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Carbon footprint assessment of ethylene oxide production *via* CO_2 electrolysis†

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Electrochemical CO_2 reduction to value-added chemicals is a promising carbon capture and utilization pathway for decarbonizing the chemicals sector. Recent advances demonstrate production of ethylene oxide—a major commodity chemical—from CO_2 *via* a tandem electrocatalysis approach with ethylene as an intermediate. This study evaluates for the first time, the carbon footprint of ethylene oxide produced by this emerging technology under various conditions. We estimate the cradle-to-gate carbon footprint to range from −2.6 to 10.2 tonnes CO_2-eq. per tonne ethylene oxide depending on the electricity supply (from zero-carbon to the 2022 Canadian average grid mix of 0.128 kg CO_2-eq. per kW h), compared to 2.32 tonnes CO_2-eq. per tonne for conventional fossil-based ethylene oxide. Negative values indicate the process emits less CO_2-eq. than captured from the CO_2 feedstock source, without implying net atmospheric CO_2 removal. Scenario analysis shows large-scale deployment could achieve emissions savings over conventional production if the process performance improves by at least 50% and is powered by low carbon electricity (<0.06 kg CO_2-eq. per kW h). We determine *via* sensitivity analyses that, in order of priority, technology improvements should focus on (i) increasing the CO_2 to ethylene faradaic efficiency, (ii) reducing the energy demand for ethylene electrosynthesis and (iii) enhancing the CO_2 single pass conversion efficiency. An examination of safety considerations highlights that the emerging pathway could enable ethylene oxide production with higher yields and safer operating conditions than the conventional pathway. This work provides valuable insights into the carbon reduction potential and development priorities for CO_2 electrolysis-based ethylene oxide, supporting efforts to decarbonize the chemicals sector.

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Sustainability spotlight

CO_2 electrolysis is an emerging technology that relies on electricity to convert CO_2 into chemicals and fuels, offering a pathway to reduce greenhouse gas (GHG) emissions in the chemical sector. Ethylene oxide, a key commodity chemical used in plastics and consumer goods, is conventionally produced from fossil fuels with high emissions. This study evaluates the carbon footprint of ethylene oxide production *via* CO_2 electrolysis, identifying key improvements—such as reducing energy demand and enhancing faradaic efficiency—to maximize its environmental benefits. By providing guidance for technology developers, this work supports the transition to sustainable chemical manufacturing, and aligns with UN Sustainable Development Goals (SDGs) 9 (Industry, Innovation, and Infrastructure) and 13 (Climate Action).

1 Introduction

Decarbonizing the chemical sector is crucial for achieving net-zero targets, as it accounts for 20% of industrial emissions and nearly 6% of total global greenhouse gas (GHG) emissions.[1,2] A notable contributor to these emissions is ethylene oxide, a major commodity chemical with a 2022 market size of US$ 54 billion, widely used as a precursor for chemicals and plastics production.[3,4] With an annual global production of 32 million metric tons, ethylene oxide production emits over 74 million metric tons of CO_2 equivalent (CO_2-eq.) per year, contributing to 2% of the chemicals sector's emissions.[1,5] Currently, commercial ethylene oxide production involves epoxidation of ethylene in air or oxygen over a silver-based catalyst and generates substantial CO_2 emissions due to the reliance on fossil feedstocks and over-oxidation of ethylene and ethylene oxide.[6]

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Carbon capture and utilization (CCU) technologies, which capture CO_2 from the air or from point-source emissions for use as a feedstock, present a promising pathway to decarbonize the chemicals industry. Electrochemical reduction of CO_2 (CO_2R) is a CCU strategy that offers the advantage of mild reaction operating conditions and potential integration with renewable electricity generation.[7] Recently, lab-scale electrochemical production of ethylene oxide from CO_2, water, and renewable electricity has been reported,[8,9] suggesting the potential for lower emissions than the commercial process.

Like ethylene oxide production *via* CO_2R, many CO_2R processes remain at early stages,[10,11] with large-scale implementation impeded by challenges in achieving high product selectivity, costly high purity CO_2 feed, and poor CO_2 conversion efficiency.[12–14] High product selectivity (*i.e.*, low generation of undesired products) reduces downstream separation energy requirements and costs.[15,16] High purity CO_2 feed is necessary for stable and efficient CO_2 conversion,[17] while poor CO_2 conversion efficiency results in considerable unreacted CO_2, leading to cost and energy intensive downstream product separations.[16] Consequently, most studies have focused on improving key performance drivers of CO_2R such as catalyst design, current density, faradaic efficiency, energy efficiency, and electrolyzer size and configuration, to address these challenges.[7,10,14,18] However there is a scarcity of environmental impact studies due to the low technology readiness levels of CO_2R processes, limiting our understanding of their environmental impacts.[19,20] It is important to understand the environmental impacts associated with unique CO_2 electrolysis processes to aid effective comparisons and make informed decisions about technology deployment policies.

Existing life cycle assessments (LCA) of CO_2R technologies have shown potential GHG reductions of up to 80% compared to conventional fossil-based routes for various chemicals,[15,21–24] and have emphasized the importance of renewable electricity in ensuring reduced GHG emissions in CO_2R processes. However, few studies have specifically assessed the environmental impacts of emerging ethylene oxide technologies.[25–29] Among these include life cycle assessments on ethylene oxide produced *via* CO_2 oxidation with mixed metal oxides[25] and *via* reaction of ethylene with H_2O_2.[26,27] Yoon *et al.*, studied costs of ethylene oxide produced *via* CO_2R and the associated gate-to-gate CO_2 footprint.[29] Recently, Rodin *et al.*, assessed ethylene oxide production *via* electrochemically produced ethylene and hydrogen peroxide, analyzing how electricity supply (wind, PV, grid mix) and CO_2 single pass conversion efficiency influence the carbon footprint of ethylene oxide.[26] However, the study did not evaluate other CO_2R performance parameters such as faradaic efficiency (FE) and ethylene conversion rate, nor consider potential changes in the technology performance at scale, which is important for processes at low TRLs given the uncertainties of their commercial-scale deployment.[19]

This study assesses ethylene oxide produced *via* an emerging CO_2R pathway that relies on a tandem electrocatalysis approach in acidic media. We identify key factors required for the pathway to be attractive from a carbon footprint perspective compared to conventional fossil-based production, with the aim of assessing its potential to reduce emissions in the chemicals sector. We perform detailed sensitivity and scenario analyses to identify key drivers of process emissions and explore the relationship between critical CO_2R performance parameters.

Additionally, we evaluate the technology's potential performance at projected industrial scale, compare its carbon footprint with that of alternative ethylene oxide production routes, and propose performance thresholds to guide the development priorities for CO_2R-based ethylene oxide. The novelty of this work lies in estimating the carbon footprint of this pathway for the first time. Additional novel contributions include estimating the carbon footprint under various electrolytic and background system conditions, employing scenario analysis on the interaction of key CO_2R performance parameters and proposing quantitative targets for the parameters at different electricity grid GHG intensities. Due to its reactive nature and several past explosion incidents in conventional production, the process safety of ethylene oxide production is of importance. Therefore, we also discuss the process safety considerations of the emerging CO_2R to ethylene oxide pathway.

2 Methods

The carbon footprint of a product refers to the total amount of GHG emissions and GHG removals in the product system. The carbon footprint in this study is determined based on a life cycle assessment using the single impact category of climate change, in accordance with the International Organization for Standardization (ISO) 14040/14044 and 14067 guidelines for life cycle assessment (LCA) and carbon footprint of products, respectively.[30,31] The LCA guidelines for CO_2 utilization, developed by the Global CO_2 Initiative are also used as a reference.[32]

2.1. Goal and scope

The goal of this study is to determine the carbon footprint of ethylene oxide produced *via* an emerging acidic media-based electrochemical CO_2 reduction (CO_2R) technology. We compare this to the conventional ethylene oxide production pathway and an alternative route, and identify conditions in which the emerging CO_2R to ethylene oxide pathway is attractive from a carbon footprint perspective.

