstream processing, and electricity. These constraints may have excluded relevant literature or overlooked other critical improvement areas in the LT CO_2E field.

3. Results and discussion

Using the previously described methodology, the 29 studies analyzed can now be compared in terms of 1) origins (i.e., family tree analysis), 2) CO_2 electrolyzer modeling and input assumptions, and 3) process design such as pre-treatment of CO_2 and water, downstream processing, and electricity. The methodological similarities and differences with other electrolysis-based TEAs were also investigated to elucidate potential improvement areas for future LT CO_2E TEAs.

3.1. *The family tree of LT CO_2 electrolysis TEAs*

The family tree analysis of the 29 papers unveils a pattern of interconnected citations, with the modeling and input assumptions in LT CO_2E TEAs frequently sourced from each other. The primary source of input assumptions continues to be low-temperature water electrolysis despite the technological developments observed in the high-

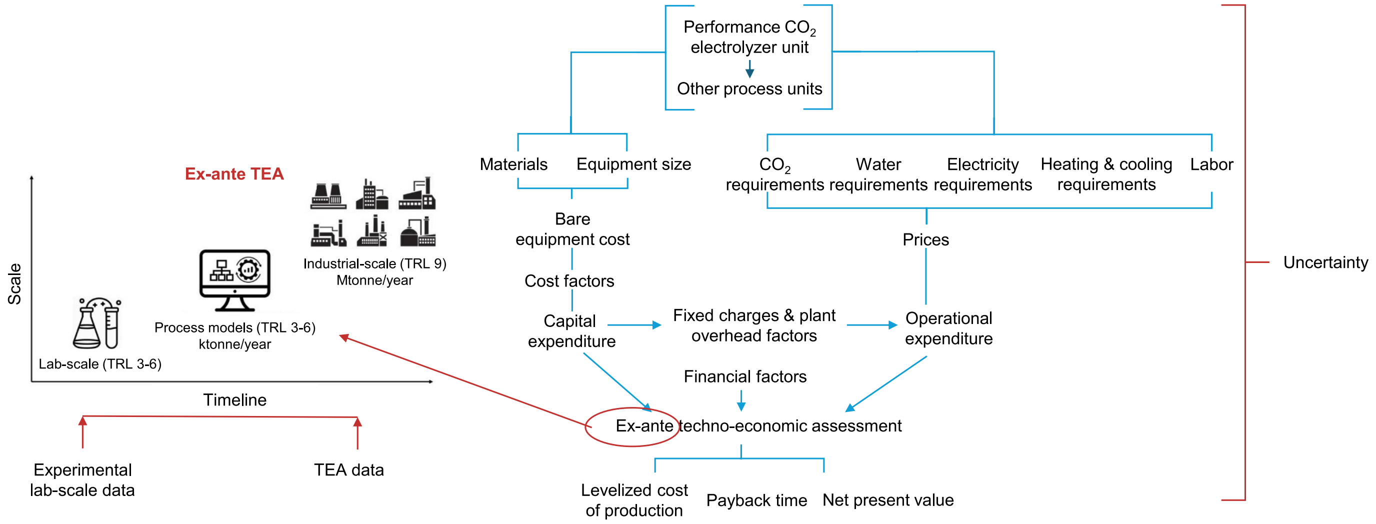

Fig. 1. Visual overview of the methodological building blocks that comprise an ex-ante techno-economic assessment of a future LT CO_2E-based plant. Note that since LT CO_2E is not yet at a commercial scale, uncertainty is high when estimating technical and economic parameters. Icons by the Noun Project.

temperature (water, CO_2, and co-electrolysis) electrolysis field. The number of citations, links between research groups, and connections with other LT electrolysis-based technologies suggest that a small yet well-connected community of academics and institutions is supporting the development of the field and, in consequence, is defining the performance benchmarks for the economic viability of future LT CO_2E plants. While interconnected citations are likely to occur for any given niche topic [67], the continued re-use of modeling and input assumptions from a few highly cited TEAs and handbooks from previous decades calls for a reevaluation to ensure alignment with the most up-to-date information regarding the techno-economic performance of LT CO_2E.

Fig. 2 shows the included 29 LT CO_2E TEAs (green and yellow boxes) in the form of a family tree to visualize the methodology-based citations between them (details in Table S2 in the SI). The analysis exhibits that the methodological assumptions used in the LT CO_2E TEAs developed after 2018 can be traced back to three TEAs: Jouny et al. (2018), Spurgeon et al. (2018), and Verma et al. (2016) [48], [51], [61] (yellow boxes in Fig. 2). Further citation tracking shows that the three key studies, in turn, originate from initial CO_2-to-formic acid/formate TEAs by Oloman et al. (2008) and Agarwal et al. (2011), Keets et al. (2012), and a mini-review by Pletcher (2015) [55], [68]– [69]. The first TEA for LT CO_2E was developed by Oloman et al. [68] in 2008 at the University of British Columbia and has shaped the current line of thinking in the LT CO_2E field.

Our analysis highlights the mixture of high TRL LT electrolysis-based technologies, namely chlor-alkali, alkaline, AEM, or PEM water electrolyzers, and fuel cells (bibliographic sources in blue boxes in Fig. 2) that have been used as the basis for LT CO_2 electrolyzer modeling, inputs, and cost assumptions (e.g., electrolyzer capital cost). The first TEA by Oloman et al. (2008) [68] based its LT CO_2E industrial-scale performance model and input assumptions on electrochemistry handbooks by Pletcher (1991 & 1993), Walsh (1993), and Goodridge (1995) [13, 70–72], which were built based on the knowledge generated from the European electroplating, water electrolysis, and chlor-alkali industries available before the 1990s.

