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Exploring circularity in sorption-enhanced methanol synthesis: a comparative life cycle assessment†

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The transition to renewable energy is driving sustainable chemical production methods, with power-to-X (P2X) technologies offering promising solutions. This study presents a comparative life cycle assessment (LCA) of adiabatic and isothermal sorption-enhanced methanol synthesis (SEMS) processes, across eight scenarios, varying in reactor configurations, electrolysis power sources, and methanol synthesis electricity use. Seven impact categories are evaluated, with results showing that all SEMS scenarios achieve significantly lower global warming potential (GWP) than the reference case of methanol production *via* steam reforming of natural gas. Adiabatic SEMS scenarios range from 71.7 to 519.7 kg CO_2 eq. per t MeOH, while isothermal SEMS scenarios range from 72 to 529 kg CO_2 eq. per t MeOH, significantly outperforming the conventional methanol production process (980 kg CO_2 eq. per t MeOH). These results indicate that SEMS-based methanol achieves 71.6–96.1% GHG savings, meeting the Renewable Energy Directive (RED II) 70% threshold for sustainability. The impacts of both corresponding adiabatic and isothermal SEMS scenarios are similar, with only slight variations. Furthermore, 61% of the impact categories analyzed across all SEMS scenarios exhibit lower impacts than the reference case. Uncertainty and sensitivity analyses revealed that electricity demand associated with water electrolysis was the dominant factor affecting system-level environmental performance of the SEM process. These findings highlight SEMS, when powered by renewable energy, as one possible solution within the P2X framework for sustainable methanol production. This study also emphasizes the critical role of integrating renewable energy into chemical processes to support industrial decarbonization and to accelerate the energy transition.

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1. Introduction

Transitioning to sustainable energy is critical to address climate change and to meet increasing global energy demands. The development of technologies that can efficiently convert electricity from renewable energy sources (RES) into clean fuels is at the core of meeting this challenge. Finland is committed to become carbon neutral by 2035, with a plan to phase out coal by 2029 and to achieve over 51% energy consumption from renewables by 2030.[1,2] These efforts include expanding wind and solar capacities and integrating them with other hybrid systems such as bioenergy and battery energy storage systems.[3]

Globally, the transport sector is the second-largest contributor to GHG emissions. In 2022, it accounted for 20.7% of global CO_2 emissions, marking a 3.23% increase year-on-year.[4] This highlights the significant need for emissions reduction in this sector. Considering this, it is important to prioritize the adoption of alternative fuels and sustainable transportation solutions to address the challenge effectively.

There is increasing interest in exploring alternative pathways for methanol production, specifically through CO_2 hydrogenation.[5] Methanol is a promising candidate for a clean fuel.[6–8] It is also a key chemical feedstock used in various industrial processes, including the production of formaldehyde, dimethyl ether, and olefins.[9–12] Methanol is also among the alternative transportation fuels due to its lower carbon footprint than crude-oil-based fuels on an equivalent energy basis.[13,14]

Previous studies on green methanol production through CO_2 hydrogenation have explored its environmental impacts, emphasizing key factors like emissions reduction and low-carbon hydrogen sources. For instance, Adnan and Kibria[15] have compared the environmental performance of one-step, two-step and three-step methanol synthesis processes. They found that an electricity emission factor below 130 g CO_2 per kWh is required for P2X green methanol production to be more

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climate-friendly compared to conventional steam reforming of natural gas. Similarly, Fernández-González *et al.*[16] found that electricity carbon intensity should be in the range of 100–140 kg CO_2 per MWh to make CCU process feasible. However, these emissions from ref. 15 and 16 are significantly higher than the Finnish grid mix, which has an average emission factor of 39.5 kg CO_2 per MWh,[17] thus, emphasizing the potential for sustainable methanol production in Finland. Based on results from ref. 16, an integrated LCA has shown that CO_2-based methanol has a lower carbon footprint of −1.272 kg CO_2 eq. per kg MeOH compared to 0.584 kg CO_2 eq. per kg MeOH from steam reforming of natural gas process. However, this is possible only when using hydrogen with low or zero carbon emissions. Similarly, CO_2 emissions from CO_2-based methanol synthesis were found to be 50% lower than those from conventional processes based on natural gas steam reforming. It was also indicated that hydrogen production through water electrolysis (WE) is the main contributor in most impact categories.[18] Rigamonti and Brivio[19] made an LCA study on the utilization of steel mill process gases to produce both methanol and electricity. The study found that for most impact categories, the positive effects of using avoided products that include average European electricity mix and producing methanol from natural gas were greater than the negative impacts caused by the process itself. They highlight the need for conducting LCA studies from the beginning of a new technology development to ensure sustainability.[19]

Another primary challenge in methanol synthesis *via* CO_2 hydrogenation is achieving a high per pass conversion. This is hindered by unfavorable equilibrium and the accumulation of water as a by-product. In conventional processes, low per-pass conversion necessitates the recirculation of unreacted gases to improve overall yield. A promising approach to address these challenges is the implementation of sorption-enhanced methanol synthesis (SEMS), which facilitates higher conversion by *in situ* water removal. This enables the design of compact, low-cost equipment with integrated high-efficiency heat utilization. In a previous article by the co-authors, a detailed analysis of a novel SEMS process, comparing two different configurations: isothermal and adiabatic was presented.[20] It was found that the adiabatic configuration is more competitive in terms of overall methanol production cost, comparable to the conventional CO_2 hydrogenation process. However, the environmental performance of the SEMS process was not assessed. Therefore, there is a need to conduct a thorough assessment of the environmental performance of the SEMS process. This analysis is essential to understand its sustainability and potential environmental impacts, providing valuable insights for its further implementation. To the best of our knowledge, this study is among the first to conduct a comparative LCA of the SEMS process, evaluating adiabatic and isothermal reactor configurations. Furthermore, to enhance the robustness of the environmental assessment, both sensitivity and uncertainty analyses were conducted. While sensitivity and uncertainty analyses have been applied in some LCA of P2X technologies,[15,18,21] their quantitative application in SEMS and circular economy contexts remain relatively underexplored. Hence, this approach contributes to filling this gap by providing a comprehensive understanding of the uncertainty and sensitivity of the system's performance.

This study aims to provide insights by conducting a comparative LCA of two SEMS reactor types (adiabatic and isothermal) with reference to Finland and a conventional methanol production process based on steam methane reforming. Each production pathway includes four scenarios, where water electrolysis and electricity for SEMS is assumed to be supplied with hydropower or wind energy. The emissions related to the manufacturing phase of the renewable energy are taken into consideration, as they are typically dominant in renewable energy production. It seeks to deepen the understanding of environmental impacts of SEMS process by evaluating the relative environmental performance of these systems. Additionally, the integration of the SEMS process within a circular economy framework by utilizing both waste CO_2 and waste heat to meet the heat demand within the systems adds a new perspective on how these technologies can reduce waste and close material loops.

