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Production of carbon nanotubes from captured carbon: An ex-ante life cycle assessment case study

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ABSTRACT

Carbon capture and utilization (CCU) plays a key role in reducing greenhouse gas emissions and reaching carbon neutrality goals. This study assesses the environmental impacts of producing carbon nanotubes (CNTs) via molten salt CO_2 capture and electrochemical transformation (MSCC-ET) and Catalytic Chemical Vapor Deposition (CCVD), both using CO_2 as feedstock. We screened and selected technologies using a parameter-based method and conducted process modeling and an ex-ante Life Cycle Assessment (LCA) to move beyond lab-scale evaluations and included the purification steps. The MSCC-ET approach showed advantages in reducing impacts related to climate change, energy, and ozone depletion impacts, specifically suggesting lower CO_2 emissions. However, traditional CCVD outperformed MSCC-ET in several other impact categories. Key contributors to the environmental impacts of MSCC-ET were found in high material use in electrolysis, purification, and electricity consumption, modeled using the Belgium grid. Moreover, including the carbon capture unit in the assessment could provide a complete view of the environmental impacts regarding CCU. For every 100 tons of MWCNTs produced, a monoethanolamine (MEA)-based carbon capture unit integrated into the MSCC-ET system could roughly reduce 716.5 tons CO_2-eq. Additionally, electricity consumption was found to constitute a significant portion of the environmental impacts. Therefore, a sensitivity analysis was conducted, revealing that changes in the electricity source, catalyst used, and CO_2 source significantly influenced the environmental performance of both technologies. Furthermore, we summarize practical insights from this study to guide the effective application of ex-ante LCA for carbon nanomaterials. The paper concludes with actionable recommendations for early-stage technology developers on optimizing energy use, improving material efficiency, and integrating recycling to enhance sustainability. In addition, we provide recommendations for LCA practitioners on incorporating dynamic systems to transition from lab-scale to industrial contexts, thereby bridging the gap between research and practical implementation.

1. Introduction

Carbon Capture, Utilization, and Storage (CCUS) technologies as critical tools for reducing carbon emissions have brought significant attention in recent years [1]. Among CCUS, Carbon Capture and Utilization (CCU) specifically enables the long-term substitution of fossil carbon in sectors that rely on carbon, while offering the potential to reduce the fossil-based footprint of the chemical derivatives sector [1]. One encouraging application of CCU is the conversion of CO_2 into carbon nanotubes (CNTs), which possess unique structural, electrical, mechanical, and chemical properties [2]. These properties make CNTs highly suitable for various applications, including electronic devices, sensors, batteries, etc [3]. Therefore, the demand for CNTs is rising rapidly, with global production expected to exceed annual kiloton levels, and expected to reach 7,000 tons by 2025 [4], driven by numerous applications in modern technology [5–9], as new manufacturers enter the market and existing ones expand [10]. Moreover, the global CNT market value also grew from $1 billion in 2014 to $4.55 billion in 2018, and is projected to reach approximately $15.02 billion by 2026 [4].

Traditionally, CNTs are produced using methods such as electric-arc discharge, laser ablation, thermal synthesis, chemical vapor deposition

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(CVD), and plasma-enhanced chemical vapor deposition, which often rely on fossil-fuel-based hydrocarbons [11,12]. Among these, CVD is favored for producing high-quality CNTs for industrial applications [13]. Alternative methods to produce CNTs from CO_2 feedstock include catalytic [14,15], electrochemical [16,17], microbial [18,19], mineralization [20,21], and photocatalytic pathways [22,23], each with varying levels of technological maturity.

The conversion of CO_2 into CNTs and similar carbon nanomaterials is mainly achieved through three pathways: metallothermic reduction, liquid metals, and electrochemical reduction. Metallothermic reduction reactions (MRRs) involve the use of reactive metals to reduce compounds and produce metals, alloys, nonmetal substances, and composites [24–26]. This method is now being explored for producing carbon nanostructures from CO_2, using metals such as Li, K, Ca, Na, and Mg under high temperatures and pressure [27,28]. These reactions avoid gaseous by-products, reducing the risk of explosions or fire. In metallothermic reduction reactions, although the reaction conditions such as temperature, pressure, and choice of metal reductants can be easily controlled, the resulting CNTs are produced in a mixture of other carbon nanoporous structures. As a result, the formation of a higher yield of CNTs synthesized through metallothermic reduction reactions remains unclear, and more effort is required to advance this emerging field [28].

Using liquid metals to transform CO_2 into carbon nanoporous structures is a novel area of research. This approach uses mechanical energy and liquid metals such as gallium and silver to reduce CO_2 [29–32]. Liquid metals offer advantages over solid catalysts by resisting active site deterioration [33]. Gallium (Ga)-based liquid metals exhibit intriguing catalytic features, such as tunability through integrating other elements and extraordinary resistance to coking [34–36]. Although liquid metals reduction of CO_2 provides a viable pathway to form carbon nanoporous structures at low or room temperatures, further research has to be conducted to analyze the morphology and yield of the different carbon nanoporous structures obtained. As a result, the large-scale production of solid carbonaceous species remains ambiguous under current circumstances.

Another viable pathway for the formation of CNTs using CO_2 input feedstocks has been the electrochemical reduction of CO_2 using molten carbonate solutions. The electrochemical conversion of CO_2 into value-added commodities has long been regarded as one of the most effective methods of digesting massive amounts of anthropogenic CO_2 [37]. In general, the efficacy of a CO_2 reduction reaction is determined mostly by the electrolyte [38], catalyst [39], temperature [40], and various stimuli such as potential, electric field, light, and mechanical force [41]. Among these parameters, the electrolyte plays a significant role in regulating the CO_2 reduction reaction in various systems ranging from room to high temperatures [37]. Electrolytes are classified into three types based on their working temperatures: room-temperature electrolytes such as aqueous solutions, liquid organic electrolytes [42], solid polymer electrolytes, ionic liquids [43], high-temperature solid oxide electrolytes, and high-temperature molten salt electrolytes [37].