A cradle-to-gate scope is applied for the carbon footprint study. A cradle-to-gate scope is recommended in assessments of products that have identical composition and technical performance with conventional counterparts, as the products are unable to be differentiated, and therefore can be assumed to have identical downstream activities and associated emissions.[33] Given the cradle-to-gate scope of the assessment, the use and end-of-life phases of the ethylene oxide are not included in the system boundaries, as the ethylene oxide produced *via* the emerging CO_2R to ethylene oxide pathway is assumed to be identical to ethylene oxide produced in the other pathways. The system boundaries assessed within this scope include upstream carbon capture and purification, the core CO_2 conversion process (ethylene electrosynthesis from CO_2 followed by

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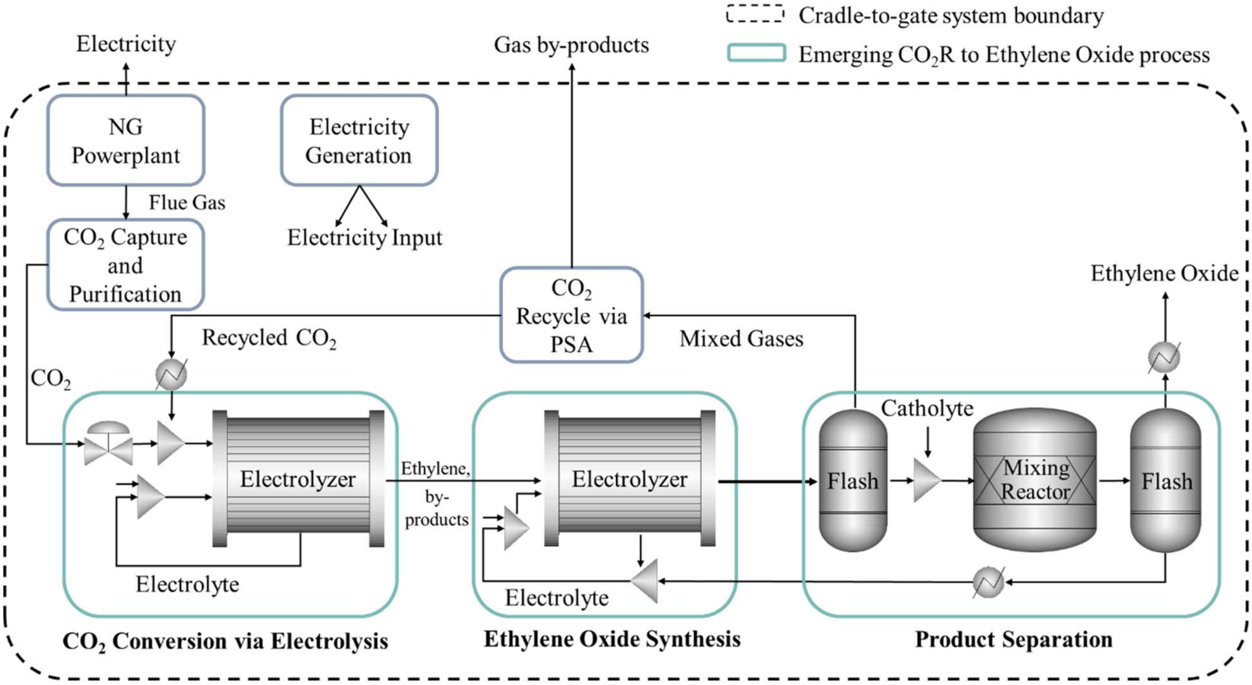

Fig. 1 Cradle-to-gate system boundary of the assessed emerging CO_2R to ethylene oxide technology. The CO_2 feedstock is assumed to be captured from a natural gas (NG) combined-cycle power plant. Liquid by-products from CO_2 reduction are assumed to be accumulated and recycled within the electrolyte at minimal concentrations. Emissions associated with the use phase and end-of-life of ethylene oxide are not included in the system boundary as they are assumed to be identical for the emerging process and alternative ethylene oxide production pathways.

ethylene oxide electrosynthesis from ethylene), and downstream ethylene oxide separation and purification. Fig. 1 shows the cradle-to-gate system boundary used in the assessment.

The carbon footprint is determined for the functional unit of 1 metric ton (t) of ethylene oxide produced *via* CO_2 electrochemical reduction. A mass-based functional unit is recommended for chemicals, such as ethylene oxide, that are traded on a mass basis.[33] In the base-case scenario, the electricity for the process is assumed to be supplied by the 2022 Canadian average electricity grid mix (where the technology is developed) with GHG intensity of 0.128 kg CO_2-eq. per kW h.[34] In the sensitivity and scenario analyses, the GHG intensity of the electricity is varied, ranging from 0 to 0.194 kg CO_2-eq. per kW h, representing lower- and higher-GHG intensity electricity grids in locations where the technology could be deployed (see Section 2.5 and 2.6).

2.2. Process system description

2.2.1. CO_2 capture. The CO_2 feed to the conversion process is assumed to be captured and concentrated from a natural gas combined-cycle power plant *via* monoethanolamine (MEA)-based post combustion capture.[35] CO_2 capture from power plants for chemicals production has the potential to simultaneously decarbonize the power and chemicals sectors by diverting potential CO_2 emissions for use as a replacement for fossil-derived raw materials used in chemicals production.[36] We examine the influence of the CO_2 feedstock source on the carbon footprint using a sensitivity analysis. The energy required for capturing the CO_2 is included in the process boundary, while the transportation and storage of the CO_2 are excluded, based on the assumption that the ethylene oxide production gate is in close proximity to the CO_2 capture facility.

2.2.2. CO_2 conversion and ethylene oxide synthesis. Several configurations exist for CO_2 electrochemical reduction to ethylene and ethylene oxide; however, the ethylene oxide assessed in this study is produced using a tandem electrocatalysis approach in acidic media. This process, currently at laboratory scale, has been previously described by the authors,[8,9,37] with additional support from recent experimental data. In summary, ethylene oxide production occurs *via* a two-step electrosynthesis process in which ethylene is first synthesized *via* CO_2 reduction in a liquid electrolyte flow cell, followed by ethylene oxide synthesis in a second flow cell (see ESI† for chemical reactions and a detailed process description). The unreacted CO_2 is separated *via* pressure swing adsorption (PSA), which is assumed to recover 90% of the unreacted CO_2.[16]

2.3. Life cycle inventory analysis

As the technology is at an early stage, data used in the assessment include extrapolations from experimental data and estimations based on process modeling and experiments. A data quality matrix classifying the data used in the base-case model, sensitivity analysis and scenario analysis is shown in the ESI.†

The material and energy data for the inventory analysis are obtained from primary laboratory experiments.[9,37,38] We also develop a Microsoft Excel model to perform stoichiometric

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calculations and a material balance on the process to obtain the CO_2 feedstock requirement and byproduct generation rates. To calculate the energy demand for separation and purification of ethylene oxide, we model the process in Aspen Plus™. Emission factors used in the assessment are obtained from literature while data for the conventional ethylene oxide process are obtained from the Carbon Minds[39] and Ecoinvent v3.8 databases.[40] Table 1 shows the process parameters used in the study and their base-case values. A more detailed description of the data for the sensitivity and scenario analyses is provided in Section 2.5 and 2.6.