2. Methods

2.1. Process description

The overall process block diagram of the sorption-enhanced methanol synthesis is presented in Fig. 1. For the LCA study, the process has been simplified to focus on the most critical parts of the process. A detailed process description can be found in the previous work.[20] A brief description of these processes is provided herein.

The two processes under consideration are identical, differing only in the reactor design (adiabatic and isothermal routes). The process involves the utilization of CO_2 and H_2 as primary inputs. The H_2 is assumed to be produced *via* water electrolysis powered by renewable energy sources. The CO_2 utilized as a feedstock in this system was derived from biogas plant. This aligns with national policies supporting biogas as a renewable energy source to meet climate goals in Finland.[1,22]

The process under the present study consists of 4 key steps, which include reaction, regeneration, separation and waste heat boiler.

(1) Reaction: heated and compressed CO_2 and H_2 are reacting in the presence of a Cu/ZnO catalyst, converting them into methanol and water. Zeolite adsorbs water, which limits one-pass conversion efficiency to methanol. The outlet gas comprises a mixture of CO_2, H_2, CO, and H_2O.

(2) Regeneration: CO_2 is fed into the reactor to remove adsorbed water from zeolite. The outlet gases include mainly CO_2, H_2, CO, and H_2O.

(3) Separation: the outlet gases from the reactor are directed to a condenser to remove water from the stream followed by flash separation units to separate methanol from non-condensing species. 90% of both CO_2 and H_2 are recycled back to the reaction stage to enhance efficiency, while the remaining 10% is purged to gas–gas separation and waste heat boiler.

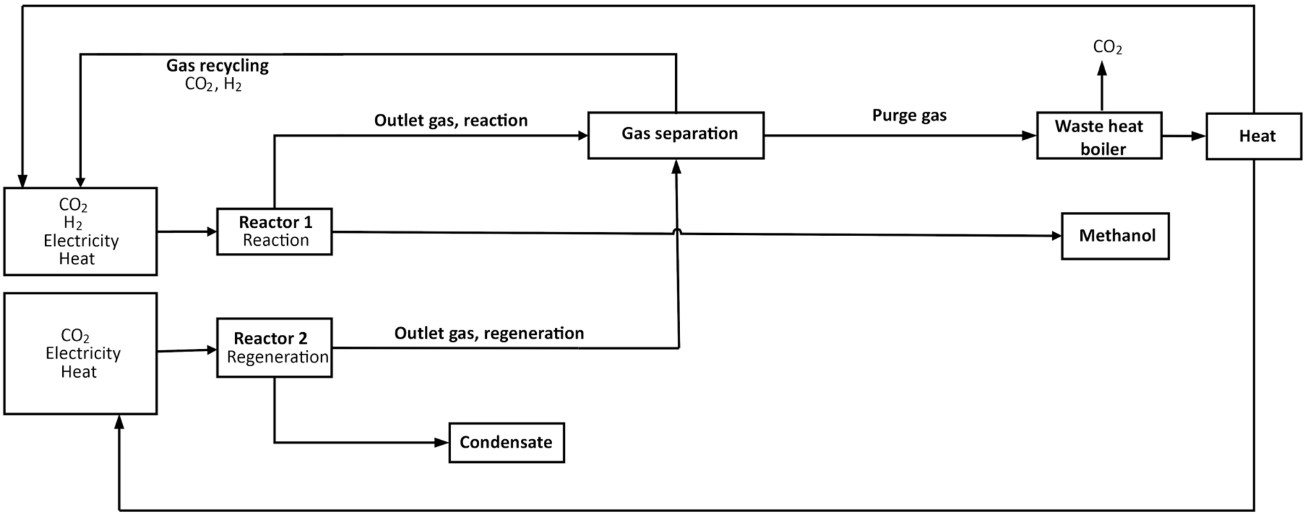

Fig. 1 Block diagram of the sorption-enhanced methanol synthesis process.

(4) Waste heat boiler: the purge gases are combusted with air in a waste heat boiler to produce steam. It has a combustion efficiency of 85%. The CO_2 emissions from combustion are considered as net emissions, reflecting reductions achieved through the combustion process. The produced steam is recycled within the system to meet the thermal energy requirements for both reaction and regeneration processes, forming a closed-loop system that improves overall process efficiency.

2.2. LCA modelling

Life cycle assessment (LCA) is applied to compare the environmental performance of adiabatic and isothermal routes with the conventional process. LCA allows for the evaluation of environmental impacts and identification of key environmental hotspots associated with products or services.

An attributional LCA was conducted following the International Standard Organization (ISO) 14040/44 guidelines.[23,24] The functional unit (FU) as a benchmark for this study is 1 t of methanol. This FU is consistent with the methodology commonly applied in similar studies.[15,18,25–27] Since sorption-enhanced methanol synthesis is at an early stage, a cradle-to-gate system boundary is considered, which include feedstock production, methanol synthesis, and emissions associated with upstream processes. This means that the analysis extends to the stage where the primary product is delivered at the factory gate. As a result, further processing, use and disposal are excluded in either system. While the CO_2 source, its capture, and hydrogen production are outside the core system boundary, their environmental impacts are accounted for to ensure a comprehensive assessment of the SEMS process. The LCI data for CO_2 capture were adapted from Eggemann *et al.*,[28] who applied pressure swing adsorption (PSA) to separate CO_2 from biogas produced *via* anaerobic digestion of manure and straw. The dataset includes electricity consumption for both desulfurization and CO_2 capture. The hydrogen production and CO_2 capture processes are shown outside the system boundary in Fig. 2 to clearly indicate their external role and ensure that their contributions to the environmental performance of the systems are accounted for comprehensively.

The life cycle inventory data for the foreground systems was derived from a previous study conducted by the co-authors of the present work where dynamic reaction cycle modelling (MATLAB R2021a) was combined with a steady-state modelling (Aspen Plus v11) of the overall process.[20] The mole fractions of gases exiting the reaction and regeneration processes, were used to calculate the mass flow rates of each component. This calculation was based on the target methanol output specified in the previous study, 1366 kg h^{-1} for adiabatic system and 2070 kg h^{-1} for isothermal system.[20] The calculations were then applied in the present work, with modification made to the original process. Since that study did not model CO_2 capture and hydrogen production, these processes are incorporated using secondary data sources.[28] In this study, generic data from LCI Ecoinvent database version 3.10 (v3.10)[29] were utilized as a background LCI database. The Ecoinvent database provides comprehensive datasets on the environmental impacts associated with various industrial processes, materials, and energy systems. It is widely used in LCA to model and quantify environmental impacts, providing a robust and reliable foundation for LCA analysis. The various scenarios were modeled and analyzed using a commercial LCA software SimaPro 9.6.0.1. SimaPro is designed to assess and model the environmental impacts of different processes, products, and systems, in line with ISO 14040/44 standards.[23,24] The LCI data for Finland were primarily obtained from the Ecoinvent database. In case where data specific to Finland were unavailable, European or global datasets were used to maintain consistency and ensure data completeness. The differences in production technologies between Finland and other countries for instance, Germany may contribute to uncertainties due to production technologies, resource availability, and regulatory framework.