The molten carbonate CO_2 (MCC) electrolyzer, unlike the room-temperature and solid-oxide CO_2 electrolyzers, does not require an ion-selective membrane to prevent electrolytic product crossover between the anode and the cathode because the cathodic products are electrolyte-insoluble solid carbonaceous materials and the anodic product is oxygen [40]. The last decade has focused on developing molten carbonates CO_2 electrolyzers, such as oxygen-evolution inert anode materials, cathodic product modulation, electrolyte engineering, and so on [37].

Despite advancements, limited research exists on the environmental impacts of mass-producing CNTs using CO_2 [44–46]. Conducting a life cycle assessment (LCA) is helpful in identifying the environmental hotspots associated with CNT production. However, most existing LCA studies are limited to laboratory-scale or small-scale production, typically considering the production of CNTs at the kilogram level [47,48], and do not include other critial processes such as purification steps. Scaling from lab to industrial production also presents challenges, such as optimizing material use and dealing with fossil-fuel-intensive electricity sources [49]. In addition, upstream activities related to these material inputs contribute significantly to the bulk of the impact categories [50]. When economies of scale are considered, perhaps a minimum production threshold could allow for the addition of recycling streams to optimize resource use.

Cucurachi et al. claim that ex-ante LCA should make it possible to anticipate probable preventable environmental impacts, and prevent environmental lock-ins [51]. Ex-ante LCA guarantees the evaluation of a technology's possible environmental impacts early in its development curve when less information is accessible and available, and opportunities for change can still be considered. Technology developers can take necessary action early in technology development, guided by the findings of LCA studies, to make investments in technologies or improvements that ultimately reduce their environmental impact. Therefore, early process decisions significantly affect the functional features and environmental impacts on technology systems [52]. Understanding how design decisions affect a technology's predicted environmental performance may help prevent avoidable environmental burdens, lower costs, avoid unwanted expenditures and replacements, and anticipate changes in environmental rules and regulations [51].

This research applies ex-ante LCA focuses on an industrial-level production scale to evaluate environmental impacts, operational efficiencies, and scalability challenges associated with CNT production using the cradle-to-gate approach. The primary aim is to evaluate and compare the environmental impact of producing CNTs using CO_2 as a feedstock via the molten salt CO_2 capture and electrochemical transformation (MSCC-ET) method versus the catalytical chemical vapor deposition (CCVD) method. Sections 2.1 to 2.3 detail the selection criteria and pathways methods for CNT production. Section 2.4 provides information on ex-ante LCA and the industrial-scale fabrication processes involved. Section 2.5 considers the influences of the carbon capture unit. Furthermore, the study outlines additional insights to capture and utilize learning opportunities for future ex-ante LCA studies of this type. In the last part of the paper, we developed a set of recommendations, categorized into suggestions for early-stage technology developers and LCA practitioners (model-based and ex-ante methodology-based).

2. Methods

2.1. Parameter selection

Electrochemical reduction of CO_2 presents numerous advantages over metallothermic and liquid metal reduction methods for synthesizing carbon nanotubes. Additionally, significant research has been conducted on the viability of MSCC-ET methods to produce CNTs and a wide range of other carbonaceous products [53]. As a result, a novel method was selected to produce carbon nanotubes using MSCC-ET techniques. There are several parameters based on which the growth of CNTs is achieved using the MSCC-ET method. We list these parameters to provide a narrower focus for selecting a novel technology system. The selected parameters and their description are proposed in Table 1. The detailed technology selection can be found in the Supporting Information (*SI*)-S1 part.

Based on the parameters highlighted above, seven novel technology pathways were screened. These pathways are highlighted in *Supplementary Material 1(SM1)* and summarized in Table 2. Of the seven studies presented, six used Li_2CO_3 as the electrolyte of choice.

The selected novel technology system is illustrated in Fig. 1. The chosen novel technology system employs the use of barium carbonate and sodium carbonate-based electrolytic solution and brass and Inconel 718 as the cathode and anode, respectively. The reaction occurs at an operating temperature of 770℃ to produce MWCNTs of 5–20 nm in diameter. The experimental purity obtained was 90% MWCNTs with

Table 1
Parameters chosen for the selection of the novel technology.