2.4. Carbon footprint calculation

The process inputs are aggregated on the basis of the functional unit, *i.e.*, 1 metric ton (t) of ethylene oxide, and the resulting carbon footprint is calculated using GHG emission factors obtained from literature (Table 1) and the Intergovernmental Panel on Climate Change 100-year global warming potentials (GWP 100) based on the AR6 synthesis report.[43] The global warming impact (GWI) estimated for the carbon footprint assessment is determined based on the GHG intensity of capturing and concentrating CO_2 from industrial flue gas, the electrical energy demand for electrosynthesis of ethylene and ethylene oxide, the energy demand for ethylene oxide separation and CO_2 recycle, and the venting of unrecycled CO_2 and waste gases. The emissions associated with construction of the electrolyzer, catalyst and other equipment are not included in the analysis due to limitations on data availability; however, earlier assessments have found these emissions to be negligible compared to the core process phase emissions.[13,15] The impact of water used as a raw material in CO_2R has been found to be negligible in several studies also using CO_2 from point source emissions as a feedstock,[15,44] so we exclude it from the inventory; however, we assess the impact of the concentration of water in the product stream on the separation energy, and thus the GWI.

In the base-case assessment, we allocate all energy inputs and other emissions associated with the conversion and separation processes to the intended product, ethylene oxide. We also perform mass- and energy-based allocations to demonstrate the sensitivity of the carbon footprint to the allocation method. We treat the CO_2 feedstock as a carbon credit—as it would otherwise be emitted from the natural gas power plant from which it was captured—and allocate this credit to all products on a mass basis. The emissions associated with the CO_2 capture unit (due to infrastructure and energy requirements) are also allocated to the products of the CO_2R to ethylene oxide process on a mass basis. Based on our assessment approach, a negative GWI result does not indicate that physical removal of emissions from the atmosphere occurs, but rather, that the process emits less CO_2-eq. than was captured from the point source, *i.e.*, that emissions are avoided because of the emerging CO_2R to ethylene oxide technology.

2.5. Sensitivity analysis

We conduct a sensitivity analysis to understand the influence of key model variables on the GWI of ethylene oxide and identify critical areas for emissions reductions within the process. The parameters for the sensitivity analysis are selected based on the results of the base-case assessment (that is, parameters found to be key contributors to the GWI in the base-case) and based on parameters shown to be critical to CO_2 electrolysis in prior literature studies. In the sensitivity analysis, one parameter was varied at a time while all other independent parameters were held constant. As the technology is still at an early stage, we employ an arbitrary variation of ± 50% for the chosen parameters to assess how sensitive the GWI is to each parameter. This variation may not fully capture the uncertainties associated with the parameters; however, it enables us to identify areas for further technology development. Following the sensitivity analysis, we identify the parameters most influential to the GWI and calculate the quantitative thresholds to be maintained for

Table 1 Key process parameters used in the study and their base-case values

Phase	Parameter	Value[a]	Units	References
Carbon capture	CO_2 capture emission factor	0.68^{3}	kg CO_2-eq. per kg CO_2	35
	Carbon capture rate	90^{3}	%	41
	CO_2 feedstock input	8.21^{2}	CO_2-eq. per t ethylene oxide	Calculated
Ethylene synthesis	Ethylene electrosynthesis energy demand	$73\,000^{1}$	kW h per t ethylene oxide	Calculated
	CO_2 SPCE	87^{1}	%	9
	Ethylene faradaic efficiency	47^{1}	%	9
	Full cell voltage	3^{1}	V	Author lab data
Ethylene oxide synthesis	Ethylene conversion rate	60^{1}	%	Author lab data
	Ethylene oxide faradaic efficiency	70^{1}	%	8
	Ethylene oxide electrosynthesis energy demand	4350^{1}	kW h per t ethylene oxide	Calculated
	Full cell voltage	2.5^{1}	V	8
Product separation	CO_2 recycle efficiency	90^{3}	%	15
	PSA energy demand	0.25^{3}	kW h m^{-3}	42
	Ethylene oxide separation energy demand	7686^{2}	kW h per t ethylene oxide	Calculated
	Electricity GHG intensity	0.128^{3}	kg CO_2-eq. per kW h	34

[a] Superscript numbers represent pedigree matrix data classification in Table S2.† SPCE: single pass conversion efficiency; PSA: pressure swing adsorption.

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Table 2 Parameter values for sensitivity analysis of the global warming impact (GWI) of the emerging CO_2R to ethylene oxide process[a]

Parameter	Low value[4]	Base value	High value[4]	Units
CO_2 capture emission factor	0.34	0.68	1.02	kg CO_2-eq. per kg CO_2
Ethylene electrosynthesis energy demand	36 500	73 000	109 500	kW h per t ethylene oxide
CO_2 SPCE	21.8	87	100	%
Ethylene faradaic efficiency	24	47	71	%
Ethylene conversion rate	30	60	90	%
Ethylene oxide electrosynthesis energy demand	2175	4350	6525	kW h per t ethylene oxide
PSA energy demand	0.13	0.25	0.38	kW h m^{-3}
Water ratio	1 : 1	2 : 1	3 : 1	mol : mol
Electricity GHG intensity	0.064	0.128	0.192	kg CO_2-eq. per kW h

[a] Superscript numbers represent pedigree matrix data classification in Table S2.† See Table 1 for sources of base values. SPCE: single pass conversion efficiency; PSA: pressure swing adsorption.

these parameters for the GWI of the emerging process to be lower than that of the conventional process. The parameter values used for the sensitivity analysis are shown in Table 2.

For the sensitivity analysis, the target production of ethylene oxide is held constant while the raw material requirements are adjusted based on the changing parameters. The parameters are varied through an approach that ensures internal consistency in the CO_2 electrolysis process. In the case of the ethylene FE, when exploring higher and lower ethylene FE, the remaining FE is distributed among the byproducts based on the distribution of the FE at the base-case. However, this may not fully represent what might be observed at the experimental scale when the FE of ethylene is varied. The FE is influenced by multiple factors such as catalyst type, electrode design, *etc.*; therefore, the results of the sensitivity analysis are expected to serve as a starting point until further progress is made at laboratory scale.[7,18]

The CO_2 SPCE is changed while holding all other parameters constant, including the FE of all products.[45] CO_2 SPCE in electrochemical reduction has characteristically been reported to be low, ranging between 10–50%.[15,26,46] However, in the emerging pathway studied here, acidic conditions in the lab-scale experiments enable CO_2 SPCE as high as 87%.[9] As many CO_2R technologies are currently limited by poor CO_2 conversion,[14,15,26] we analyze a wider range of variation in the CO_2 SPCE, from 100% to as low as 21.8%.

The energy demand for electrosynthesis is a function of several parameters such as the cell voltage, FE, and number of electron transfers needed. For the sensitivity analysis, we assume an improvement or loss in performance by decreasing and increasing the energy demand, respectively, without making direct changes to the FE, or the voltage. Thus, the changes in electrosynthesis energy demand could be attributed to changes in either of these functions or additional process changes. In the case of varying the FE, the electrosynthesis energy demand is changed in response to the changing FE, while holding the cell voltage constant.

2.6. Ethylene oxide technology scale up and scenario analysis

As the emerging CO_2R to ethylene oxide technology is still at an early stage, a scenario analysis on the future performance of the technology at scale is recommended to reasonably compare it with the conventional pathway.[19,33] We scale the model to an annual production of 450 kt of ethylene oxide – the capacity of an existing ethylene oxide/ethylene glycol plant in Alberta, Canada – and perform a scenario analysis to identify conditions under which the emerging CO_2R to ethylene oxide pathway would be attractive from a carbon footprint perspective.

Three scenarios were initially analysed for the future performance of the technology at scale: an improved performance scenario, in which we assume a 50% improvement in performance of the process relative to the base-case; a reduced performance scenario, in which we assume a 50% decline in performance relative to the base-case due to scale up, and a maintained performance scenario, in which the scaled up process is assumed to perform identically to the base-case performance observed at lab scale. Due to the relationships among the model parameters, there are likely to be trade-offs between the performance of parameters. For example, prior studies showed that an improvement in the FE at a larger scale may be obtained only at the expense of the CO_2 SPCE due to difficulties related to reactant loss, larger flow directions and solubility limitations encountered at large scale.[45,47] However, prior assessments of environmental impacts of CO_2R technologies have varied the CO_2 SPCE and FE independently,[15,36,44] failing to capture how their trade-off influences the GWI. Therefore, to address this gap, we explore two additional scenarios: a high conversion rate/low FE scenario, and a high FE/low conversion rate scenario.