Processes and materials specific to the systems under study, such as green methanol synthesis, regeneration, and waste heat

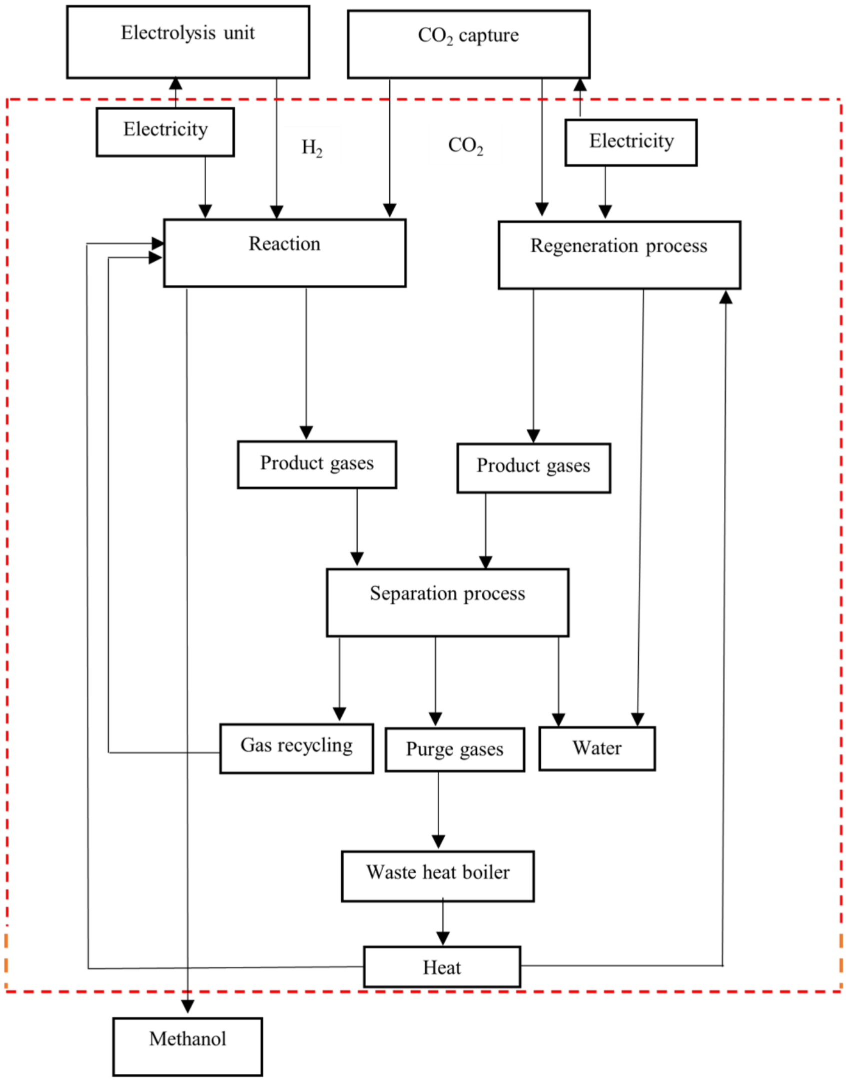

Fig. 2 A simplified LCA system boundary for SEMS process.

boiler combustion were not available in Ecoinvent database. Therefore, custom-made processes and flows were created within SimaPro to represent the missing data elements. For each unavailable element, relevant data was sourced from literature, experimental data or industrial reports. These custom-made elements were integrated into the broader system to ensure the completeness and accuracy of the LCA. Each new process or flow was documented with its corresponding data sources and assumptions to maintain transparency and reproductivity. This approach ensured that the unique aspects of the systems under study were appropriately captured, even when pre-existing datasets were insufficient. The LCI for adiabatic SEMS and isothermal SEMS pathways are presented in Tables 1 and 2, respectively.

In this study, the SEMS process produces methanol on a small-scale in a decentralized plant, which is located close to hydrogen and CO_2 sources. This setup reduces the need for transportation costs and associated emissions. To simplify the comparative assessment, only material and energy consumption during the operational phase are considered across all scenarios. The infrastructure required for the adiabatic and isothermal SEMS is assumed to have a negligible contribution to the overall environmental impact, as both reactor systems are designed to deliver equivalent functionality. Therefore, infrastructure-related impacts are excluded from the system boundaries of this study. In general, a key challenge in accurately assessing the environmental performance of products incorporating recycled materials is the potential for double-counting material flows. To address this challenge, we adopted a net material flow approach to account for the environmental impacts of both virgin and recycled materials.

In this study, the ReCiPe 2016 Midpoint level methodology (hierarchical perspective)[30] was used to evaluate the environmental impacts of the processes. The midpoint level focuses on quantifying specific environmental impact categories, such as global warming potential, eutrophication, and fossil resource scarcity, without aggregating them into broader damage categories. The method aligns with commonly accepted policy

Table 1 Life cycle inventory of inputs and outputs of adiabatic SEMS

	Reaction	Regeneration	Waste heat boiler
Input			
CO_2 (kg h^{-1})	2541.67	822.52	6819.04
H_2 (kg h^{-1})	354.17	0.27	44.61
CO (kg h^{-1})	—	0.50	702.70
H_2O (kg h^{-1})	—	—	53.51
Electricity (kWh h^{-1})	52	1440	—
Output			
MeOH (kg h^{-1})	1366	—	—
Regeneration outlet gas (kg h^{-1})	—	874.68	—
Heat (MJ h^{-1})	—	—	14 077.98
Emissions			
CO_2 (kg h^{-1})	—	—	1341.08
H_2O (kg h^{-1})	—	—	481.08

principles, emphasizing temporal aspects and global-scale impact mechanisms.[30] Relevant environmental impact categories were further analyzed based on the recommendations of LCA in the European context.[31] In total, seven (7) impact categories were selected because they are primarily required by the environmental authorities in Finland for assessing the environmental impacts of new plants.[32] The results were then compared with the conventional methanol production process *via* steam reforming of natural gas. The reference conventional process based on steam reforming of natural gas was taken from Ecoinvent v3.10.[29]

2.3. Scenario study

In this study, eight scenarios for methanol production by adiabatic and isothermal SEMS were evaluated. Each production pathway includes four scenarios, where hydrogen is assumed to be supplied exclusively by either hydropower or wind energy. This scenario-based approach is adopted given that hydrogen production is energy-intensive, and its environmental footprint depends on the energy source. We compare methanol synthesis using these renewable electricity sources to assess their impacts on sustainability. The scenarios are summarized in Table 3, which outlines the combinations of electricity use for hydrogen production, and the in SEMS processes. This approach allows for a thorough assessment of the environmental impacts.