NO.	Parameter	Description	References
1	Process type	The process type describes whether the electrochemical reduction process takes place in a continuous or batch operation. The flow of a single product unit between each phase of the process without pausing in time, substance, or length is a continuous process. Each device in the continuous process has a specific processing function and runs steadily. A batch process involves a series of processes that must be completed in a precise order. Because there is a time lag between distinct parts of the process in a batch process, shutdown time occurs frequently.	[54–56]
2	Products produced	Products produced may include multi-walled carbon nanotubes (MWCNTs), single-walled carbon nanotubes (SWCNTs), carbon spheres, and hollow carbon nanotubes. Based on specific operating parameters such as temperature, the input source of hydrocarbon stream and the type of catalyst, the resulting carbon nanotube type varies.	[57–59]
3	Purity of CNTs obtained	A typical electrochemical reduction process produces a mixture of CNTs and byproducts. The purity of CNTs indicates the percentage purity of pure carbon present in a sample containing a mixture of CNTs and other byproducts.	[60,61]
4	Type of molten carbonate	The type of molten carbonate used influences the morphology of the carbon nanotubes and its growth process at the cathode. The molten carbonates possess high ionic conductivity and low vapor pressure to reduce CO_2.	[62,63]
5	Type of CO_2 source	Multiple sources of CO_2 are available for CCU pathways, and the selection of the type of CO_2 source would influence the associated emissions for the CCU pathway under study.	[64]
6	Operating temperature	The reaction temperature influences the alignment characteristics and diameter of the synthesized nanotubes.	[65]
7	Size of CNTs produced	SWCNTs typically have diameters of 0.5–1.5 nm MWCNTs have diameters ranging from 10 to 100 nm. MWCNTs with more than 100 nm diameters can also be formed by altering operational conditions.	[66,67]
8	Technology Readiness Level (TRL)	The Technology Readiness Levels present levels between 1–9. The higher the levels, the more mature the technology is. The TRL scale measures a technology's maturity with an eye toward operational application in a system setting.	[68]
9	Duration of electrolysis	The duration of the current supplied to the electrolysis reaction can range from several minutes to hours. The duration of the current supplied to the electrolyzer influences the morphology and the growth process of the CNTs.	[62]
10	Current density/ Voltage/Current applied	The amount of current supplied to the electrolyzer influences the growth and morphology of the CNTs formed on the active area of the cathode.	[17]
11	Byproducts	Amorphous carbon and small quantities of cathode-based metal particles can be found in the solid carbon obtained as part of the electrolysis reaction.	[69]
12	Operating pressure	The operating pressure is assumed to be 1 atm unless otherwise stated.	[62]
13	Anode and cathode material	The growth of CNTs is influenced by the cathode.In contrast, electron transfer for the reduction of CO_2 to form CNT takes place at the anode.	[70]
14	Proposed application	Based on factors such as purity, morphology, and size of the synthesized CNTs.	[70]

byproducts surfacing from the brass cathode. While the use of lithium carbonate as an electrolyte is found to be the most common route to produce CNTs using CO_2 as an input feedstock, the non-lithiated electrolyte has the singular advantage in its unit economics – the costs of using barium carbonate and sodium carbonate per ton is 97.7% and 96.4% cheaper than that of lithium carbonate [73].

Moreover, given that lithium is a critical material required for the transition of the global economy to renewable energy technologies, the production volume of lithium saw the greatest increase of 208% in the recent decade [74]. As a result, the concentration of lithium-based exports and imports is high in upstream supply chain segments. This has resulted in export restrictions being placed. 10% of the global critical raw materials are subjected to at least one export restriction measure – with export restrictions on ores and minerals growing faster than other supply chain segments [75]. This affects both the availability and prices of these materials. Therefore, using barium and sodium carbonate as electrolytes is well-positioned for fabricating CNTs using CO_2 as an input source.

2.2. Selection of conventional technology systems

Similarly, we proposed a selection for a conventional CVD method using the parameter selection method (see *SI*-S2.1, 2.2 and Table S1). The selected technology (also referred to as the reference or conventional technology in this study) system employs a CCVD technique to manufacture carbon nanotubes using natural gas as the hydrocarbon source, as can be seen in Fig. 2. The continuous production of MWCNTs was achieved using transition metals such as nickel and molybdenum on magnesium oxide support as the catalyst at an operating temperature of 975℃. The process takes place in an incline-mobile bed reactor, and the diameter of the produced MWCNTs ranges between 5 to 20 nm [76]. A high carbon production of MWCNTs corresponded to a low purity level of 90%. The byproducts produced materialize from the Ni-Mo/MgO catalyst. The advantage of using natural gas as the hydrocarbon source instead of ethylene lies in unit economics – where the cost per ton of natural gas is 99.9% cheaper than that of ethylene [77,78]. More importantly, although ethylene is a more prevalent hydrocarbon source than natural gas, the natural gas network is better developed. The advantages of using an inclined mobile-bed reactor lie in its adaption to suit the sluggish kinetics of CNT synthesis and its large product-catalyst volumes [76]. Additionally, the use of Ni-Mo/MgO as the catalyst of choice improves CNT selectivity and catalyst surface renewal characteristics, which are important factors, especially when economies of scale are applied. This resulted in the selection of the novel and conventional technology systems presented by Wang et al. using MSCC-ET

Table 2
Seven novel technology pathways assessed in this study.

Metal Used	Electrolyte	Temp (°C)	Current Density (mA/cm^2)	Time (hours)	Average Diameter (nm)	Yield (%)	Notes	References
Stainless steel, brass	Li_2CO_3	750	100	1	Steel: 29, Brass: 126	–	Utilized low-value waste metals, synthesized MWCNTs for battery electrodes and sensors.	[17]
Various metal wires	Li_2CO_3	750	100	1	27.5	99	Blended standard gas phase with electrolytic growth, used ALD for Ni anode coating.	[62]
Ni nanoparticles	Li_2CO_3, Li-Na carbonate	500–750	50–300	–	24–69, 100–200	> 90	Explored different current densities and electrolyte compositions for CNTs, CSs, and honeycomb structures.	[71]
Inconel, Muntz Brass	Li_2CO_3 with Fe_2CO_3 additive	770	0.15	4	100–500	97	Focused on high-purity CNTs, used C_2CNT process for nano-filtration applications, aligned CNTs.	[70]
Not specified	Li_2CO_3 with additives	–	0.1	4	22–42	>90	Achieved thinner CNTs with Li_2O and calcium metaborate, observed synergy between calcium and metaborate.	[72]
Steel electrodes	Li_2CO_3 with Li_2O	750	–	1.5	–	–	Employed sun thermal electrochemical process (STEP), optimized CNTs and CNFs for battery applications, and focused on energy storage capacities and defect control in synthesized nanostructures.	[2]
Nichrome C, brass	Na_2CO_3, $BaCO_3$	770	0.1	5	5–20	80–90	Used non-lithiated electrolyte, demonstrated MWCNT synthesis with atmospheric CO_2.	[73]

method [73] and Douven et al. using the CCVD method [76], respectively.