Most LT CO_2E TEAs [45,48,51,55,56,61,73–80] developed after Oloman et al. cite the US Department of Energy Hydrogen Analysis model (DOE H2A for water electrolysis) [81] as the basis for their LT CO_2E assumptions. However, despite citing the same reference, the TEAs select different LT water electrolyzer designs (e.g., PEM, alkaline) to represent the LT CO_2 electrolyzer. For example, Jouny et al. [48] considered commercial-scale CO_2 electrolyzers to be similar to alkaline water electrolyzers, whereas Spurgeon et al. [51] selected PEM water electrolyzers. Also, some TEAs [49,50,82] do not specify the type of electrolyzer design that was considered. The type of electrolyzer design

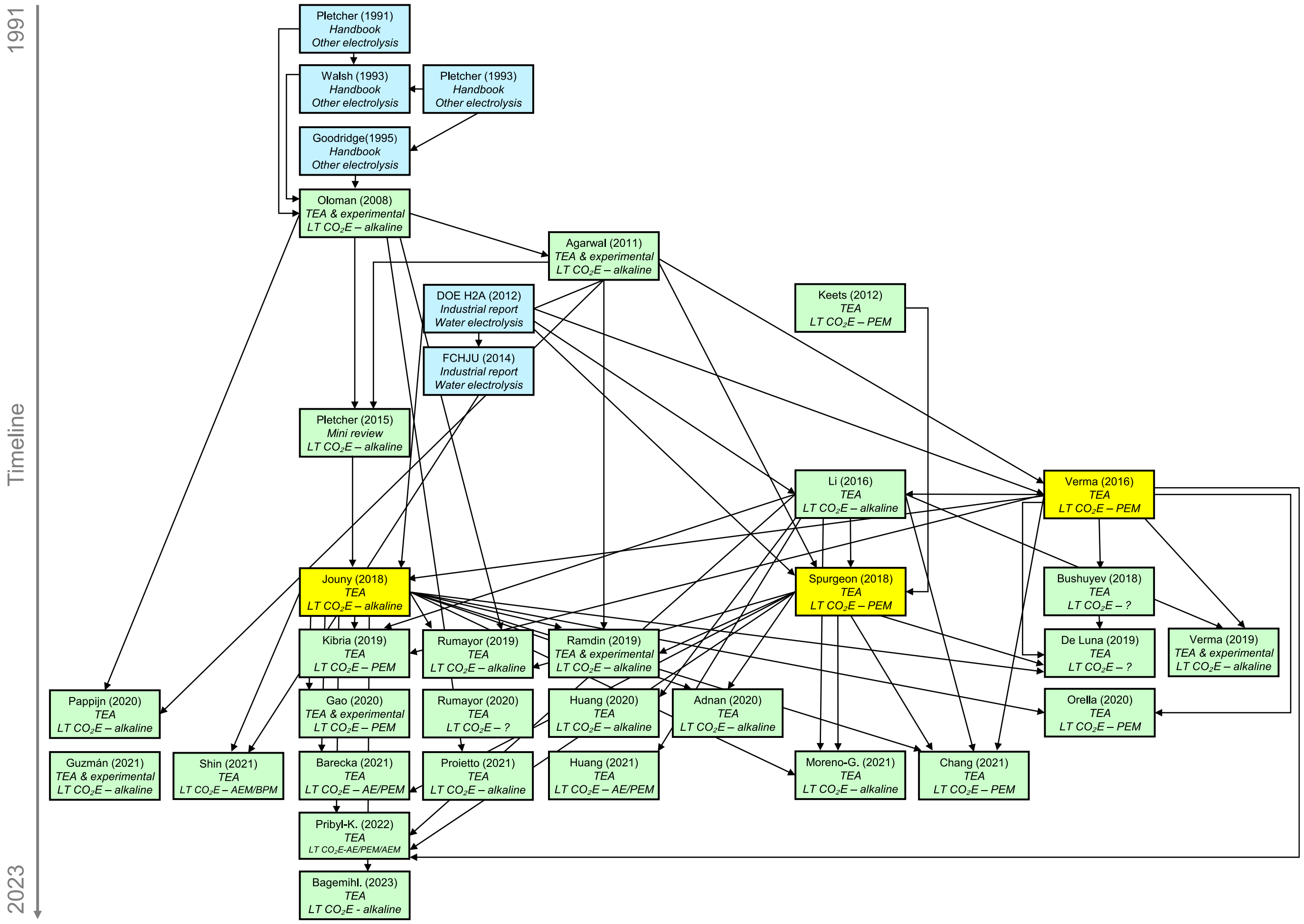

Fig. 2. Family tree of LT CO_2E TEAs based on 29 articles between 2018 and 2023 and 6 handbooks and industrial reports. The family tree depicts methodological citations between the studies. Legend: The green boxes represent the LT CO_2E studies, the yellow boxes are the most cited TEAs, and the blue boxes are the handbooks and industrial reports focusing on other electrolysis technologies. Each box includes information regarding the assumed electrolyzer design type (i.e., alkaline (AE), anion exchange membrane (AEM), bipolar membrane (BPM), proton exchange membrane (PEM)) and research group. The citation data analyses corresponding to each bibliographic source are summarized in Table S2 in the SI. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

is a key assumption because the electrolyzer cost-performance model is the main building block within the ex-ante LT CO_2E TEA methodology (Fig. 1) and, thus, an important factor influencing the overall TEA outcome. Insights from water electrolysis [17,30] indicate differences between electrolyzer designs exist at three levels: cell, stack, and balance of plant level. Therefore, differences in plant performance, overall equipment list, and bare equipment costs make the estimation of the industrial-scale LT CO_2 electrolyzer cost and performance dependent on the selected electrolyzer design.

Although HT electrolysis has existed alongside LT electrolysis for some time, the analysis of studies indicates a lack of knowledge exchange between both fields. A review by Küngas [20] compared the state of the art for both HT and LT CO_2E technologies and pointed to a lack of commonly accepted figures of merit as a potential reason for the limited knowledge sharing. Küngas [20] highlighted the progress made for HT CO_2E in terms of reaching commercial-scale efficiencies, durability, and current densities. A recent perspective by Detz et al. (2023) [40] applied an experience curve methodology to compare designs of high- and low-temperature CO_2 electrolyzers. They also mention that high-temperature solid-oxide electrolysis for CO and syngas production is the most advanced and closest to achieving break-even levelized production costs compared to fossil-based benchmarks. Given that HT electrolysis is presently the most mature technology for CO_2E, one can question why LT CO_2E TEAs are not using the knowledge available in HT CO_2E TEAs [36–39,83–85].