2.4. Uncertainty analysis

Uncertainty is inherent in every stage of LCA, influencing the accuracy and reliability of results. It can stem from factors such

Table 3 Scenarios of the analyzed SEMS

Scenario	Reactor type	WE electricity source	Electricity for SEMS
A1	Adiabatic	Hydro	Hydro
A2	Adiabatic	Hydro	Wind
A3	Adiabatic	Wind	Hydro
A4	Adiabatic	Wind	Wind
B1	Isothermal	Hydro	Hydro
B2	Isothermal	Hydro	Wind
B3	Isothermal	Wind	Hydro
B4	Isothermal	Wind	Wind

Table 2 Life cycle inventory of inputs and outputs of isothermal SEMS

	Reaction	Regeneration	Waste heat boiler
Input			
CO_2 (kg h^{-1})	3851.21	1240.6	6819.04
H_2 (kg h^{-1})	536.65	42.03	44.61
CO (kg h^{-1})	—	49.41	702.70
H_2O (kg h^{-1})	—	169.32	53.51
Electricity (kWh h^{-1})	92	2184	—
Output			
MeOH (kg h^{-1})	2070	—	—
Regeneration outlet gas (kg h^{-1})	—	29 061.04	—
Heat (MJ h^{-1})	—	—	29 061.04
Emissions			
CO_2 (kg h^{-1})	—	—	1376.79
H_2O (kg h^{-1})	—	—	1521.83

as input data, methodological choices, modeling approaches, and scenario assumptions. While uncertainties do not invalidate an LCA, however, acknowledging and communicating these uncertainties is essential for contextualizing LCA outcomes. This study accounts for potential uncertainties arising from data variability and assumptions by conducting an uncertainty analysis using Monte Carlo simulation. The simulation, performed with 1000 iterations at a 95% confidence level, evaluates the combined effects of input data variability and parameter uncertainty. Monte Carlo analysis was carried out using SimaPro v.9.6.0.1 to assess uncertainties across all eight SEMS scenarios.

2.5. Sensitivity analysis

Besides uncertainty analysis, a sensitivity analysis was conducted to examine how these assumptions and choices affect the model outcomes and to evaluate the robustness of the results. In this study, perturbation analysis was applied to determine how variations in individual parameters influence the overall results. Perturbation analysis identifies the most influential parameter by calculating the sensitivity ratio (SR) according to eqn (1):[33]

$$SR = \frac{\frac{\Delta \text{result}}{\text{initial result}}}{\frac{\Delta \text{parameter}}{\text{initial parameter}}} \quad (1)$$

Parameters with SR values above 0.8 significantly influence LCA outcomes, with values greater than 1.0 indicating critical importance and values below 0.2 suggesting minimal impact.[34]

3. Results

The results of the Life Cycle Impact Assessment (LCIA) for all eight scenarios and reference case are presented in Table 4. Detailed contributions of each unit process to the impact categories can be found in Fig. SI–SVII of the ESI.† As shown in Table 4, all SEMS system scenarios exhibit significantly lower in most impact categories compared to reference case, primarily due to heat integration and avoided emissions. These factors effectively offset overall environmental burdens.

In this study, the global warming potential (GWP) of SEMS is 46–93% lower than the reference case. For ozone formation, human health (OFH) impact category, five scenarios were lower than the reference case, while scenarios A4, B3 and B4 were higher by 23.4%, 6.2% and 29% respectively. With regard to freshwater eutrophication (FE), only A1 and B1 show lower FE impacts compared to the reference process. In marine eutrophication (ME) impact category, six of the SEMS scenarios exhibited lower impacts than the reference case. However, B4 shows a 3% increase compared to the reference case, while A4 remains nearly unchanged, with only 0.4% reduction. In contrast, A3 and A4 were lower than reference case at 22.1% and 16.2% respectively. Similar to FE, only two scenarios show lower land use (LU) than the reference case, with A1 at 48.8% and B1 at 39.7%. Notably, fossil resource scarcity (FRS) is significantly reduced in all scenarios (85.7–98.5%). Furthermore, in terms of water consumption (WC), scenarios A1–A2 and B1–B2 exhibit reductions of 34.7–75.3% and 34.5–72.6%, respectively, compared to the reference case.

In LCA studies, a hotspot refers to a specific life cycle stage with significant environmental impact. Identifying hotspots within emerging technologies allows practitioners and decision-makers to better prioritize optimization efforts. Among all eight SEMS scenarios in this study, electricity for hydrogen production by water electrolysis is the primary hotspot due to its high energy demand. However, the carbon intensity of electricity consumption are lowered because the electricity used for hydrogen production is sourced from renewable energy. Additionally, the closed-loop nature of the systems helps further to reduce impacts by preventing emissions from being released into the atmosphere. Efficient heat generation and utilization within the systems contribute to overall impact reduction by minimizing waste heat and enhancing energy efficiency. Contributions analysis of all assessed processes across all impact categories in the LCA is available in Fig. SI–SVII and Table SI in ESI.†

3.1. Environmental impact analysis

Regarding process contribution, methanol synthesis is the major contributor across all eight scenarios and seven impact categories analyzed. This is primarily due to its higher net

Table 4 Comparisons of the 7 impact categories analyzed with reference case, per t of MeOH

Scenario	GWP (kg CO_2 eq.)	OFH (kg NO_x eq.)	FE (kg P eq.)	ME (kg N eq.)	LU (m^2a crop eq.)	FRS (kg oil eq.)	WC (m^3)
A1	70.68	0.25	0.020	0.002	5.03	13.88	0.71
A2	174.47	0.57	0.136	0.009	11.30	39.33	1.87
A3	415.91	1.34	0.405	0.026	25.88	98.54	4.59
A4	519.71	1.67	0.520	0.033	32.14	124.00	5.75
B1	74.12	0.26	0.021	0.002	5.93	14.56	0.79
B2	171.49	0.57	0.129	0.009	11.81	38.42	1.88
B3	446.08	1.44	0.435	0.028	28.39	105.78	4.96
B4	543.45	1.75	0.544	0.034	34.27	129.66	6.06
Ref.[a]	980.00	1.35	0.071	0.033	9.83	907.98	2.87

[a] Reference case from Ecoinvent v3.10 [29] and references therein.

material input compared to the other subprocesses. The electrolytic hydrogen used in the methanol synthesis unit contributes to the overall material and energy requirements, thereby influencing the environmental impacts in each scenario. As a result, material inputs become the dominant factor influencing environmental impacts, making methanol synthesis the major contributor in the net values approach. However, the process contributions vary across scenarios.