2.3. Supporting purification systems for technology pathway

For the selected novel technology pathway by Wang et al., they reported the details of the process engineering focusing on the electrolyzer, fabricating MWCNTs from flue-gas-based CO_2 as the CO_2 source [73]. We modified the supporting purification process engineering systems to model a system representing an industrial-scale (continuous fabrication) MSCC-ET process, with several purification steps as introduced in the subsections from 2.3.1 to 2.3.5. Similarly, for the conventional technology pathway, the supporting process can be found in *SI*-S3. The processes are illustrated in Fig. 3A for the novel technology pathway and Fig. 3B for the conventional one. Meanwhile, we carried out mass balance calculations for both technology pathways in *SM2* and *SM3*. However, the capture of CO_2 from the industrial flue gas is not described in their study since the researchers used atmospheric CO_2 [73].

2.3.1. Acid treatment process

The acid treatment procedure is the initial stage in the purification of MWCNTs and other byproducts. MWCNTs and byproducts – metal particles from the cathode plate and electrolyte – are obtained during electrolysis. Mortazavi et al. described soaking the MWCNTs and byproducts in 5 M sulphuric acid at 337°C. The MWCNTs and byproducts are immersed for 3 h to minimize MWCNT degradation [69]. Even though Mortazavi et al. outlined a process for purifying MWCNTs and byproducts derived through a conventional technology approach such as CVD [69]. The acid treatment technique described by Mortazavi et al. was applied to purify MWCNTs and byproducts produced by the novel technology approach. In this study, the acid treatment apparatus is the same for the conventional technological pathway as described in the *SI*-S3.3 part.

2.3.2. Froth flotation process

Froth floatation is the second stage of the purification process in the novel technology pathway. The froth flotation approach extracts the MWCNTs from the numerous by-products created during the acid treatment process [49]. Surfonic L24-7 was chosen as a sample non-ionic surfactant by Lertrojanachusit et al. [49]. In a liter of Surfonic L24-7, 0.25 g of solid content (MWCNTs and other by-products) were mixed, and 250 mL of the mixed solution was transported to a froth floatation column with air injected at the bottom. A mass flow controlled was used to set the airflow rate at 170 cm^3/min, and was operated at room temperature. At regular intervals, the collapsed foam was scraped off. In this study, the froth floatation technique was modified for the novel technology pathway [49]. The apparatus used for the froth floatation process is the same as described in the *SI*-S3.4 froth floatation for the conventional technological pathway.

2.3.3. Washing

The surfactant Surfonic L24-7 was removed from the collapsed froth by washing it multiple times in distilled water until the pH of the solution is neutral. The device used for the washing process was the same as the one described in the *SI*-S3.5 Washing for the purification of MWCNTs obtained using the conventional technological pathway.

2.3.4. Solid-liquid drying

Solid-liquid drying is the last step in the purification process. According to Mortazavi et al., drying was carried out at 130°C for 8 h in an oven. The dried mixture comprises less than 10% metal particles and 90% pure MWCNTs [69]. The drying apparatus used to purify MWCNTs in the novel technology pathway is the same as described in the *SI*-S3.6 Solid-liquid drying.