In the case of the high FE/low conversion rate scenario, we assume that both the CO_2 conversion and the ethylene conversion rate are low, while the FE of ethylene is high, and the corresponding energy demand for electrosynthesis is low (due to the increased FE). Likewise, in the case of the high conversion rate/low FE scenario, we assume that both the CO_2 conversion and the ethylene conversion rate are high, while the FE of ethylene is low, and the corresponding energy demand for electrosynthesis is high (due to the reduced FE). In these two scenarios, we assume a lower energy demand for ethylene electrosynthesis in the case of high FE, and a higher energy demand for ethylene electrosynthesis in the case of a lower FE, to ensure internal consistency in the scenarios. However, the

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Table 3 Values and performance scenarios analyzed in the scenario analysis of the CO_2 electrolysis process parameters[a]

Parameter	Improved^{4}	Maintained^{1}	Reduced^{4}	High FE/Low conversion rate^{4}	High conversion rate/Low FE^{4}	Units
Ethylene electrosynthesis energy demand	36 500	73 000	109 500	36 500	109 500	kW h per t ethylene oxide
CO_2 SPCE	95	87	55	55	95	%
Ethylene faradaic efficiency	70.5	47	23.5	70.5	23.5	%
Ethylene conversion rate	90	60	30	30	90	%
Ethylene oxide electrolysis energy demand	2175	4350	6525	4350	4350	kW h per t ethylene oxide

[a] Superscript numbers represent pedigree matrix data classification in Table S2.† SPCE: single pass conversion efficiency.

energy demand and FE for ethylene oxide synthesis is held constant in the two scenarios above as its impact on the GWI was found to be small relative to the other parameters varied in the scenario analysis.

With the exception of the maintained performance scenario, which is based on experimental data obtained at the laboratory scale, the data used to perform the scenario analysis are based on the authors' assumptions of the process performance using extrapolations of the experimental data. The values for the scenario analysis of the CO_2 electrolysis parameters are shown in Table 3.

As the emerging CO_2R to ethylene oxide process is expected to compete with other technologies in the future, it is recommended by Müller *et al.* to "define scenarios representing the status quo, a fully decarbonized future and a transition scenario for the energy and electricity inputs".[33] Therefore, we perform a scenario analysis on the background system by varying the electricity GHG intensity and the CO_2 feedstock source. We explore CO_2 from direct air capture (DAC), natural gas power plants (base-case), and bioethanol production *via* fermentation of corn, as these represent common CO_2 sources used in carbon capture and utilization pathways such as CO_2 electrochemical reduction.[48] Extended methods and results on the scenario analysis of the background system are presented in the ESI.†

2.7. Comparison with conventional and alternative ethylene oxide production pathway

We compare the GWI of the emerging CO_2R to ethylene oxide pathway with that of alternative ethylene oxide production pathways. The alternative pathways include conventional fossil-based ethylene oxide,[6] and a process developed by the Center for Environmentally Beneficial Catalysis (CEBC) at University of Kansas, which utilizes hydrogen peroxide as a liquid oxidizing agent to produce ethylene oxide from fossil-derived ethylene,[27] see ESI† for process descriptions.

For the comparison, we adjust the carbon footprint of the conventional ethylene oxide process to employ the base-case Canadian electricity grid mix used in modeling the emerging CO_2R to ethylene oxide process. We do not include the alternative pathways in the background scenario analysis where the electricity source is varied, as these pathways use minimal electricity (less than 2% of total emissions) in their processes.[27,40] Emissions from the conventional ethylene oxide pathway are largely due to direct CO_2 emissions as well as fossil-based ethylene synthesis. In the CEBC-ethylene oxide pathway, 60% of the emissions are contributed by H_2O_2 production.

3 Results and discussion

3.1. Base-case carbon footprint results for ethylene oxide production

A contribution analysis of the GWI of the emerging CO_2R to ethylene oxide pathway is shown in Fig. 2. In the base-case model, the net GWI is estimated as 10.2 t CO_2-eq. per t ethylene oxide when all emissions are allocated to ethylene oxide and the Canadian average electricity grid GHG intensity (0.128 kg CO_2-eq. per kW h) is assumed. Mass and energy-based allocation result in lower GWIs: 4.3 and 3.3 t CO_2-eq. per t ethylene oxide, respectively, as a portion of the emissions are allocated to the co-products.

The ethylene electrosynthesis phase in which CO_2 is converted to ethylene is the main driver of the GWI, accounting for nearly 60% of the carbon footprint. This is consistent with results of other carbon footprint assessments of CO_2R systems. For example, the CO_2 conversion phase has been found to be the main driver of the GWI, accounting for 60% and 85% of the GWI in CO_2 electrolysis to ethylene and ethylene oxide, respectively, due to the large energy demand for electrosynthesis.[15,26] This is particularly the case for gaseous products of CO_2 electrolysis. In comparison, in the case of liquid products of CO_2 electrolysis, the energy demand for product separation is usually higher than the energy demand for CO_2 conversion due to the low concentration of liquid products in the product stream. This leads to the separation phase having a larger influence on the GWI than the CO_2 conversion phase when liquid products are synthesized.[16] The energy demand for ethylene electrosynthesis can be reduced by improving the cell voltage and FE towards ethylene.

The ethylene oxide separation phase contributes 5% of the total GWI. The carbon footprint of this phase is a function of the product selectivity and the energy demand for product separation. Low product selectivity leads to high volumes of undesired byproducts in the product stream, resulting in a high energy demand and associated emissions to separate and purify the desired product.

The carbon capture phase is also a notable contributor to the GWI. This phase depends on the CO_2 SPCE and the GHG intensity of the CO_2 feedstock. At a high CO_2 SPCE such as is

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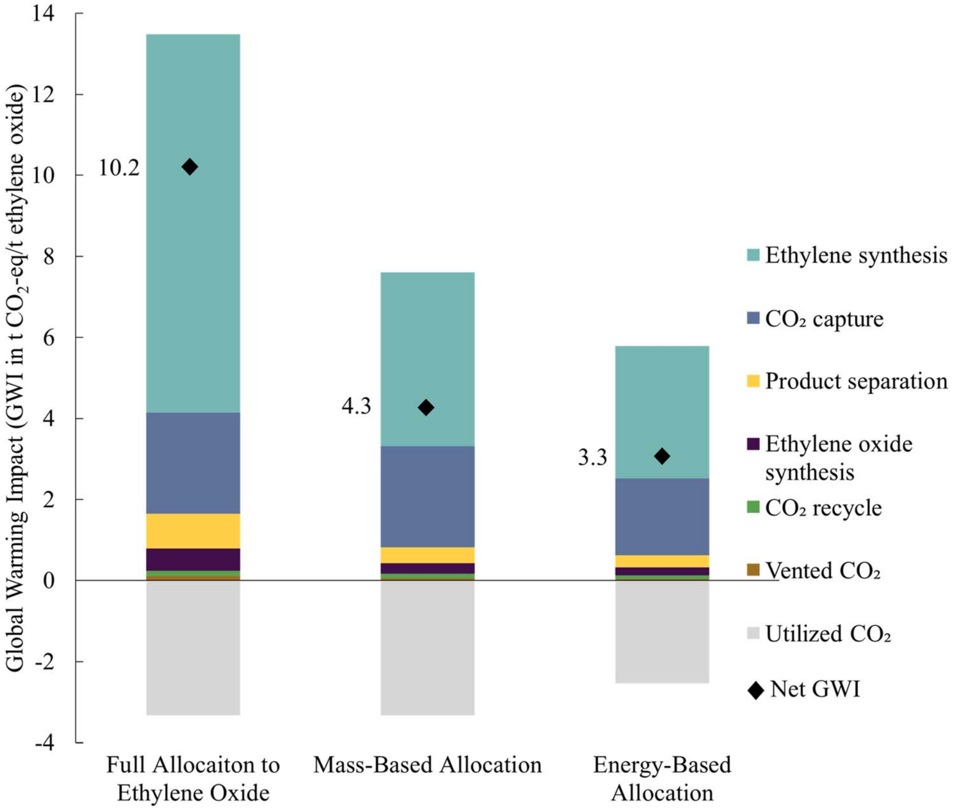