Global warming potential (GWP) measures the climate impact of greenhouse gas emissions, expressed in kilograms of carbon dioxide equivalent (kg CO_2 eq.). Among the scenarios, A1 has the lowest GWP impact at 70.7 kg CO_2 eq. per t MeOH, followed by B1 at 72 kg CO_2 eq. per t MeOH, while B4 shows the highest impact, with 529.24 kg CO_2 eq. per t MeOH produced. Methanol synthesis contributes most significantly to scenario A1 accounting for 85.8% of the overall impacts in the scenario. Its contribution is 82.1% in B4 scenario (see Fig. 3(a)). The waste heat boiler contribution across all scenarios range from 2.24 kg CO_2 per t MeOH in B1 to 24 kg CO_2 eq. per t MeOH in A4. Its GWP impact is modest, mainly due to heat integration in the process. Water electrolysis is the primary contributor to the GWP in methanol synthesis. Its share of GWP is about 98% for A3 and B3, 89% for A1, A4, B1, and B4 and 54% for A2 and B2. The other significant source of GWP is CO_2 feed. Water electrolysis and CO_2 feed together account for nearly 100% of the GWP in methanol synthesis.

Ozone is formed in the stratosphere when UV radiation split oxygen molecules, creating ozone that protects against UV radiation. In the troposphere, ozone is secondary pollutant formed by reaction between nitrogen oxides (NO_x) and volatile organic compounds (VOCs) under sunlight. In this impact category, the lowest impacts are observed in A1 at 0.25 kg NO_x eq. per t MeOH, and the highest impact is B4 at 1.75 kg NO_x eq. per t MeOH. This is mainly due to emissions from turbine manufacturing, particularly NO_x and VOCs from steel production, which contribute more to OFH impacts. With regard to impact distribution, methanol synthesis was the main contributor in all the scenarios (60–97%). The contributions of regeneration across the scenarios varies from 8.7% to 30.7% (Fig. 3(b)).

In the FE impact category (Fig. 3(c)), phosphate is a key contributor because it originates from phosphorus-containing emissions, such as those from wastewater discharge, agricultural runoff, or industrial processes. This process can lead to algal blooms, oxygen depletion, and significant harm to aquatic ecosystems. FE impacts are expressed in phosphorus equivalents (kg P eq.), quantifying the potential for nutrient loading. In this study, scenarios A1 and B1 exhibit lower impacts of FE at 0.02 and 0.021 kg P eq. per t MeOH respectively relative to reference case. Environmental savings from avoided CO_2 emissions and heat integration reduce total impacts in the remaining six scenarios combining hydropower and wind electricity, as well as in those that use wind electricity alone. However, these impacts remain higher than the reference case, primarily due to phosphorus runoff from material production and construction phases of wind turbines.

In the marine eutrophication category (Fig. 3(d)), nitrogen compounds, mainly nitrates and ammonia, are the primary contributors as they promote excessive algal growth. These emissions come from agricultural runoff, wastewater, and

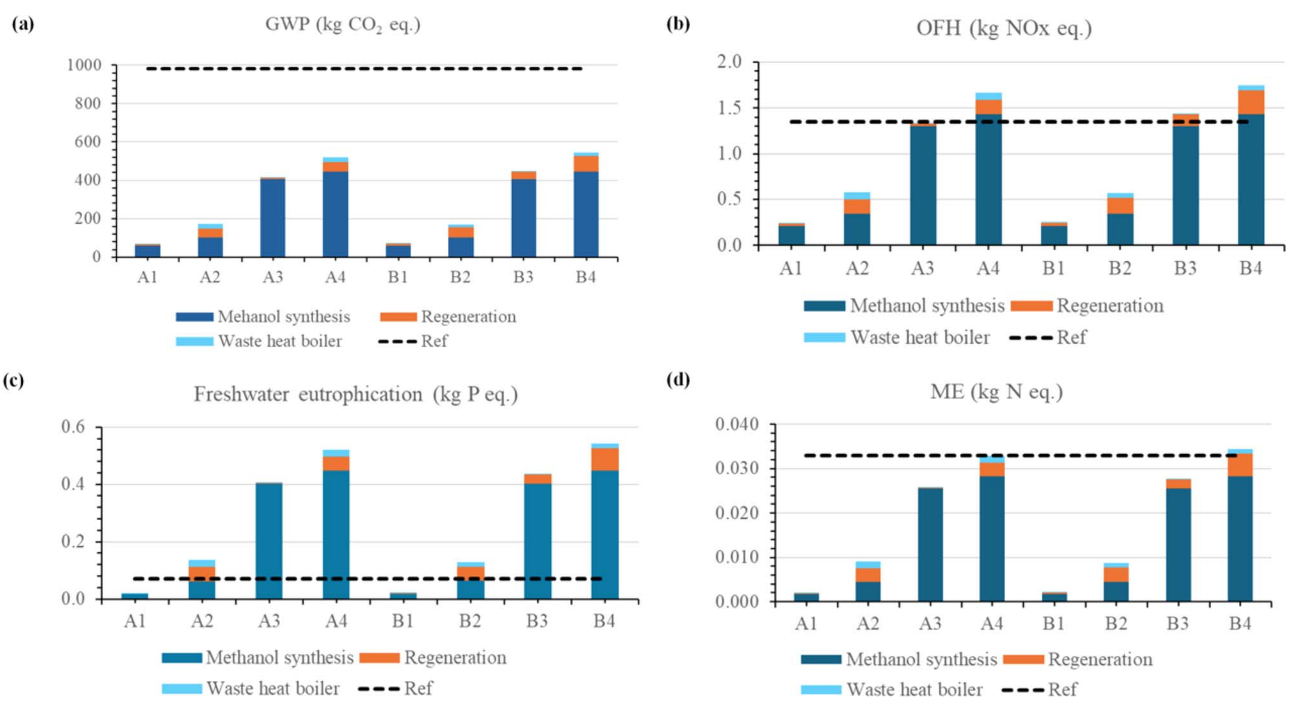

Fig. 3 Process contributions analysis of life cycle stages to the impacts in adiabatic (A1–A4) and isothermal (B1–B4) SEMS scenarios. (a) Global warming potential, GWP, (b) ozone formation, human health, OFH, (c) freshwater eutrophication, FE, (d) marine eutrophication, ME.

atmospheric deposition, leading to oxygen depletion in marine ecosystems. Similar to FE impact category, methanol synthesis unit is the major contributor in all the eight scenarios ranging from 49% to 98.8%. However, the highest impacts were observed in B4 at 0.034 kg N eq. per t MeOH, then followed by A4 at 0.033 kg N eq. per t MeOH. This is mainly due to emissions associated with material and infrastructure requirements, which lead to increased nitrogen runoff during turbine production and construction.