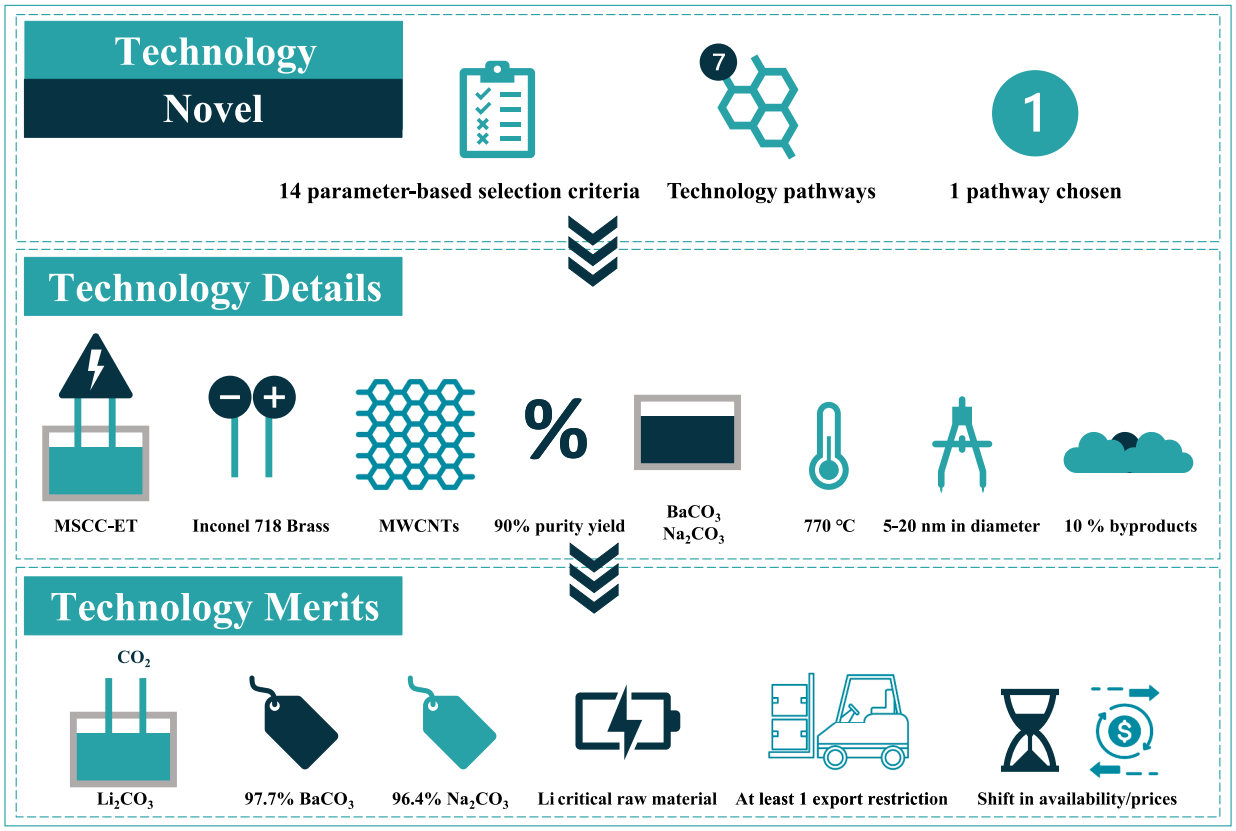

Fig. 1. Technology screening and selection of novel technology pathway.

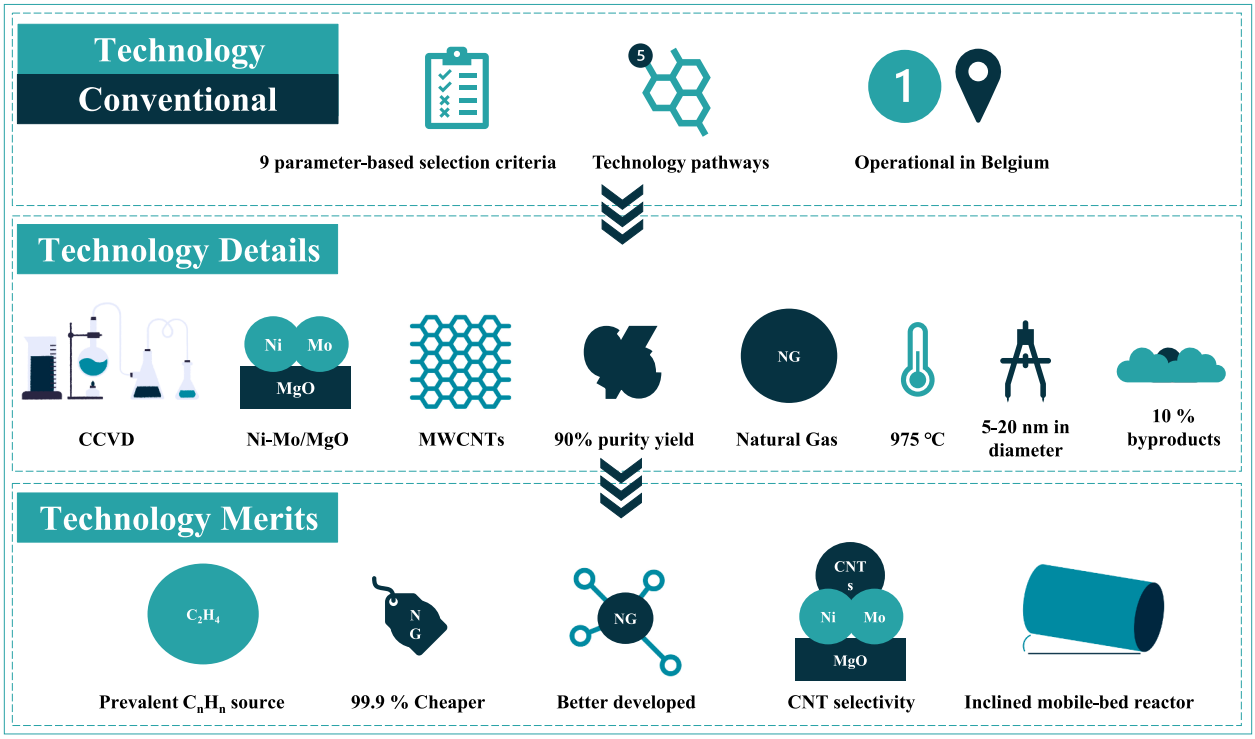

Fig. 2. Technology screening and selection of conventional technology pathway.

2.3.5. Compressors and heat exchangers

Centrifugal air compressors can handle high flow rates at a range of pressures and can run continuously for extended periods. In addition, they are more compact, need fewer maintenance visits, and use fewer consumables than similar products. The compressor used in the novel model was the centrifugal compressor, with an efficiency of 0.85. The