3.2. CO_2 electrolyzer models

The analysis of the 29 electrolyzer models used in the LT CO_2E TEAs further highlights how the electrolyzer modeling assumptions have evolved from a combination of reactor requirements described in a few handbooks from the 1990s related to industrial-scale LT electrolysis processes (i.e., electroplating, water electrolysis, and chlor-alkali), more details in the upcoming section. Alongside this, differences were found between longstanding performance targets, for example, for ethylene (current densities >200 mA/cm^2) determined by LT CO_2E TEAs using single-cell reactor models with linear cost-performance relationships considering the five figures of merit independently of each other, when compared with the latest TEAs using more extensive electrolyzer models (>100 mA/cm^2) [86], more details to follow. Moving forward, it is essential that TEAs use the best available modeling approaches to provide updated research and development targets as the field progresses. Additionally, it is crucial to present the electrolyzer modeling limitations alongside the TEA results, for instance, by including the context and conditions under which the experimental input data applies (e.g., electrolyzer costs and performance for specific operating parameters, types, and target products).

Origins and justifications behind the electrolyzer assumptions: The foundations of the electrolyzer modeling and input assumptions of the first LT CO_2E TEAs can be traced to the electrochemistry handbooks by Pletcher et al. (1991 & 1993), Walsh et al. (1993), and Goodridge et al. (1995) [13,70–72]. In particular, the book by Walsh, *"A first course in electrochemical engineering"* [70], designed to serve as a practical guidebook for chemical engineers, proposed a range of electrolyzer cost-performance equations that consider the figures of merit (e.g., current density, faradaic efficiency, energy efficiency) related to the performance of an electrochemical cell as independent in order to quickly determine cost-performance tradeoffs at plant level. For instance, a high current density minimizes the size of the electrolyzer, thereby reducing the bare equipment costs of the electrolyzer and, in turn, the overall plant CAPEX. A high Faradaic efficiency leads to a high product purity and minimizes the number of by-products formed and the downstream processing units needed for the separation of the final product, which decreases the CAPEX and OPEX. A high energy efficiency decreases the amount of electricity needed to reach the desired product yield, thereby decreasing the OPEX. Whereas high durability will reduce maintenance and replacement costs for the electrolyzer and minimize downtime of the plant (a conceptual representation is shown in Fig. 3).

Citations in the paper by Oloman et al. [68] pointed out that the equations in Walsh's book [70] and input assumptions in Pletcher's book [13], such as current density >100 mA/cm^2 (related to chlor-alkali technologies), were used as a starting point for the first hypothetical design of a future LT CO_2E to formic acid/formate plant. Oloman et al. [68] proposed a list of LT CO_2E reactor requirements based on lessons learned from industrial-scale electrochemical and thermochemical processes, as shown in Table 1.

Oloman's TEA used lab-scale experimental results [88,89] to propose a crude (single-cell) reactor model to estimate the techno-economic performance of a commercial formic acid/formate plant converting 100 tonnes of CO_2 per day. The model considers the cathode as a 1-D, unipotential, adiabatic, two-phase, plug-flow reactor and uses experimental-based expressions for the stoichiometry, equilibria, and kinetics of the electrochemical reactions to solve the steady-state material and energy balances in a spreadsheet. The electrolyzer model was validated using experimental results (3 tonnes CO_2/day), and the model was then extrapolated to 100 tonnes CO_2/day.

The mini-review by Pletcher [69] reiterated Oloman's [68] design requirements for industrial-scale LT CO_2 electrolyzers by stating that "*the current density must be* $>$ 100 mA/cm^2 *for most electrolytic applications*". Additionally, Pletcher [69] mentioned that *"it is commonly advantageous to push towards a higher value"*. A high current density minimizes the size of the electrolyzer unit, thereby reducing capital investment costs (as described at the beginning of Section 3.2). This line of thinking is also described in the handbook by Walsh [70], in which cost-performance equations related to an electrochemical cell act as capital cost estimates for the overall plant. Pletcher and Walsh are co-authors of the book *"Industrial Electrochemistry"* [13], which builds on knowledge gained from industrial practice related to chlor-alkali technologies. Pletcher's mini-review [69] further stressed how higher current densities should be reached at the lab-scale and highlighted the importance of the experimental findings of Koplar et al. [90], who reached a current density of 200 mA/cm^2 for formate via gas diffusion electrodes (GDEs).

The LT CO_2E TEAs developed after 2018, all sharing the same heredity, also used electrolyzer modeling assumptions such as 1) a single-cell reactor model (i.e., the performance of a single electrolyzer cell at lab-scale is extrapolated to the performance of an industrial-scale stack), and 2) linear cost-performance relationships considering the five figures of merit independently of each other. Common input assumptions used in the 29 LT CO_2E TEAs include a current density >200 mA/cm^2, Faradaic efficiency $>90\%$, voltage ~ 2.0V, durability $>10{,}000$ h, and an energy efficiency $>70\%$.

Despite the similarities in electrolyzer modeling assumptions and inputs, there are orders of magnitude differences in overall plant CAPEX when comparing carbon monoxide studies with each other and ethylene studies with each other (Figs. 4 and 5). Moreover, the expected cost-performance relationships (e.g., higher current density and faradaic efficiency imply lower plant CAPEX) are not observed. As discussed in the family tree section, the studies have different assumptions on which electrolyzer design (e.g., PEM, alkaline) is most suitable as an analogy for a CO_2 electrolyzer. However, the results in Figs. 4 and 5 do not highlight robust differences in overall plant CAPEX between PEM and alkaline types.