Land use measures the extent of land occupation and transformation required for a process, often expressed in square meters per year (m^2a crop eq.). It reflects the environmental impact on ecosystems, biodiversity, and natural landscapes due to industrial infrastructure and energy production. In this study, the lowest LU impact belongs to A1 at 5 m^2a crop eq. per t MeOH followed by B1 at 5.9 m^2a crop eq. per t MeOH, which are lower than reference case. In this impact category, methanol synthesis is also the major contributor with 72.9% in B1 and 97% in A3 (Fig. 4(a)). The higher LU impacts in the wind-based SEMS scenarios stem mainly from the space needed for the installation of wind infrastructure. In contrast, methanol production *via* steam reforming of natural gas requires less land for refineries and transportation infrastructure.

Fossil resource scarcity (FRS) quantifies the consumption of non-renewable fossil resources, typically expressed in kilograms of oil equivalent (kg oil-eq.). It is a key measure of how much fossil fuel reserve is consumed during a process. In this study, FRS impacts are significantly reduced approaching negligible levels primarily due to feed gas recycling, heat integration, and the extensive use of renewable energy (Fig. 4(b)). This highlights the dominant role of feed gas recycling and renewable energy in minimizing fossil fuel consumption in SEMS process.

Water consumption is a critical environmental impact category, measuring the total amount of freshwater used in a process, and is typically expressed in cubic meters (m^3) of water per functional unit. This metric accounts for both direct and indirect water use throughout the life cycle of a product or process, including water for cooling and produced water as a side product in alternative MeOH synthesis. In this study (Fig. 4(c)), the highest impacts are caused by scenario B4 at 6.1 m^3 per t MeOH. Again, the distribution of process impacts varies between the scenarios. In scenario A1, which has the lowest impact, methanol synthesis accounts for 81% of the contribution. This is lower than in A3, where methanol synthesis contributes the highest proportion at 97%.

3.2. Uncertainty analysis

The results of the uncertainty analysis are reported using mean, median, standard deviation (SD), coefficient of variation (CV), and standard error of the mean (SEM). Characterization results for four key impact categories that include GWP, LU, FRS and FE across all eight SEMS scenarios are shown in Table 5. These categories were selected for their relevance to the alternative methanol synthesis process under study, while comprehensive results for all impact categories are provided in Table SII of the ESI.† The variations of the values range from moderate low (CV > 25%) to moderate (CV ≤ 50%), indicating moderate variability that remains within an acceptable range, confirming the reliability of the obtained results. However, in scenarios A1 and B1,

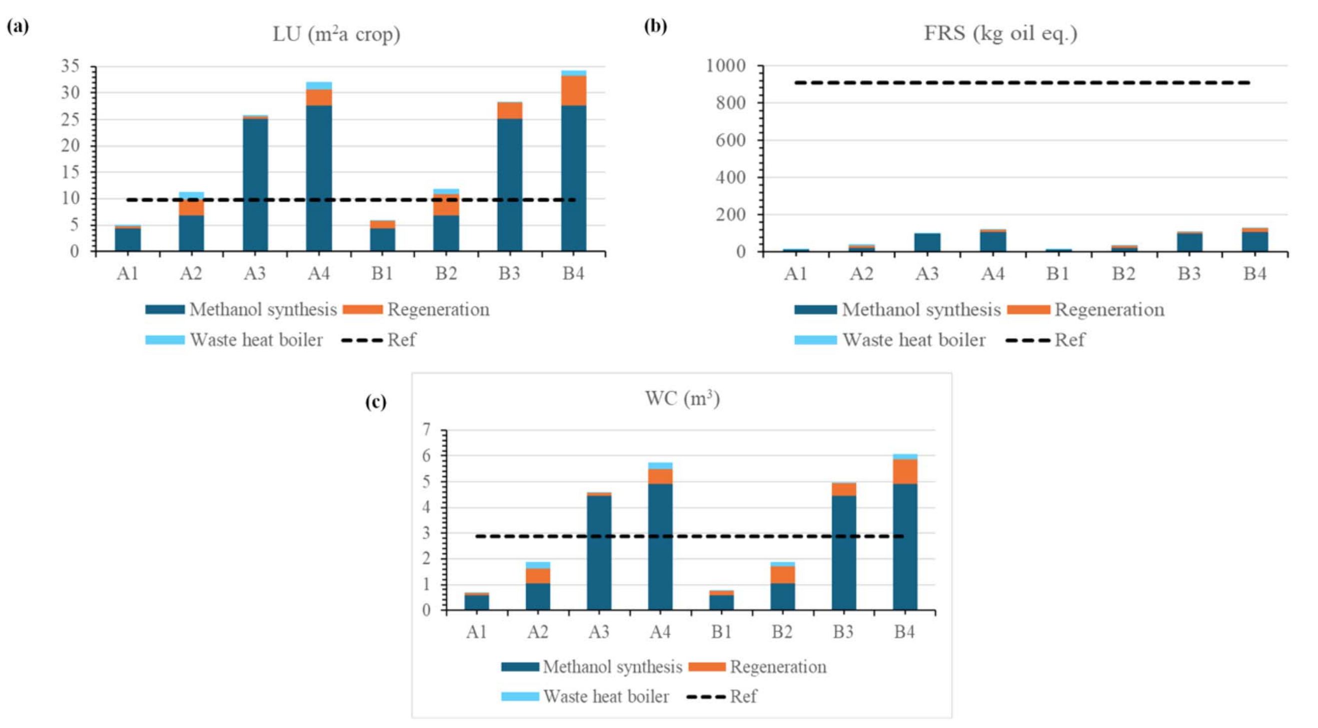

Fig. 4 Process contributions analysis of life cycle stages to the impacts in adiabatic (A1–A4) and isothermal (B1–B4) SEMS scenarios. (a) Land use, LU, (b) fossil resource scarcity FRS, (c) water consumption, WC.