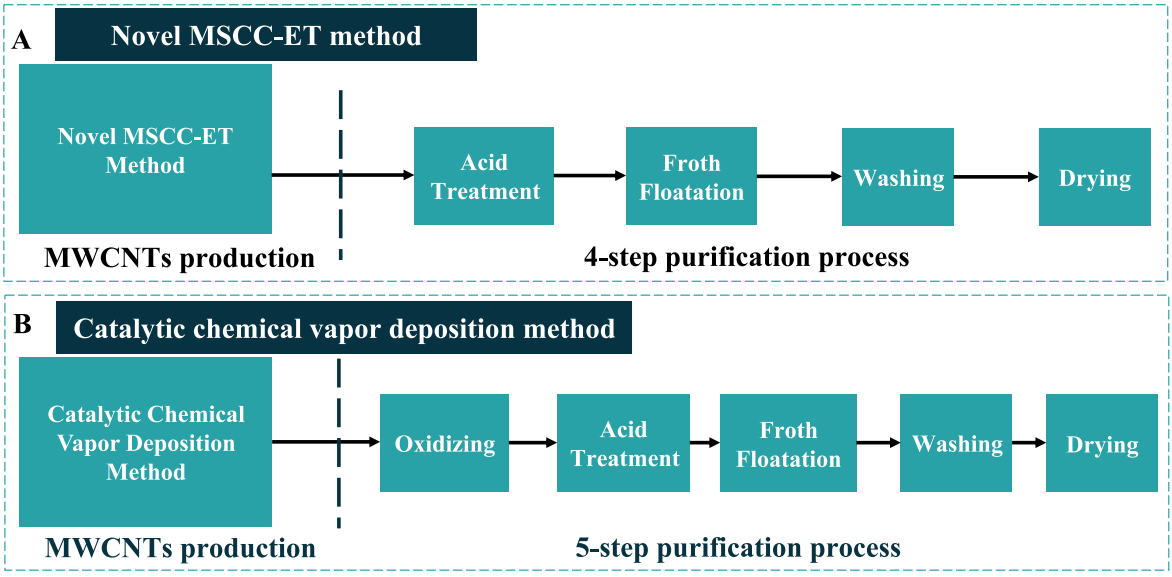

Fig. 3. (A) the processes and supporting systems for the novel MSCC-ET technology pathway and (B) the processes and supporting systems for the conventional CCVD technology pathway.

devices used to compress gases (heating and cooling gasses), and liquids were the same as those described in the *SI*-S3.8 Compressors and 3.9 Heat exchangers, respectively, with Fig. S1 and S2 for illustrating the process flow diagram of a heat exchanger (heating or cooling) to a material.

2.3.6. *Additional steps for the CCVD pathway*

The additional oxidation, use of cyclone separator and pressure swing adsorption (PSA) steps in the CCVD purification process served specific purposes that reflect the differences in impurity types between CCVD and MSCC-ET processes. The oxidation step in CCVD was employed before the acid treatment to selectively remove amorphous carbon and other loosely bound graphitic impurities [49]. This could prepare the material for acid treatment by ensuring that the acids were more focused on removing metal catalysts rather than dealing with a mixture of carbon types (refer to *SI*-S3.2).

The cyclone separator, on the other hand, was used after drying steps to physically separate larger particulate impurities [79,80]. In MSCC-ET, these impurities are generally lower due to the nature of the molten salt process, making such extensive purification steps unnecessary. Following the application of compressor, PSA was used to purify the hydrogen produced during the thermal decomposition of methane. PSA can separate hydrogen from the natural gas mixture, which contains unreacted methane and other components such as nitrogen, ethane, CO_2, and propane. This method effectively adsorbs these heavier gases under high pressure which allows pure hydrogen to be recovered and reused in the process (see *SI*-S3.10) [81].

2.4. *Ex-ante life cycle assessment (ex-ante LCA)*

Ex-ante LCA performs an LCA of a new technology before it is commercially implemented in order to inform R&D decisions to make this new technology environmentally competitive as compared to the incumbent technology mix [82]. This study follows the framework of ISO 14040, including goal and scope definition, inventory analysis, impact assessment, and interpretation [83]. Where necessary, it expands upon ISO 14044 to accommodate the prospective nature of ex-ante LCA [84]. We refer to the individual sections for the relevant assumptions and further to the supplementary materials, which include mass balance calculations, and the relevant specific technological descriptions with assumptions.

2.4.1. *Goal and scope*

The goal of the LCA study is to evaluate the environmental impacts of MSCC-ET for converting CO_2 into CNTs, comparing it with a conventional CVD CNT fabrication technology. The study is comparative in nature and follows the definition of ex-ante LCA as defined by Cucurachi et al. [51]. The geographical scope for both systems is Europe, and specifically Belgium. The study covers background processes for resources such as electricity, water, and raw materials in a temporal period of 2022–2023. The study focuses on MWCNTs with a 5–20 nm diameter. The novel technology system was based on the research of Wang et al. [73], with a TRL between 3–4, while for the reference system we considered the study of Douven et al. [76], with a TRL of 9. As a result, utilizing upscaling methods based on expert consultancy from the industry, the novel technology system was scaled up to match the conventional technology system for comparison. Notably, carbon dioxide capture and catalyst preparation were not included. Furthermore, both systems' post-growth purification processes were modeled, as introduced above. The study applied the CML impact assessment methodology. Additionally, a second regionally relevant methodology was used for comparability and geographical representativeness, detailed in the sensitivity analysis section.

The system boundary is a cradle-to-gate approach. The functional unit is defined as the production of 100 tons of MWCNTs with a 90% purity. An example of the use case of 90% purity yield of MWCNTs obtained is in cementitious materials, where the impurities present do not pose a perceived risk in its application [85]. In the use case scenario, the products obtained from the novel and conventional technology systems appear identical in their chemical structure and composition, adhering to the functional unit recommendations of Langhorst et al. [86] and Ramirez Ramirez et al [87]. Therefore, the chosen 90% purity yield of MWCNTs suggests that 90% of the product obtained from the novel and conventional technology processes comprises MWCNTs, with the remaining 10% representing residual particles of the cathode and spent catalyst, respectively. The life cycle impact assessment (LCIA) was performed with the CML v4.8 impact assessment family [86]. It divides the impact into 11 midpoint impact categories: AC (acidification) – kg SO_2 eq; CC (climate change) – kg CO_2 eq; FETP (freshwater ecotoxicity) – kg 1,4 DCB eq; METP (marine ecotoxicity) – kg 1,4 DCB eq; TETP (terrestrial ecotoxicity) – kg 1,4 DCB eq; NRER (non-renewable energy resources) – MJ; EP (eutrophication) – kg PO_4 eq; HTP (human toxicity) – kg 1,4 DCB eq; MMR (material resources, metals/minerals) – kg Sb

eq; ODP (ozone depletion) – kg CFC-11 eq; POFP (photochemical oxidant formation) – kg ethylene eq.