Capturing the interdependency of figures of merit: The main simplification in the LT CO_2E cost-performance relationships is the independent consideration of the five figures of merit. Although experimental studies [41,53] show that the figures of merit are correlated and depend on the electrolyzer's operating conditions (i.e., temperature and pressure), the current LT CO_2E modeling approaches are usually limited by their inability to effectively account for the interdependencies between all five figures of merit and operating conditions. For instance, the voltage vs. current dependency affects the projected electricity

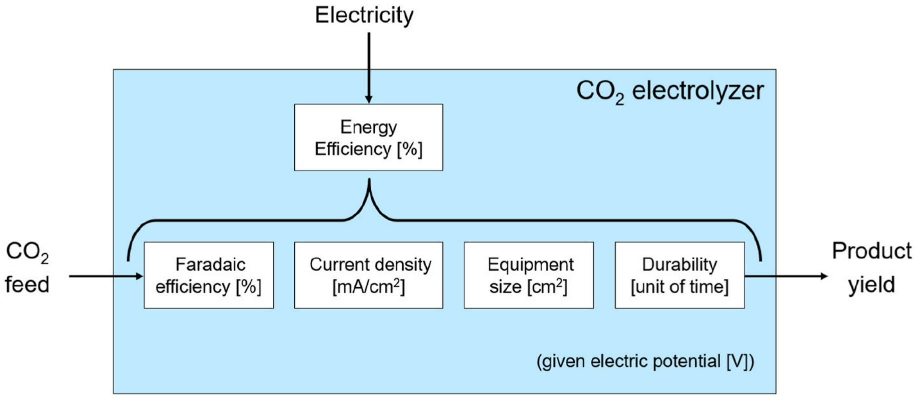

Fig. 3. A conceptual representation of an electrolyzer model considering the five figures of merit (i.e., the most commonly used lab-scale performance indicators in the 29 LT CO$_2$E TEAs), and the electrolyzer output. The diagram shows an electrolyzer with two main inputs, namely pure CO$_2$ and electricity, and the product yield as the output. Note: This is a simplified diagram to represent the five figures of merit. In reality, the performance parameters are not independent of each other and are influenced by many other factors. The five figures of merit are based on the following definitions [87]. Faradaic efficiency: the selectivity of reducing CO$_2$ toward a certain product. Current density: the amount of electric current flowing through the active area of the electrode at a given potential. Equipment size: cell dimensions in relation to the active area of the electrode. Durability: the lifetime of the electrolyzer. Energy efficiency: the ratio between energy stored in desired products vs. input electricity needed.

Table 1
Design requirements for industrial-scale CO$_2$ electrolyzers by Oloman et al. [68].

Characteristics	Hypotheses and thresholds
Mode of operation	Continuous
Current density	>100 mA/cm^2
Energy efficiency	<500 kWh/kmol formate/formic acid
Gas space velocity	>100/hour
Operating time	>4000 h/year
Operating conditions	P = 100–1000 kPa (abs) T = 283–373 K
Feed gas composition	Greater than 90 vol% CO₂ (dry basis); trace impurities less than 0.1 vol%
CO$_2$ conversion per pass	>10%
Product catholyte (formate/formic acid) concentration	>1 kmol/m^3
Capital cost (uninstalled)	<US$10 000 (2008)/m^2 geometric cathode area

consumption in the electrolyzer unit, which is a major OPEX factor affecting the overall TEA outcome.

Furthermore, temperature, pressure, and concentration gradients exist along the three dimensions of the electrochemical cell (i.e., length, width, and height) [91]. The gradients affect the rate and direction of the reactions. The current modeling efforts often consider a 0-D model in which the electrochemical conversion behavior in relation to the specific electrochemical reaction environment cannot be evaluated (e.g., the relationship between the electricity losses due to Ohmic resistances and the width of the cells). Consequently, the projected performance of the future industrial electrolyzer based on single-cell 0-D models could be overestimated.

Additionally, the single-cell 0-D electrolyzer models developed in LT CO$_2$E TEAs are often used to perform sensitivity analyses in which the current densities, Faradaic efficiencies, and potentials are altered (i.e., increased to values that are sometimes not yet possible at the experimental level) to determine initial break-even performance targets for several LT CO$_2$E-based products (for instance, positive net present values). For example, Jouny et al. [48] found that a current density threshold between 200 and 400 mA/cm^2 must be reached to reduce the overall plant capital cost and enable the economic viability for various LT CO₂E products (i.e., carbon monoxide, formic acid, methanol, ethanol, ethylene, n-propanol). The performance benchmarks determined by the three key TEAs [48,51,61] (e.g., $j > 200$ mA/cm^2, FE>90%) have often been adopted in the latter literature, used as established values in commercial plant scales, and possibly as indicative targets for the experimental community. However, these are subject to high uncertainty. Future LT CO₂E models that incorporate the fundamental knowledge regarding the reaction mechanisms behind the figures of merit for the specific target product will help overcome these limitations [92].

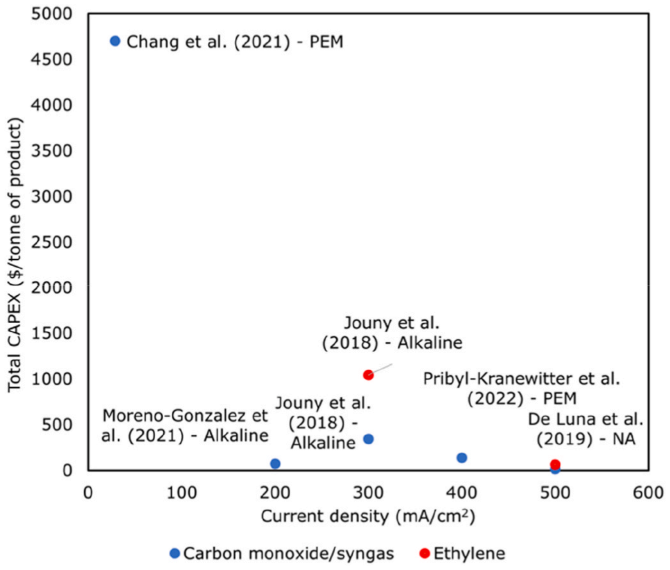

Fig. 4. Overview of the assumed current densities (mA/cm^2) plotted against the projected overall CAPEX of a future LT CO$_2$E plant (USD/tonne of product) for different LT CO$_2$E TEA papers. Comparisons focus on carbon monoxide studies with each other and ethylene studies with each other. Cost figures are represented in the currency (USD) and year used in each paper.