Table 5 Uncertainty analysis results for the eight SEMS scenarios across impact categories: GWP, LU, FRS, and WC, using the Monte Carlo method (SD: standard deviation; SEM: standard error of mean; CV: coefficient of variation)

Scenario	Impact category	Unit	Mean	Median	SD	SEM	CV (%)
A1	GWP	kg CO_2 eq.	60.10	55.65	22.84	0.72	38.00
	LU	m^2a crop eq.	4.28	4.13	1.06	0.03	24.75
	FRS	kg oil eq.	11.88	11.17	4.48	0.14	37.69
	FE	kg P eq.	0.02	0.02	0.01	0.00	59.04
A2	GWP	kg CO_2 eq.	99.79	94.49	30.05	0.95	30.12
	LU	m^2a crop eq.	6.66	6.39	1.64	0.05	24.56
	FRS	kg oil eq.	21.46	20.29	6.77	0.21	31.57
	FE	kg P eq.	0.06	0.06	0.02	0.00	40.40
A3	GWP	kg CO_2 eq.	414.35	367.84	200.79	6.35	48.46
	LU	m^2a crop eq.	25.31	22.95	11.37	0.36	44.93
	FRS	kg oil eq.	98.04	87.24	49.12	1.55	50.10
	FE	kg P eq.	0.40	0.36	0.18	0.01	45.48
A4	GWP	kg CO_2 eq.	446.10	400.55	214.96	6.80	48.19
	LU	m^2a crop eq.	27.76	25.26	12.45	0.39	44.86
	FRS	kg oil eq.	107.17	99.13	50.82	1.61	47.42
	FE	kg P eq.	0.45	0.41	0.21	0.01	46.00
B1	GWP	kg CO_2 eq.	60.08	55.47	22.93	0.73	38.17
	LU	m^2a crop eq.	4.26	4.14	1.02	0.03	24.01
	FRS	kg oil eq.	11.77	10.94	4.27	0.13	36.25
	FE	kg P eq.	0.02	0.01	0.01	3×10^{-4}	56.91
B2	GWP	kg CO_2 eq.	100.21	95.56	30.63	0.97	30.57
	LU	m^2a crop eq.	6.72	6.47	1.64	0.05	24.36
	FRS	kg oil eq.	21.45	20.38	6.83	0.22	31.83
	FE	kg P eq.	0.061	0.06	0.023	0.001	37.54
B3	GWP	kg CO_2 eq.	404.86	361.96	187.83	5.94	46.39
	LU	m^2a crop eq.	25.01	23.13	10.79	0.34	43.15
	FRS	kg oil eq.	96.18	85.41	45.24	1.43	47.04
	FE	kg P eq.	0.41	0.38	0.18	0.01	43.90
B4	GWP	kg CO_2 eq.	454.87	405.92	221.53	7.01	48.70
	LU	m^2a crop eq.	28.13	25.62	12.98	0.41	46.14
	FRS	kg oil eq.	108.53	95.81	52.54	1.66	48.41
	FE	kg P eq.	0.46	0.41	0.23	0.01	50.39

FE exhibits higher uncertainty (CV > 50%). In LCA, estimations of water use for alternative technologies such as SEMS, are sensitive to methodological choices, system boundaries, direct and indirect water use.[35] Inconsistencies in the background data and the characterization factors could further lead to uncertainty. Therefore, improving the Ecoinvent background database is critical for addressing these uncertainties[36] identified in FE impact category. This is particularly important given the water-intensive nature of hydrogen production *via* electrolysis. Overall, the key contributors to model uncertainty are water and electricity consumption, both of which are critical to the sustainability and scalability of alternative methanol synthesis using CO_2 and green hydrogen.

3.3. Sensitivity analysis

The results of the sensitivity analysis for all scenarios and impact categories are presented in Fig. SIII–SIX of the ESI.† In the Figures, SR values are color-coded with green for SR < 0.2, yellow for 0.2 < SR ≤ 0.8, and red for SR > 0.8. Fig. 5 shows the SR values for GWP of all the eight scenarios. The results revealed that hydrogen production *via* electrolysis in methanol synthesis and the heat demand in the regeneration step were the most influential parameters (SR > 0.88) across most SEM scenarios. These factors were particularly significant under varying energy source strategies. In scenarios A2 and B2, which utilized hydropower for hydrogen production and wind power for SEM operation, the SR values for hydrogen varied from 0.24 to 0.57 (Fig. 5), reflecting the increased relative impact of electricity use in SEM operation. Consistent with uncertainty analyses results in Section 3.2, electricity demand associated with water electrolysis is the dominant factor affecting system-level environmental performance of SEM process. This influence is primarily attributed to the substantial electricity consumption of the electrolysis process. In addition, heat demand during regeneration was consistently significant across most of the SEMS scenarios and impact categories, highlighting the critical role of thermal integration measures implemented in this study to improve overall system efficiency.

4. Discussion

This study presents a comparative LCA of sorption-enhanced methanol synthesis (SEMS) under various scenarios, emphasizing the environmental impacts of different operating conditions, specifically comparing adiabatic (A1–A4) and isothermal (B1–B4) configurations. The results presented in this study are significantly influenced by process optimization of various

Scenario	A1	A2	A3	A4	B1	B2	B3	B4
Methanol synthesis								
CO_2	0.10	0.45	0.02	0.10	0.10	0.45	0.02	0.10
H_2	0.89	0.54	1.98	1.79	0.89	0.54	0.98	0.89
Electricity	0.00	0.01	0.00	0.00	0.00	0.01	0.00	0.00
Heat	0.77	0.66	0.33	0.31	0.78	0.67	0.34	0.32
Regeneration								
CO_2	0.30	0.30	0.29	0.30	0.18	0.27	0.05	0.18
H_2	0.01	0.00	0.04	0.01	0.38	0.08	0.82	0.39
CO	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00
H_2O	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Electricity	0.69	0.70	0.67	0.69	0.42	0.64	0.12	0.43
Heat	0.99	0.95	0.99	0.95	0.99	0.95	0.96	0.92
Waste heat combustion								
CO_2	0.63	0.93	0.24	0.70	0.40	0.83	0.11	0.48
H_2	0.26	0.05	0.72	0.29	0.41	0.11	0.83	0.49
CO	0.11	0.02	0.04	0.02	0.15	0.04	0.03	0.02
H_2O	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Fig. 5 Sensitivity ratio (SR) for global warming potential (GWP) of eight SEMS scenarios (green for SR < 0.2, yellow for 0.2 < SR ≤ 0.8, and red for SR > 0.8).

integrated processes such as methanol synthesis unit, and regeneration.[20] Heat integration was instrumental in reducing energy consumption and improving the overall efficiency of the system, which led to reductions across all environmental impact categories. The potential for energy savings, as observed in this study, aligns with findings from Zhang *et al.*,[37] who demonstrated that heat integration can lead to energy savings of 23.7%. This provides a reliable benchmark for the effectiveness of process optimization in SEMS systems. Thus, in the present study, the impact of both corresponding adiabatic and isothermal SEMS scenarios are almost identical.