Fig. 4 provides an overview of the process flow diagrams created for the novel and conventional reference technology systems. Only the foreground processes of both systems are presented in Fig. 4A and B to ensure readability. Complete flowcharts, including background activities, are presented in *SM4* and *SM5*. Additionally, heat exchangers are crucial components in chemical processes. However, their simulation in LCA is sometimes overlooked. The modeling of heat exchangers required for heating and cooling purposes is illustrated in *SM6*. As illustrated in Fig. 4A, the CO_2 gas enters the electrolysis chamber, where the MWCNTs, impurities from the cathode, and byproducts such as O_2 and unused CO_2 are formed. The byproducts leave the electrolysis chamber as waste, whereas the MWCNTs and the cathode-based impurities enter the 4-step purification process represented by acid treatment, froth floatation, washing, and drying.

Fig. 4B represents the process flow diagram of the conventional technology where a mixture of H_2 and Natural gas enters the thermal decomposition reaction chamber. Due to the thermal decomposition of methane, MWCNTs, amorphous carbon, the unused mixture of H_2 and Natural gas, and spent Ni-Mo/MgO catalyst products are produced, following which, the unused H_2 and natural gas mixture is recycled back to the reaction chamber. The MWCNTs, amorphous carbon, and spent Ni-Mo/MgO catalyst enter the 5-step purification process represented by oxidation, acid treatment, froth floatation, washing, and drying.

2.4.2. Life cycle inventory

A key issue in ex-ante LCA studies is the scale-up of an emerging technology pathway to a full-scale model using available lab knowledge and aligning it with established reference technology systems [82]. Typically, learning curves based on market experience are not directly applicable to these new systems, given the absence of peer-reviewed studies on market penetration [82]. In this study, despite expert consultancy, upscaling methods recommended by Parvatker and Eckelman [88] and Tsoy et al. [89] were used to establish inventory data for both the novel and conventional technology systems. Parameter-based upscaling was employed to align production processes with the functional unit of 100 tons of MWCNTs per year, adjusting material, energy, and operational parameters for industrial settings. Manual calculations, as recommended by Lertrojanachusit et al. were also applied in determining the surfactant amount required for the froth flotation process [49]. Traversing the data/time requirements, accuracy hierarchy and the decision tree methods, three types of upscaling methods were applied depending on the foreground process being modeled. In addition, recycling processes were integrated across purification steps to improve resource efficiency and reduce waste. For modeling the electrodes and the catalyst in the novel and reference technology systems, suitable proxies from the ecoinvent database were considered. Moreover, manual calculations based on stoichiometric relations were employed for other foreground processes under consideration, which was sufficient given the relatively straightforward nature of these calculations and the available data. The manual calculations and modellings were then examined and verified by expert consultancy.

The modeling of the selected novel and reference technology systems comprises several sub-systems. These sub-systems of both the technology pathways are divided into three parts – production of the MWCNTs, purification of the MWCNTs to achieve a 90% purity yield, and electricity consumption of sub-systems, adhering to the chosen functional