Improvements via multi-scale modeling: Experimental studies to support reactor design and advancements in operating conditions are already being accompanied by more in-depth modeling efforts. For instance, the models by Kas et al. [91] and Videla et al. [93] considered the various phenomena affecting the electrolyzer performance for the synthesis of carbon monoxide via the incorporation of multi-scale sub-models based on different theoretical models such as Maxwell-Stefan equations for dynamic mass transport and the Navier-Stokes equations for fluid dynamics.

The use of more extensive electrolysis modeling approaches (i.e., modeling the electrolyzer and the stack) is starting to gain attention for

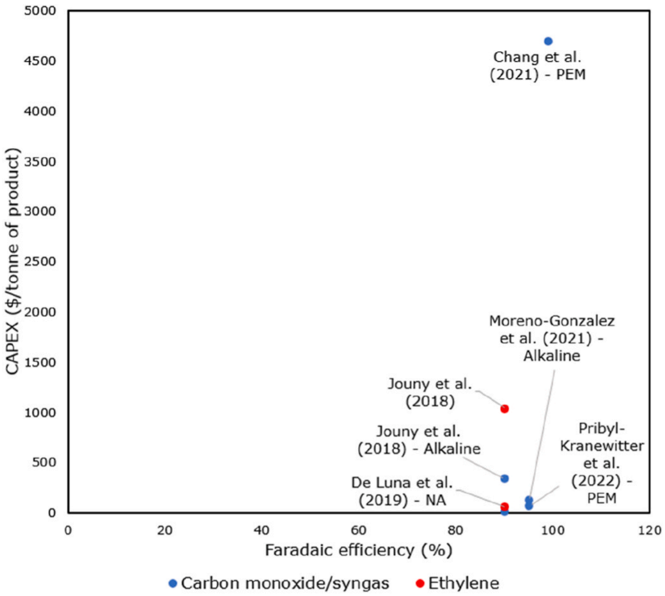

Fig. 5. Overview of the assumed Faradaic efficiencies (%) plotted against the projected overall CAPEX of a future LT CO_2E plant (USD/tonne of product) for different LT CO_2E TEA papers. Comparisons focus on carbon monoxide studies with each other and ethylene studies with each other. Cost figures are represented in the currency (USD) and year used in each paper.

LT CO_2E TEAs [22]. For example, Bagemihl et al. [86] proposed a pseudo 2-D modeling approach to capture the interconnections between the mass transfer effects and the reactor length. The work combines three levels, namely cell, electrolyzer, and plant level, to perform a techno-economic evaluation of a CO_2-to-ethylene plant. Using this model, it was found that an increase in CO_2 reacting with the electrolyte to bicarbonates (i.e., CO_2 cross-over effects) occurs at high current densities, causing the operation at lower current densities to be more advantageous. The outcome of this study indicated a possible lower optimal current density for ethylene ($\sim$100 mA/cm^2) than previously reported values in the three key TEAs (>200 mA/cm^2) [48,51,61]when optimizing the NPV of the plant.

Continuing the advancement towards more accurate electrolyzer models that capture the interdependency between the five figures of merit is only possible via multi-scale modeling efforts validated by experimental data. To achieve this, the working relationship between TEA modelers and electrolysis experimentalists needs closer collaboration. Interdisciplinary TEA teams working simultaneously on the cell, electrolyzer, and plant levels with ongoing data sharing and early feedback sessions between the modeling and experimental efforts will allow for the needed validation and refinement of electrolyzer models for LT CO_2E TEA research.

Lifetime of the electrolyzer unit: The overall plant CAPEX estimation is significantly influenced by the lifetime of the electrolyzer unit. Most TEAs discuss potential lifetimes of around 10,000 h for CO_2 electrolyzers due to catalyst degradation [48], but achieving stability beyond 190 h remains a major technical challenge [47], requiring, for instance, accelerated aging tests alongside long-term stability tests to obtain realistic lifetime estimations working under (flexible) conditions. Lessons from the chlor-alkali industry indicate the electrocatalyst needs to be replaced every 5–7 years [94,95] and also stated in the TEA by Shin et al. [79]. Although the potential effects of frequent replacement of the electrolyzer on the overall economic viability of the future LT CO_2E plant are usually mentioned, they are often not considered in the cost estimations in the TEAs. It is usually unclear in the current LT CO_2E plant CAPEX estimations whether or how the additional investment costs are considered within the methodological assumptions (e.g., as a recurring CAPEX or as a contributing item to the OPEX).

3.3. Process design for future LT CO_2E plants

The analysis of the 29 LT CO_2E process flow diagrams (PFDs) reveals that the impact of the upstream pre-treatment of CO_2 and water and downstream product purification sections have been considered at three different levels of detail, 1) no purification or separation steps, 2) ideal separator blocks with 100% efficiency, or 3) in-depth modeling of multiple equipment units. Focusing on the design and performance of the overall process, instead of only the electrochemical reactor, even if it is the core unit operation, is key to moving away from the preliminary conceptual phase and continuing the process development for LT CO_2E. Additionally, future commercialization of LT CO_2E will likely require plant designs integrating storage solutions (e.g., tanks or batteries) to allow for coupling with intermittent renewable electricity sources, as well as heat integration, recycling, and waste valorization, all aspects not yet included in the current LT CO_2E PFDs.

The evolution of the process flow diagrams: The first LT CO_2E TEA by Oloman et al. [68], as well as others [49,50], focused entirely on the electrolyzer unit. Oloman's initial process flow diagram considered the electrolyzer in combination with mixers and separators that assume 100% efficiency at each process step [68]. Agarwal et al. [55] expanded the process design of the TEA by Oloman et al. [68] by including CO_2 capture and performing a value chain analysis (i.e., a gate-to-gate TEA of a CO_2 to formic acid/formate plant). The works by Pletcher and Keets et al. [96,69] further expanded the analysis by coupling it with renewable electricity (no yet considering storage tanks or batteries to deal with the intermittency of renewable electricity sources) and considering other CO_2E-based products (e.g., methanol).