Fig. 2 Base-case global warming impact (GWI) and contribution analysis for ethylene oxide production (cradle-to-gate boundary) through the emerging CO_2R pathway; with full allocation to ethylene oxide and with mass- and energy-based allocation. The diamond represents the net GWI. "Utilized CO_2" represents the credit to the CO_2R to ethylene oxide system for carbon capture. Results assume 2022 Canadian average electricity grid mix GHG intensity of 0.128 kg CO_2-eq. per kW h.[34]

observed in the base-case, the CO_2 feedstock requirement is higher due to less unreacted CO_2 being available for recycle. In such instances, the GHG burden of the capture phase is considerable if the GHG intensity of the CO_2 feedstock is also high. However, in our model, this burden is offset by allocating a credit to the emerging CO_2R to ethylene oxide system for capturing CO_2. In Fig. 2, the "Utilized CO_2" represents the CO_2 emissions that are avoided due to the capture of carbon from the NG combined-cycle power plant for ethylene oxide production.

The mass- and energy-based allocation methods attribute lower emissions to ethylene oxide, leading to 57% and 67% reductions of emissions relative to the base-case, respectively. Note that the base-case model does not include recycle of unreacted ethylene, which would result in a slightly lower total GWI of 9.7 t CO_2-eq. per t ethylene oxide, with 6.7 t CO_2-eq. and 6.0 t CO_2-eq. of this being allocated to ethylene oxide when mass and energy-based allocation, respectively, are applied (see ESI†). We proceed with modeling the sensitivity and scenario analyses using the base-case results with full allocation to ethylene oxide as a reference. Given that the CO_2R pathway is an emerging technology, and the product distribution is dynamic as the process is refined, this approach represents a conservative assessment of the current process performance.

3.2. Sensitivity analysis

The FE of ethylene production has the largest influence on the GWI, followed by the electricity GHG intensity and the energy demand for ethylene electrosynthesis (Fig. 3). The FE of ethylene contributes to the energy demand for ethylene electrosynthesis, the ethylene production rate per unit of CO_2 converted, and the energy demand for subsequent separation of byproducts.

As shown in Fig. 3A, a step increase in the ethylene FE reduces the GWI, whereas an equivalent step decrease in the FE causes a larger increase in the GWI. This is likely attributed to the higher energy demand associated with poor FE (*i.e.*, increased energy consumption during ethylene electrosynthesis and separation of byproducts due to low ethylene production), which outweighs the potential energy savings achievable with higher FE.

The GHG intensity of the electricity and the energy demand for ethylene electrosynthesis are critical for reducing the carbon footprint, while monitoring the ethylene FE is critical to prevent a large increase in the carbon footprint due to losses in FE. The influence of the GHG intensity of the electricity grid on the GWI is explored further in Section 3.3.

The sensitivity analysis on the influence of the CO_2 SPCE shows a non-linear relationship between the CO_2 SPCE and the

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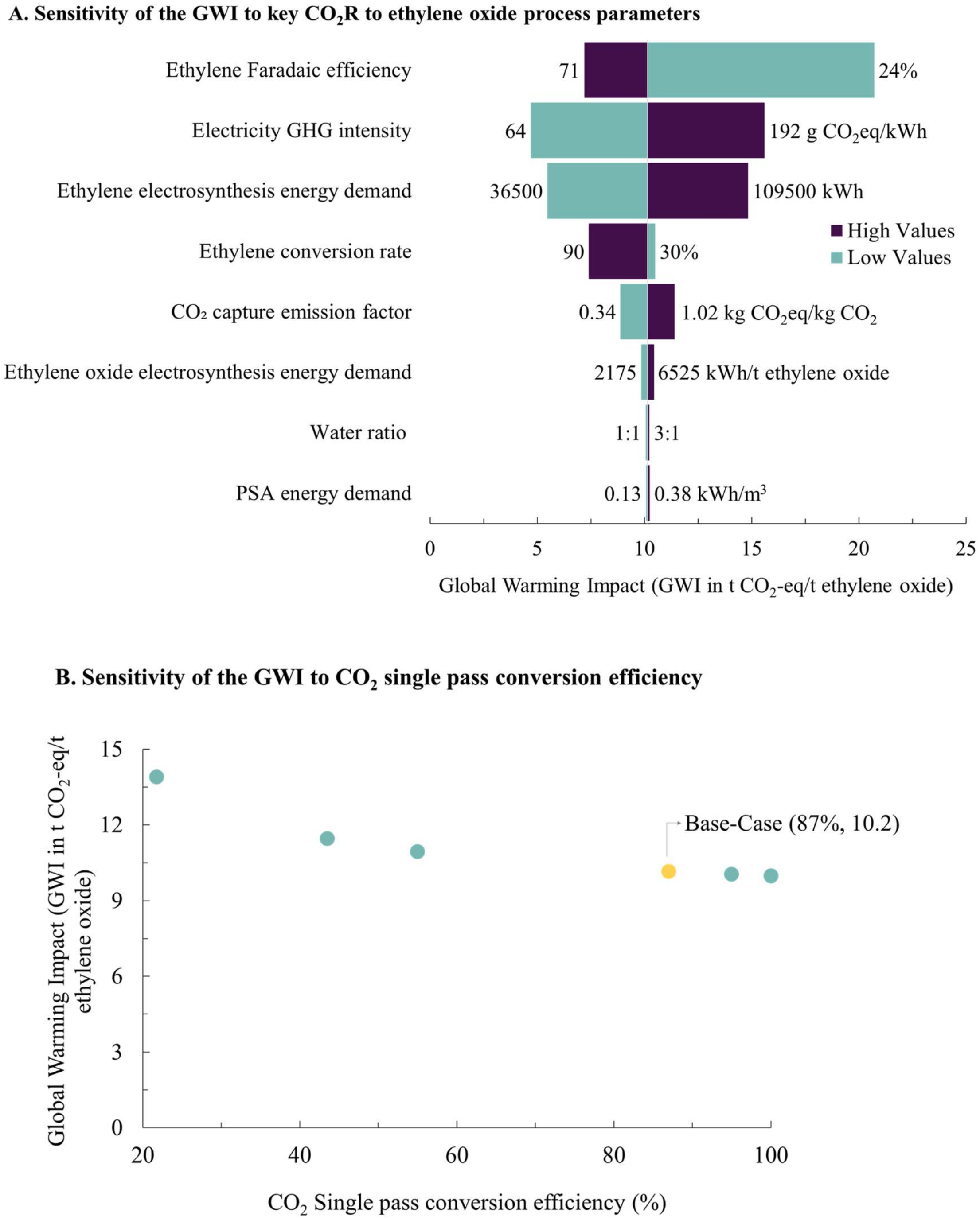

Fig. 3 (A) Tornado plot illustrating sensitivity of the global warming impact (GWI) to key CO_2R to ethylene oxide process parameters. The bars represent the deviation of the GWI from the base-value of 10.2 t CO_2-eq. per t ethylene oxide when each parameter is varied by ± 50%. The base-case value for each parameter is shown in Table 1, while the data labels on Fig. 3 represent the high and low values used for the sensitivity analysis. (B) Global warming impact (GWI in t CO_2-eq. per t ethylene oxide) as a function of the CO_2 single pass conversion efficiency (SPCE). The yellow dot represents the base-case SPCE (87%) and GWI (10.2 t CO_2-eq. per t ethylene oxide).