Overall, findings in this research reveal notable environmental advantages for the SEMS systems over the conventional process, with those utilizing hydropower performing better than those based on wind electricity in all the impact categories. The GWP is the impact category that has been studied the most among all LCA studies. In this study, the obtained GWP values of all SEMS scenarios (71–529 kg CO_2 eq. per t MeOH) are lower than value reported by,[27] who obtained 686.44 kg CO_2 eq. per t MeOH. Comparing the results with,[18] it is clear that significant portion of CO_2 emissions in all the scenarios are attributed to the electricity demand for hydrogen production by water electrolysis. In their system,[18] a 77% reduction in fossil depletion is obtained compared to 85.7–98.5% obtained in our study. Similar to our approach, system expansion of recycling materials and avoidance of external heat usage has been included. Therefore, with the same approach and taking into account environmental savings from outlet gas utilizations into this impact category, the results are consistent. As mentioned in Section 2, the main co-product in this study are CO_2, H_2, CO, and H_2O, which are further utilized in the downstream processes. Therefore, by accounting for the utilization of these co-products, the environmental savings from this approach are reflected in the results, ensuring consistency with the overall sustainability goals of the study, and the principles of circular economy.

Similar to findings reported in ref. 18, our study also shows that while alternative methanol production significantly reduces fossil fuel depletion, water consumption does not always decrease. In four out of eight scenarios, WC remained higher than reference case, aligning with previous studies that attribute this to the water demand of hydrogen production *via* electrolysis. This highlights a need for further optimization of electrolysis efficiency and water management strategies to enhance overall sustainability of SEMS process. In comparing the results of scenarios using the same reactor configuration, the scenarios using wind electricity results in higher environmental impacts than those with hydropower. This difference can be primarily attributed to the life-cycle characteristics of the respective energy sources. Wind energy systems typically have nearly 87% of lifecycle GWP emissions from production of tower (48%) and foundation (39%).[38] Conversely, hydropower systems generally benefit from lower life-cycle impacts, particularly when infrastructure is amortized over a long operational lifespan.[39] Furthermore, the intermittency of wind power may necessitate additional grid management or storage systems, further influencing the overall impact.[40] In this study, the obtained GWP of the adiabatic scenarios (A1–A4) ranged from 71–519 kg CO_2 per t MeOH, and the isothermal scenarios (B1–B4) ranged from 72–529 kg CO_2 eq. per t MeOH. These results were consistent with the literature, where the utilization of wind energy was responsible for the higher GWP impact contribution of methanol synthesis.[41] Therefore, these results emphasize the necessity of integrating detailed energy source-specific analyses into sustainability assessments of chemical production pathways to optimize environmental performance. Moreover, our study, using 100% renewable electricity in Finland, demonstrates that adiabatic SEMS scenario utilizing hydropower emits 12.3 kg CO_2 per MWh and 13 kg CO_2 per MWh in the isothermal scenario. In comparison, in the adiabatic SEMS scenario using wind power emits 90.2 kg CO_2 per MWh and 95.7 kg CO_2 per MWh in the isothermal scenario. In e-methanol pathway, where the manufacturing phase is not considered, the emissions reported by the Methanol Institute are approximately 0.92 kg CO_2 per MWh for wind power and 1.76 kg CO_2 per MWh for hydropower-based production.[42] This highlights the importance of taking the electricity source's manufacturing impact when producing fuels with renewable electricity.

In general, emissions of NO_x, phosphorus and nitrogen in SEMS process are minimal. However, indirect emissions arise from upstream industrial activities associated with the production of wind turbines and hydropower infrastructure as contained in the ecoinvent dataset.[29] These emissions were characterized using the ReCiPe method, which assigns impact factors to emissions based on their potential to contribute to various environmental and health-related effects.[30] Thus, the results are consistent with the life cycle inventories available in ecoinvent, where wind power generally has higher impacts due to indirect emissions from turbine manufacturing, transportation, and installation.[29]

The revised Renewable Energy Directive (RED II)[43] sets a minimum 70% GHG emission savings threshold for renewable fuels of non-biological origin (RFNBOs), using a fossil fuel comparator of 94 g CO_2 eq. per MJ. The LCA results of this study indicate GHG emissions between 3.6 and 26.7 g CO_2 eq. per MJ, corresponding to 96.1% to 71.6% GHG savings respectively. The best case scenario, achieving 96.1% savings, is based on hydropower, while the worst case scenario, achieving 71.6% savings, is based on wind electricity. However, as the EU is increasingly prioritizing wind energy as a key component of its renewable energy strategy, future scenarios may shift as wind technology continues to improve and economies of scale are realized. With the European Union's goal of installing 300 GW of offshore wind capacity by 2050,[43] there is a clear need for ongoing assessment of renewable energy sources, particularly in their potential contribution to alternative methanol synthesis, one of the key pathways to decarbonizing industrial processes.

5. Conclusions

This study performed a comparative LCA of adiabatic and isothermal sorption-enhanced methanol synthesis. Eight scenarios were analyzed, differing in reactor configurations, electricity sources for hydrogen production, and in SEMS process. The LCA results indicate that both corresponding adiabatic and isothermal SEMS scenarios are almost identical. Scenarios utilizing hydropower exhibited the lowest overall environmental impact. These reductions were driven by renewable energy use, feed gas recycling, and avoided external heat use. The adiabatic SEMS scenarios exhibit GWP values ranging from 71.7 to 519.7 kg CO_2 eq. per t MeOH, and the isothermal SEMS scenarios range from 72 to 529 kg CO_2 eq. per t MeOH, both representing substantial reductions compared to the reference case of 980 kg CO_2 eq. per t MeOH from steam reforming of natural gas. Furthermore, all SEMS scenarios achieve GHG emission savings between 71.6% and 96.1% relative to fossil fuels, thereby meeting the 70% savings threshold for sustainability set by the RED II. In addition to reducing GHG emissions, 61% of the impact categories across all scenarios show lower values than the reference process. The impact categories of marine eutrophication, and fossil resource scarcity have shown significant improvement overall. However, impacts on ozone formation, freshwater eutrophication, land use, and water consumption varied depending on electricity sources and reactor designs. These findings highlight the need for careful optimization to minimize trade-offs in these categories. Future research incorporating full life cycle impacts, including the methanol use phase, would provide a more comprehensive environmental assessment.

Data availability

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

Author contributions

A. R. Dahiru: conceptualization, methodology, visualization, validation, investigation, formal analysis, writing – original draft. A. Vuokila: conceptualization, validation, methodology, investigation, writing – original draft, writing – review & editing, supervision. E. Laasonen: writing – review & editing. A. Laari: conceptualization, writing – review & editing. T. Koiranen: conceptualization, supervision. M. Huuhtanen: conceptualization, writing – review & editing, supervision.

Conflicts of interest

The authors declare no conflicts of interest.

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