The TEAs by Jouny et al., Spurgeon et al., and Verma et al. [48,51,61] and the majority of TEAs developed after 2018 often include 1) a CO_2 capture process considering monoethanolamine (MEA) or direct air capture (DAC), 2) a 0-D electrolysis model, and 3) a product separation step based on either pressure-swing adsorption (PSA) or distillation (Tables S3 and S4 in the SI). Moreover, significant differences in assumptions regarding the CO_2 and electricity costs were found between TEA studies. Yet, most LT CO_2E TEAs conclude in their sensitivity analyses that the CO_2 and electricity costs are the main factors influencing the overall plant OPEX [51,54,77].

A comparison of the OPEX shares related to CO_2 and electricity costs was made to determine whether their impact on the overall plant OPEX is within similar ranges across studies. Fig. 6 highlights that for five

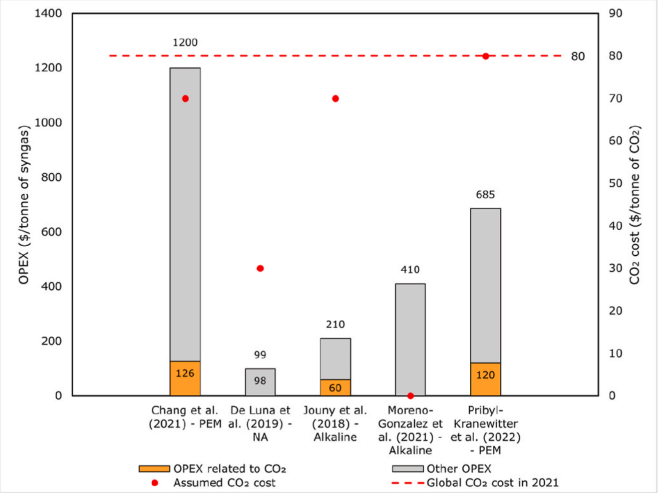

Fig. 6. Overview of the assumed CO_2 costs (USD/tonne of CO_2) against the projected total OPEX of a future LT CO_2E plant producing syngas (USD/tonne of syngas). The global CO_2 cost in 2021 was added as a reference based on data from the World Bank [97]. Note that the CO_2 cost and OPEX data are represented in the currency (USD) and year used in each paper.

TEAs studying syngas, the OPEX portion related to CO_2 costs varies between 0 (CO_2 at no cost) and 28% of the overall plant OPEX; further details are discussed in the upcoming section. A more striking result is found in Fig. 7, showing that the OPEX share related to electricity costs ranges between 18 and 81% of the total OPEX; more details to follow. Although variations in input assumptions, such as CO_2 and electricity costs, CO_2 conversion rates, and energy efficiencies, can explain some discrepancies, significant differences in OPEX shares between TEAs suggest that the influence of the CO_2 and electricity cost on the future plant OPEX remain unclear, hindering progress towards targeted LT CO_2E research. Moreover, as noted in Section 3.1 regarding the family tree, the studies differ in assumptions regarding electrolyzer design (e.g., PEM, alkaline). Yet again, the results in Figs. 6 and 7 reveal no robust differences in shares of OPEX related to CO_2 and electricity between PEM and alkaline electrolyzer types.

Pre-treatment of CO_2: The majority of TEAs developed between 2018 and 2022 consider coal-fired power plant or biobased plant flue gases as CO_2 point sources. Most include a CO_2 capture process considering monoethanolamine (MEA) based on the works by Raksajati et al., Finkenrath, and Rubin et al. [99–101]. Direct air capture (DAC) is usually considered based on Zeman, Keith et al., and Sanz-Pérez et al. [102–104]. The detailed composition of the captured CO_2 stream is often disregarded, and the CO_2 stream is usually described as "pure" [50, 54,75,76,105,106], or "concentrated" [48,73,77]. Major flue gas impurities (e.g., SO_x, NO_x, carbon monoxide, methane, hydrogen, nitrogen, metals) are mentioned by several studies [54,76,77,80,106,107] but their impact is not taken into account in their calculations. CO_2 from DAC can be relatively pure (with traces of nitrogen and oxygen) [104]. Only two of the analyzed papers specified the concentrations of major contaminants in the CO_2 feed [74,107], but did not include cleaning units in the PFD. The full compositions of the CO_2 feed stream consisting of both major and trace impurities are described, for instance, in the National Academies of Sciences, Engineering, and Medicine (NASEM) report [108].

The potential effects of impurities on the durability of the CO_2E unit have often been mentioned but not considered in most TEAs. Impurities can have three possible effects on the CO_2 electrolyzer: 1) act as diluents (e.g., nitrogen), 2) act as reducible species (e.g., oxygen), and 3) act as catalyst poisoning species (e.g., NO_2, SO_2, H_2S) [105,109]. Pappijn et al. [105] and Ramdin et al. [57] indicated that experimental studies have shown that even slight impurities can alter the catalytic performance significantly or deactivate the catalyst entirely. To scale up CO_2E for industrial applications, Pappijn et al. [105] mentioned that the effect of impurities on the electrolyzer performance needs further experimental investigation. Other studies [48,57,77,80,110] have remarked that captured industrial flue gases cannot be directly used in the CO_2 electrolyzer, but will require purification steps beforehand. However, the types of cleaning steps required were not further specified or modeled. Neither was the possibility of assuming more polluted streams at the expense of, for instance, a lower lifetime of the CO_2 electrolyzer (i.e., resulting in higher capital costs).