GWI (Fig. 3B). The GWI plateaus closer to 100% CO_2 SPCE, showing no further notable drop in GWI with improvements in the CO_2 conversion efficiency. Relative to the ethylene FE, electricity GHG intensity and ethylene electrosynthesis energy demand, the CO_2 SPCE has a smaller influence on the GWI. The 79% change in CO_2 SPCE over the range varied decreases the GWI by about 30%, whereas a 50% change in the FE, electricity GHG intensity and the ethylene energy demand change the GWI by more than 60%. Nevertheless, the CO_2 SPCE remains a critical parameter for CO_2R processes as it also impacts process costs (*e.g.*, costs of product separation and CO_2 recycle), although quantifying the process costs is outside the study scope.

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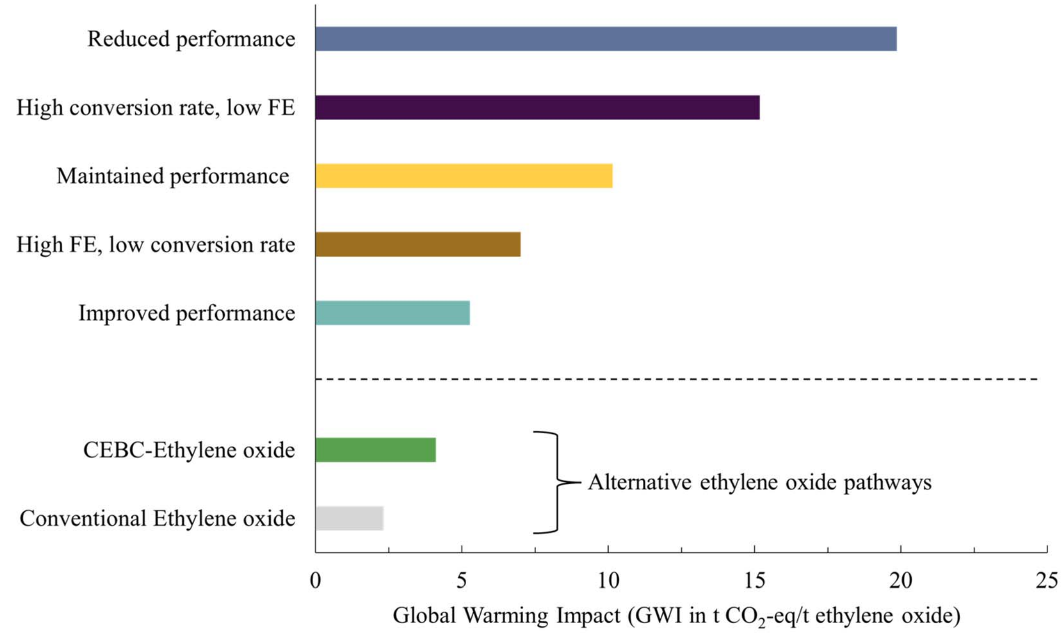
3.3. Scenario analysis and comparison with alternative pathways

Fig. 4 shows the GWI of the performance scenarios for the emerging CO_2R to ethylene oxide process alongside those of the alternative pathways assessed. In the *improved performance* scenario, where the electrolysis parameters are enhanced by 50% compared to lab-scale values, the GWI is reduced by 48% relative to the base-case. However, the GWI in this scenario

A. Scenario analysis at base-case electricity (0.128 kg CO_2-eq/kWh)



B. Scenario analysis at low-carbon electricity (0.025 kg CO_2-eq/kWh)

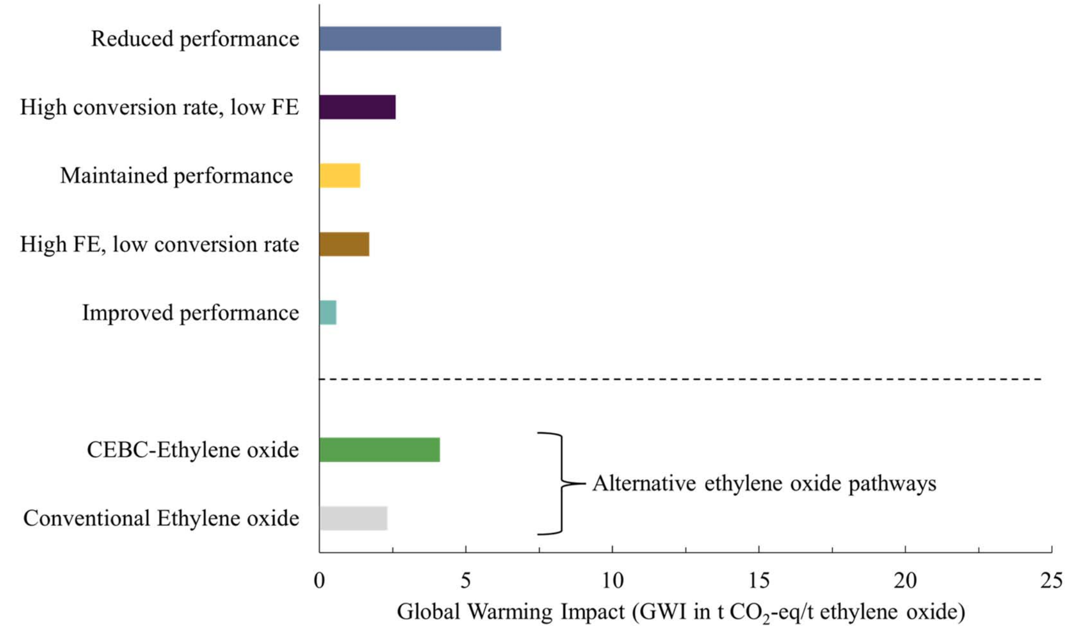


Fig. 4 Scenario analysis showing the global warming impact (GWI) of the scaled up CO_2R to ethylene oxide process at various performance scenarios and the GWI of alternative pathways for ethylene oxide production (A) with the base-case electricity (Canadian 2022 average grid mix, 0.128 kg CO_2-eq. per kW h) and (B) with low-carbon electricity (Ontario grid, 0.025 kg CO_2-eq. per kW h). *Improved performance* represents 50% enhancement in process variables relative to the lab-scale performance, *maintained performance* represents performance identical to the lab-scale performance, *reduced performance* represents 50% loss in performance of relative to the lab-scale. The conventional ethylene oxide pathway is oxidation of fossil-based ethylene in air over a silver catalyst;[6] CEBC-ethylene oxide is the oxidation of fossil-based ethylene using hydrogen peroxide, developed by the Center for Environmentally Beneficial Catalysis at University of Kansas.[27]

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remains higher than that of the conventional and CEBC-ethylene oxide processes when using the base-case electricity GHG intensity of 0.128 kg CO_2-eq. per kW h (Fig. 4A). When powered by a lower-carbon electricity source, such as the Ontario grid (0.025 kg CO_2-eq. per kW h) where the lab-scale technology is being developed, the *improved performance* scenario has a 74% lower GWI than the conventional pathway (Fig. 4B) even with the conservative approach of allocating all emissions to ethylene oxide.

From the scenario analysis, we observe that a change in the FE of ethylene production has more influence on the GWI than the equivalent change in CO_2 SPCE. Interestingly, the *high FE/low conversion rate* scenario performs better than the *maintained performance* scenario, although the CO_2 SPCE is as low as 55% and the FE was only increased to 70%. Other studies have predicted ethylene FE as high as 90%[15] in sensitivity analyses, making the "high FE" in our scenario analysis a conservative estimate. Improvements in the FE toward ethylene beyond 70% will further reduce the process GWI.

As seen in the *high conversion rate/low FE* scenario, even with 95% CO_2 SPCE, the penalty on the GWI due to the low FE (23.5%) is much more significant. This is because although high CO_2 SPCE is generally desirable, if the CO_2 is converted to more undesired products than the intended product (as in the case of a lower FE to ethylene), this leads to a higher energy demand for ethylene electrosynthesis and product separations, as well as a higher CO_2 feed requirement to meet the plant capacity. As this is an electrochemical process, the electrical energy demand is arguably one of the most important parameters.

The GWI of the *maintained performance* scenario becomes lower than that of the conventional pathway when the electricity GHG intensity is lower than 0.04 kg CO_2-eq. per kW h. This demonstrates that if deployed with a lower-carbon electricity grid, even without further improvements to the electrolysis performance, the emerging CO_2R to ethylene oxide technology could prove attractive compared to the conventional fossil-based pathway from a carbon footprint perspective. However, cost considerations will likely dictate the need for further improvements in the process performance.

The analysis of the influence of the background system shows that the GWI of ethylene oxide production is more sensitive to the GHG intensity of the electricity supply than to the CO_2 feedstock source (Fig. S4†). Given the critical nature of the electricity GHG intensity on the GWI of the process, we examine the influence of the electricity grid intensity beyond the initial boundaries of the sensitivity analysis and explore current and future electricity grids, particularly in the top ethylene oxide producing regions (Fig. 5).

While the importance of low-carbon electricity in CO_2R processes is well established, this study advances that understanding by linking grid GHG intensity to potential deployment

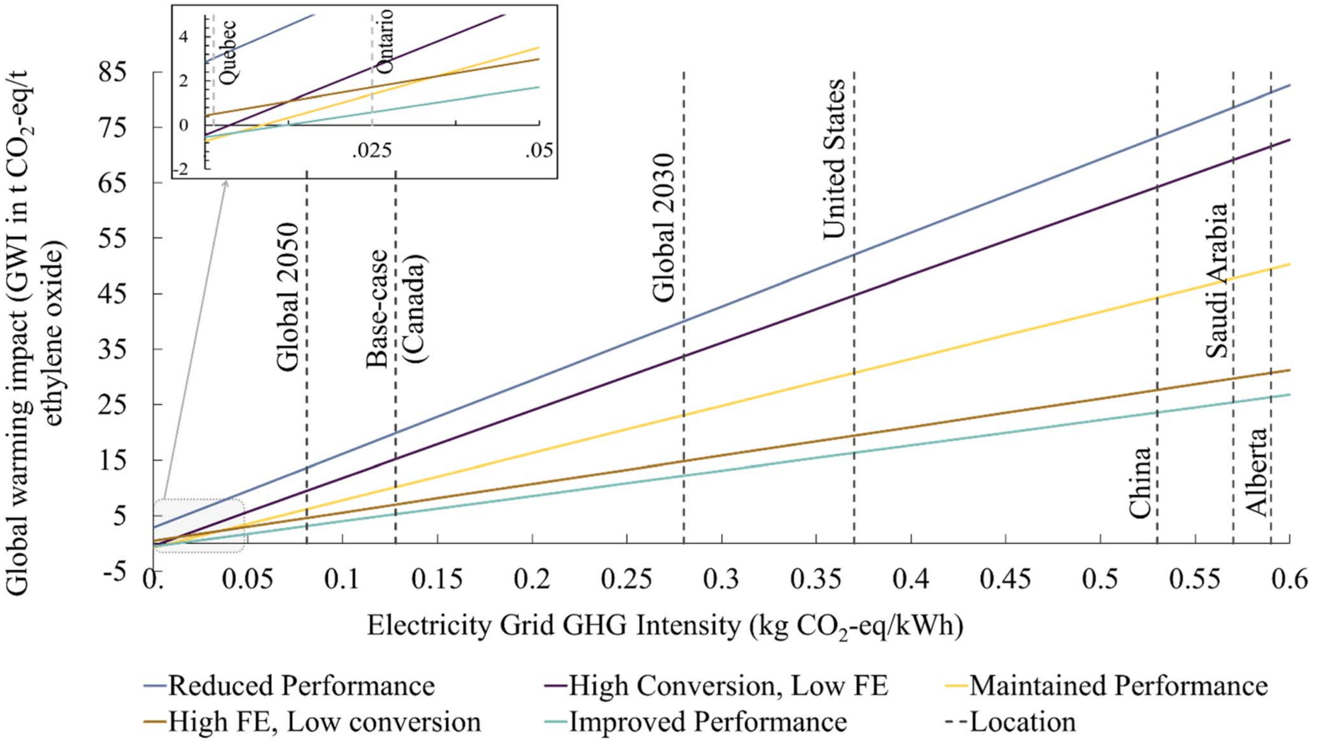

Fig. 5 Global warming impact (GWI) of the emerging CO_2R to ethylene oxide technology at various performance scenarios as a function of the GHG intensity of the electricity supply. The vertical dashed lines represent the average GHG intensity of electricity supply (2022) in the top four ethylene oxide producing countries; (United States, Saudi Arabia, China, and Canada), and the Global 2030 and 2050 target electricity supply GHG intensities.[34] *Improved performance* represents 50% enhancement in process variables relative to the lab-scale performance, *maintained performance* represents performance identical to the lab-scale performance, *reduced performance* represents 50% loss in performance of relative to the lab-scale. The GWI of the conventional ethylene oxide pathway (not shown) is 2.32 tonnes CO_2-eq. per tonne ethylene oxide.

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regions with existing ethylene oxide markets. Our findings highlight that the priority for process improvements should be tailored to the electricity grid GHG intensity. As shown in Fig. 5, because of the impact of electricity grid GHG intensities, the location where the emerging CO_2R to ethylene oxide pathway is deployed informs the process performance conditions necessary to make the technology attractive relative to the conventional pathway from a GWI perspective. For example, if deployed with the average Canadian electricity grid (0.128 kg CO_2-eq. per kW h), performance improvements greater than 50%, as modeled in the *improved performance* scenario, would be required for the technology to have a lower GWI than the conventional pathway. At this grid GHG intensity, improvements in the faradaic efficiency of ethylene should be prioritized over improvements in the CO_2 single pass conversion efficiency. Conversely, the CO_2 single pass conversion efficiency plays a larger role in reducing the GWI if the process is to be deployed in locations with lower GHG intensity electricity grids (below 0.0125 kg CO_2-eq. per kW h).

As the electricity GHG intensity gets closer to zero, for example, in Quebec, the *maintained performance* scenario becomes the most attractive from a carbon footprint perspective. This is because performance improvements in the CO_2 SPCE and ethylene FE become inconsequential to the GWI as the carbon footprint associated with the ethylene electrosynthesis phase (the main driver of the process GWI) approaches zero due to zero-carbon electricity supply for electrolysis. Therefore, if prioritizing a zero-carbon electricity source, at the present stage of development, the emerging CO_2R to ethylene oxide technology can offer GHG savings relative to the conventional pathway, even with a conservative allocation approach.

It is challenging to predict the performance of the technology at a commercial scale and there is a need for further assessment of the interaction of the parameters due to scale up. Nevertheless, the scenario analysis, particularly the *conversion/FE* scenarios, provides insight into the impacts of potential trade-offs that might be observed at scale. Further, given the challenge of predicting the performance of the technology at scale, we quantify thresholds for each critical parameter, within which the CO_2R to ethylene oxide process GWI remains below the GWI of the conventional pathway for a specific electricity grid scenario (Table 4).