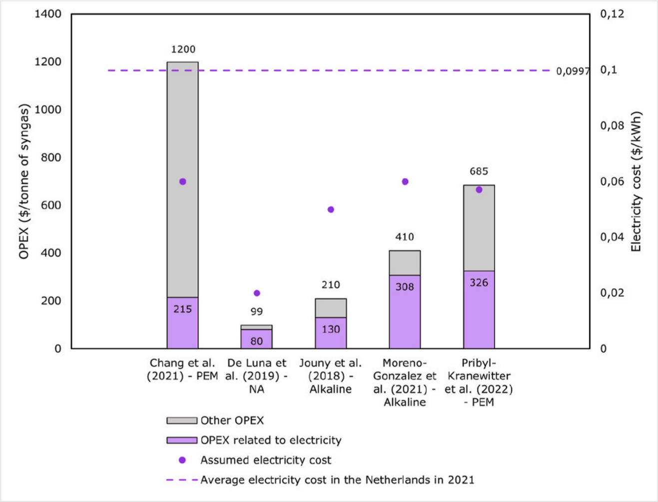

Fig. 7. Overview of the assumed electricity costs (USD/kWh) against the projected total OPEX of a future LT CO_2E plant producing syngas (USD/tonne of syngas). The average electricity cost in the Netherlands in 2021 was added as a reference based on data from Eurostat [98]. Note that the electricity cost and OPEX data are represented in the currency (USD) and year used in each paper.

In some studies, CO_2 is considered an operating cost (the price of a feedstock), while in others, the CO_2 capture unit is included in the cost estimations (affecting both the CAPEX and OPEX). A compilation of the CO_2 prices in the analyzed TEAs shows that prices lie between 0 and 80 USD/tonne [19,48,57,73,106] (comparison available in Fig. 6 for papers considering the synthesis of syngas and Table S3 in the SI). Barecka et al. [74] mentioned that including separation steps to reach the required purity will be costly and that reducing costs for carbon capture processes will be challenging. They highlighted that the most used CO_2 price of ~40 USD/tonne may be too optimistic.

Other works, such as Pappijn et al. [105], considered a negative CO_2 price of minus 30 USD/tonne, indicating that they assumed the LT CO_2E plant would get paid for handling the CO_2. This assumption implicitly means that the study considers the LT CO_2E plant as a waste handling plant instead of a chemicals production plant. It is important to note that considering the CO_2 electrolysis plant as a CO_2 waste management strategy, will not be possible, for instance, under the current Emission Trade System (ETS) regulation [111]. For the cases treating CO_2E plants as a waste management strategy, the CO_2 needs to be permanently disposed of, which cannot be claimed when the CO_2 is used to produce chemicals [111]. Therefore, using a negative CO_2 price to offset the OPEX is no longer a relevant assumption for LT CO_2E TEAs.

A different issue that calls attention is the use of different names for CO_2 prices, i.e. "CO_2 price" [74,112], "carbon tax" [113], "carbon emission cost" [51], "cost of CO_2 avoided" [77] or "carbon credits" [51, 68]. It is, however, generally not explained what the CO_2 price includes in most TEAs. As a result, it is unclear whether the CO_2 price is considered to be the price paid to the CO_2 capture facility per tonne of captured CO_2 [19,48,57,73,106] or the price that the CO_2 source would need to pay if the CO_2 is not captured. Additionally, it is unclear whether the price includes the costs for transportation of CO_2.

The outcomes of studies regarding the influence of the CO_2 price on the overall techno-economic performance are also very different. Several studies pinpointed that the CO_2 price has a limited impact on the economics of the process for methanol [82], ethylene [105], carbon monoxide and formic acid/formate [114]. Whereas, Rumayor et al. [54] and Spurgeon et al. [51] stated that a carbon emission cost is essential to making CO_2E processes profitable for syngas, formic acid/formate, and ethanol [51,54] (which is in contradiction with the current ETS regulation [111]). Huang et al. further specified that only processes with products that require the least amount of electrons and those with high energy efficiencies are sensitive to changes in CO_2 price, while products requiring more electrons are more sensitive to electricity price changes [77].

Pre-treatment of water: The purity of water, as the purity of CO_2, is also crucial for LT CO_2E. Typically, water fed in experimental electrolysis is ultra-pure water [115]. However, in the TEAs, a lack of specification exists regarding the water type (i.e., demineralized, deionized, ultra-pure), the purity requirement (i.e., no-go contaminants), as well as the costs related to water purification units. The study by Roh et al. [116] mentioned that deionized process water is supplied to the electrolyzer and included the water purification units in the process flow diagram. Yet, the CAPEX and OPEX of water purification were excluded [116].

Water electrolysis and the chlor-alkali industry typically use water from a water pre-treatment unit located within the plant gate. Therefore, the units required for the pre-treatment of water for CO_2E should also be included in the PFDs, and their CAPEX and OPEX considered in the TEA. As an example, a 1 GW water electrolysis study by the Dutch Institute for Sustainable Process Technology (ISPT) [15] considered that the demineralized water will be produced via reverse osmosis within the plant gate for an alkaline system, while the ultra-pure water needed for a PEM will be produced through continuous polishing using ion-exchange resins (i.e. additional plant CAPEX and OPEX).

Downstream processing: The downstream processing steps have been studied at various levels of detail throughout the 29 LT CO_2E TEAs (Table S4 in the SI). Studies by Greenblatt et al., Jouny et al., Chang et al., Sisler et al., de Luna, and Ramdin et al. [48,50,53,62,117,118] have highlighted the complexity of downstream separation of products from by-products and wastes, such as unreacted CO_2 and electrolytes, and how different electrolyzer designs (e.g., alkaline vs. catholyte-free) will each require unique separations. Even if Faradaic efficiencies and single-pass conversions continue to improve beyond 50% [19,50], realistic downstream processing will remain key for the commercial application of CO_2E (e.g., due to the continued need for recirculation of the electrolytes [119]).

Key differences between the TEAs lie in the type of CO_2E products studied and the separation units assumed for downstream processing. As an example, three key TEAs are discussed here. Spurgeon et al. [51] focused on the production of liquid products via both a direct (i.e., CO_2E to ethanol and formic acid with a CO_2/CO gas separation via PSA considering scaling relationships based on Li et al. [78]) and an indirect route (i.e., CO_2E to obtain syngas followed by a Fischer-Tropsch and naphtha upgrading unit using scaling relationships based on Liu et al. [120]). Jouny et al. [48] considered products formed via the direct CO_2E route (i.e., carbon monoxide, formic acid, ethylene, propanol, methanol, and ethanol) and gas separation through a PSA. The PSA-scaling relationships for cost estimation were based on the work by Paturska et al. [121,122], which compared different biogas upgrading technologies (with a different gas composition than the one downstream of the CO_2E). Verma et al. [61] estimated the product separation costs for carbon monoxide, formic acid/formate, methanol, methane, ethanol, and ethylene using Sherwood plots, which correlate the separation costs inversely to the product concentration in dilute streams without mentioning the types of separation equipment used.