The threshold for each parameter is determined independently, assuming the other critical parameters remain at their base-case values. For example, assuming the Ontario grid scenario, and all other parameters remaining at the base-case value, the ethylene FE would need to be at least 35% for the emerging CO_2R process GWI to be below the conventional ethylene oxide emissions of 2.32 t CO_2-eq. per t ethylene oxide. The quantification of these thresholds serves as a guideline for technology developers when targeting large-scale performance of the technology.

3.4. Process safety considerations

Commercial ethylene oxide production presents significant safety challenges, including the risk of runaway reactions and ethylene oxide decomposition due to its reactivity and thermal properties.[49,50] The oxidation of ethylene to ethylene oxide is extremely exothermic, and therefore necessitates stringent process controls to prevent safety incidents. These control measures limit the ethylene conversion to below 10% to avoid overoxidation of the ethylene oxide, costing nearly US$1.2 billion annually due to reactant loss.[51,52] Even with the complex safety control systems for commercial ethylene oxide production, multiple deadly explosions have occurred over the years, exceeding the incident rate of other hydrocarbon oxidation systems by about 80%.[50] Notably, recent incidents in Spain (2020) and the US (2023) underscore the persistent challenges in maintaining safe operations.

Efforts to address the safety concerns in commercial ethylene oxide production have led to technological advancements. For example, microreactors have been proposed for silver-catalyzed ethylene oxidation to ethylene oxide.[53] Microreactors employ more efficient mass and heat transfer conditions to avoid known reactor hotspots and can safely handle gas compositions within the explosive limits.[53] However, despite lab-scale demonstrations of this process, large-scale implementation remains pending, and the environmental impacts of the process are yet to be quantified. The CEBC-ethylene oxide process is another advancement, which conducts ethylene oxide synthesis in the liquid phase, to avoid the flammable mixture of gaseous ethylene oxide/ethylene in air.[27] A comparison of its environmental impacts and costs with those of the conventional process revealed higher CO_2-eq. emissions and process costs, which could impede large scale implementation despite improved safety.

The carbon footprint assessment of the emerging CO_2R to ethylene oxide technology in this study showed that the emerging technology could compete with the conventional pathway when coupled with low carbon electricity input and

Table 4 Quantitative CO_2R process parameter thresholds for the GWI of the emerging CO_2R to ethylene oxide process to be below the GWI of conventional ethylene oxide (2.32 t CO_2-eq. per t ethylene oxide).[39]

Parameter	Base-case values	Ontario grid (0.025 kg CO_2-eq. per kW h)	Quebec grid (1.3×10^{-3} kg CO_2-eq. per kW h)	Zero-carbon grid (0 kg CO_2-eq. per kW h
Ethylene faradaic efficiency	47%	≥ 35%	≥ 8%	≥ 7%
Ethylene electrosynthesis energy demand	73 000 kW h per t ethylene oxide	≤ 109 500 kW h per t ethylene oxide (≤4.75 V)	≤ 2.3×10^6 kW h per t ethylene oxide	Agnostic to energy demand
CO_2 SPCE	87%	≥48%	≥23%	≥22%

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future process improvements. This section discusses the safety aspects of the technology. Historically, explosive incidents in commercial ethylene oxide production have predominantly resulted from runaway reactions. Most plants operate within the explosive limits (2.6 to 100% volume in air[54]) and at high temperatures (between 200 °C to 300 °C) and pressures (10–30 bar).[55,56] These operating conditions are dangerously close to the onset temperature and autoignition temperature (300 °C and 428 °C respectively) of ethylene oxide.[51,57] In contrast, the emerging CO_2R to ethylene oxide pathway operates at ambient temperature and pressure.[8] The CO_2R to ethylene oxide process therefore offers higher ethylene conversion rates (60% *versus* 10% in the commercial pathway) and higher ethylene oxide yields, while operating further from the explosive limits. While the ethylene oxide is produced using 2-chloroethanol as an intermediate, this pathway does not present safety concerns as the intermediate is not released but completely converted to ethylene oxide (see ESI†).

Another area prone to explosions in the commercial ethylene oxide process lies within the distillation columns for separating high purity ethylene oxide. Pure ethylene oxide can decompose explosively and is also highly sensitive to impurities.[54,58] Therefore, it is recommended to use inert gases, such as nitrogen in the separation and storage of ethylene oxide,[50] and to avoid contamination of ethylene oxide in the separations process.

4 Conclusion

Commercial ethylene oxide production emits more than 74 million metric tons of CO_2-eq. annually, constituting approximately 2% of emissions from the chemicals sector. We analyzed ethylene oxide produced *via* an emerging CO_2 electrolysis technology, determining for the first time its carbon footprint and conditions under which the technology could be attractive from a carbon footprint perspective when compared to conventional ethylene oxide production.

We project that the technology could have a lower carbon footprint than the conventional pathway (2.32 tonnes CO_2-eq. per tonne ethylene oxide) when the process is coupled with a very low GHG intensity electricity supply (<0.04 kg CO_2-eq. per kW h), and could completely offset the emissions of the conventional pathway when paired with zero-carbon electricity supply (assuming no embodied emissions). We advance the understanding of how electricity grid GHG intensity influences the process by demonstrating its varying impact on the emerging CO_2R technology across locations and identifying which process parameters should be prioritized under different grid conditions. Further, we provide quantitative guidance on key parameters for process advancements to reduce emissions. While directly relevant to the ethylene oxide system, this approach can be broadly applied to other CO_2R processes, making the work of significance to technology developers in the field of CO_2 electrolysis.

This study demonstrates the potential of CO_2 electrolysis as a pathway for ethylene oxide production to offer GHG savings relative to conventional fossil-based production, advancing efforts to decarbonize the chemicals sector. The findings of this study are beneficial to CO_2 electrolysis technology developers broadly, and can inform policy decisions, guiding governmental bodies and other stakeholders in setting climate targets and net-zero goals. Further, in the context of ethylene oxide production, our examination of the safety considerations highlights the potential of CO_2 reduction technology to achieve higher yields with safer operating conditions than the current commercial pathway.

4.1. Limitations and future directions

This study assesses a specific process for ethylene oxide production *via* CO_2 electrochemical reduction, despite the existence of other configurations. To improve this specific pathway, future research could explore a three-step electrosynthesis approach that involves first reducing CO_2 to carbon monoxide (CO). This approach has successfully improved CO_2 SPCE and FE when producing other C_1 and C_2 products.[15,45,59] However, the energy demand of synthesizing CO and the infrastructure implications of a 3-step electrolysis process must be carefully considered.

The study relies on limited lab-scale data, with assumptions to fill in data gaps, leading to uncertainties, particularly regarding the performance of the technology at commercial scale and the comparisons with alternative ethylene oxide production pathways. Other limitations include the exclusion of the liquid byproducts generated in the CO_2 electrolysis process due to their minimal quantities. However, the accumulation of liquid products in the electrolyte over time could inhibit the electrolyzer performance. Therefore, we recommend that the electrolyte is replaced after a threshold of 10% v/v accumulation of product is reached.[15] Embodied emissions in the electrolyzer, catalyst and other equipment are also excluded due to data limitations, though prior studies suggest these emissions are small compared to operational emissions,[15] particularly with electrode lifetimes over 210 hours.[44,60]

Data availability

The data supporting this article have been included as part of the ESI.†

Author contributions

Conceptualization: A. B., H. L. M., B. A. S.; data curation: J. E. H., A. B.; formal analysis: A. B., H. L. M., B. A. S.; funding acquisition: H. L. M.; investigation: A. B.; methodology: A. B., H. L. M., B. A. S.; resources: H. L. M., B. A. S., E. H. S.; software: A. B.; supervision: H. L. M., B. A. S., E. H. S.; validation: all authors; visualization: A. B.; writing – original draft: A B.; writing – review & editing – all authors.

Conflicts of interest

There are no conflicts of interest to declare.

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