The CAPEX and OPEX of the future LT CO_2E plant will depend on the required purity of the products to be sold. Yet, most TEAs do not mention the required purity of their main product(s) nor of any by-products that are assumed to be sold, even though the price of chemicals changes based on the purity grade (i.e., quality level), reflecting the difficulty and costs of the separation steps involved [63]. Contamination limits or allowances related to environmental regulations of the waste streams (e.g., temperatures, pH levels, impurities) are not specified in any of the revised TEA papers. Moreover, the upgrading of by-products, electrolyte recycling, waste handling and conditioning, as well as heat integration are usually excluded from the analysis. The impact of the downstream processing units on the techno-economics of future LT CO_2E plants remains unclear and needs further investigation. Additional information is available in comparison Table S4 in the SI.

Electricity & flexibility: Most LT CO_2E TEA models assumed that future industrial-scale CO_2E plants will be powered with intermittent renewable electricity (i.e., wind and solar). It is often highlighted that electricity will be a major OPEX component, and when varied in sensitivity analyses, it is generally assumed that future prices of renewables will be cheaper than the current price of grid electricity (Fig. 7). However, water electrolysis research highlights that calculating electrolysis-based production costs solely based on the (expected) annual average electricity cost by region oversimplifies electricity expenses and overlooks location-specific conditions or utility tariff structures (i.e., industrial and commercial electric utility rates) [123]. Moreover, lessons from other electrolysis-based technologies indicate an economic tradeoff between oversizing the plant capacity (i.e., higher CAPEX) vs. dynamic operation (i.e., varying the electrolyzer power loads to benefit from the price volatility of intermittent renewable electricity) [124–126]. Therefore, future TEA models should increasingly consider design and operation in a unified way.

Our analysis of the process flow diagrams in the current LT CO_2E TEAs (Section 3.3) found that most plants are designed for continuous operation as they do not yet take into account how the connection between a future LT CO_2E plant and an intermittent renewable electricity source will affect the overall plant scale, process design, CAPEX, and OPEX (e.g., the installation of batteries, or any other type of electricity or material storage solutions [127,128]). Moreover, the TEAs that include downstream processing often consider thermochemical separation equipment (i.e., distillation units) which are designed for continuous operation and limit the flexibility of the overall plant. Additionally, the electrochemical plant will require rectifiers (i.e., AC to DC transformers), and the flexibility of this equipment is also still unknown [15,129,130]. The rectifier units were not yet mentioned or included in the 29 LT CO_2E TEAs. As an illustration, the projected cost contribution related to the power supply and electronics was found to be ~17% of the bare equipment costs in the Dutch ISPT 1 GW water electrolysis plant study [15].

To move the field forward, LT CO_2E TEAs should increasingly consider 1) the dynamic operation of the electrolyzer unit (e.g., range of current density of operation or a maximum current density, and modularity) and 2) a plant designed to accommodate flexibility, due to its importance on the overall process economics (e.g., as is starting to be done in the HT CO_2E field [131]). As previously illustrated in the multi-scale modeling section (Section 3.2), as we move toward higher TRLs with time, more refined TEAs modeling the cell, stack, and plant levels simultaneously in close collaboration with experimentalists will be a necessary step to capture flexibility tradeoffs that traditional TEAs cannot grasp.

4. Conclusions

A systematic literature review was performed to investigate the methodological assumptions used in 29 techno-economic assessments (TEAs) for low-temperature CO_2 electrolysis (LT CO_2E). This work aimed to study the origins and justifications behind the methods to pinpoint which modeling and input assumptions need further improvement to ensure that only the highest quality and updated TEAs support decision-making in the LT CO_2E field as it moves to higher technology readiness levels (TRLs). Three main takeaways were found as a result.

Firstly, the current methodological justifications seem stuck in the past due to the continuation of modeling and input assumptions established by a limited number of studies and LT electrolysis-based technologies from previous decades. The fast-paced knowledge evolution in both HT and LT electrolysis calls for a reexamination of whether the previously employed assumptions should still form the basis of the modeling and input assumptions for LT CO_2E TEAs moving forward, considering the best available information today.

Secondly, previously held LT CO_2E techno-economic performance benchmarks derived using single-cell reactor electrolyzer models with linear cost-performance relationships and independent consideration of the five figures of merit do not align with the latest TEAs using more extensive modeling approaches. Given that the results of TEAs significantly influence fundamental electrolyzer design decisions and set performance benchmarks for the experimental community, it is imperative to proceed using the most advanced electrolyzer modeling approaches available. At the same time, the limitations of the TEA outcomes should be clearly defined alongside the TEA results to grasp under which context the insights are meaningful.

Finally, the majority of LT CO_2E TEAs are in the preliminary conceptual phase, mainly considering only the CO_2 electrolysis unit. To advance toward more in-depth process designs, future LT CO_2E TEAs should include the pre-treatment of CO_2 and water, the latest advancements in electrolyzer modeling approaches and input data, product separation steps, as well as heat integration, recycling, and waste valorization. Only then can LT CO_2E TEAs build on past efforts and ensure that the latest advancements and highest quality information are used to effectively guide the research and development efforts required for LT CO_2E to advance to a higher TRL.

CRediT authorship contribution statement

Josephine Vos: Conceptualization, Methodology, Software, Formal analysis, Investigation, Writing – original draft, Visualization. **Andrea Ramírez:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Mar Pérez-Fortes:** Conceptualization, Methodology, Writing – review & editing, Supervision, Resources, Project administration.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2025.115454.

Data availability

The systematic literature review data and analysis of low-temperature CO_2 electrolysis techno-economic assessments are available as Supplementary Information (SI) and can be accessed on Zenodo: https://doi.org/10.5281/zenodo.14604472.

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