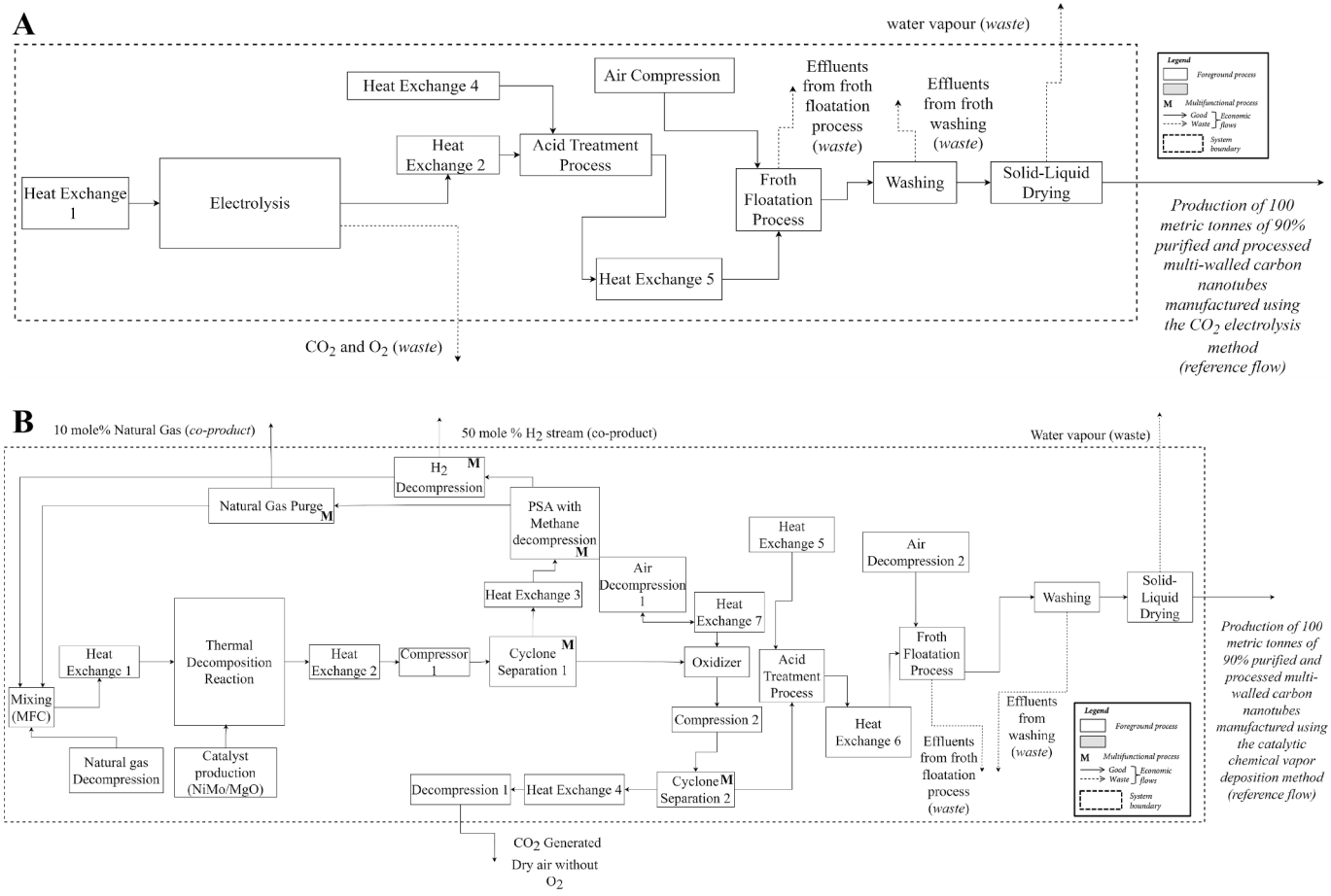

Fig. 4. (A) Process flow diagram of the novel technology system (with foreground processes) and (B) process flow diagram of the conventional technology system (with foreground processes).

unit. The sub-systems for the novel technology pathways sub-systems are detailed in *SM7* while that for the reference technology are in *SM8* and *SM9*, providing justifications for the assumptions taken and data collected wherever necessary.

As mentioned above, LCA models are inherently static and often struggle to effectively represent dynamic systems [90]. Therefore, practitioners frequently utilize process simulation software such as Aspen Plus to mitigate this limitation. In this study, as mentioned above, Aspen Plus V11 was specifically employed to model the heat exchangers within both technology systems at industrial level, as detailed in *SM10*. The rationale for using Aspen Plus to simulate the process engineering conditions of the heat exchangers is twofold. Firstly, other LCA studies of relevant and applicable heat exchangers cannot be retrofitted into the process conditions of both technology pathways, as these pathways are specific to the raw materials considered, operating temperatures and pressures, and the composition of the end product and functional unit (MWCNTs).

Secondly, since water served as a critical input for the operation of the heat exchangers, simulating the heat exchanger in Aspen Plus facilitated the determination of the required cooling water flow rate and consequently, the applicable water volume. This was achieved by converting the electrical energy input to thermal energy, using this heat duty as input for the heat exchanger simulation in Aspen Plus, specifying other process conditions (such as inlet and outlet temperatures, stream compositions, and flow rates), and running the simulation to determine the required cooling water flow rate and, ultimately, calculate the water volume. However, other foreground processes were not simulated using Aspen Plus. This decision was made because efficiency considerations for these processes were not included due to their minimal impact on the overall environmental assessment, thereby placing them outside the scope of this study.

Activity Browser was employed as the LCA calculation software [91], and the Ecoinvent v3.8 database was recruited as the database for background information [92]. Since the production scenario was assumed to be in Belgium, background data for materials and energy were prioritized using Belgian-specific datasets. If data specific to Belgium are unavailable, regional European (RER) or Global (GLO) datasets were used as a substitute.

2.4.3. Life cycle impact assessment and interpretation

Challenges exist in LCIA and Interpretation for novel technologies, including unforeseen impacts in future states, the relevance of characterization factors over time, increased modeling uncertainty, and the potential for unknown impacts [82]. This study focuses on comparing the environmental impacts of the novel technology with a reference system rather than broader future socio-technological contexts. Therefore, these future-oriented challenges fall outside the scope of this study. Instead, to address uncertainties relevant to the present impact assessment and interpretation phases, we used the Global CO_2 Initiative's LCA standards, which provide robust guidelines for evaluating current impacts [93].

To ensure the consistency of the novel and conventional technology systems analyzed in this study, the consistency checklist was applied [94]. The results of the consistency and completeness check are demonstrated in the *SI*-S5 part. Furthermore, contribution analysis was conducted based on the LCA results to identify hotspots responsible for a higher share of impacts, highlighting the results at the process level for each impact category (graphical presentation in *SI*-S6). Sensitivity analysis was performed based on:

a. Change in electricity source
b. Change in materiality requirement
c. Change in CO_2 source

The LCA results were selected as baseline results. In addition, we expanded the sensitivity analysis to include assessments of data quality and key assumptions. Specifically, we introduced changes in the impact assessment methodology and allocation methods to examine their influence on the results. The results of these analysis are provided in *SI-S7*.

2.5. LCA of CCU and influence of carbon capture unit

CO_2 is normally classified as an emission in traditional LCAs; hence, employing CO_2 as an input feedstock can create methodological issues. These difficulties are more visible in CO_2 acquired from point sources rather than direct air capture. The key argument for CCU technology developers and policymakers to push for CCU technologies has been the claims associated with the environmental benefits of CCU products [95]. Nevertheless, the debate for conducting an environmental impact assessment of such technologies is twofold. First, even if the environmental impact of global warming and resource depletion is reduced by implementing a CCU technology, the environmental impact of other impact categories, such as ecotoxicity, acidification, or ionizing radiation, may increase, leading to burden-shifting [96]. Second, CCU solutions may increase CO_2 emissions and resource use because these choices often require a large amount of energy to extract CO_2 and transform it into the final product [96].

Rosental et al. performed a cradle-to-gate LCA on the production of organic compounds such as methanol, ethylene, benzene, toluene, and mixed xylenes. According to their findings, replacing all plants in Germany for fabrication of the researched items with CCU processes would result in a 2–7% increase in total primary energy demand for the entire country [97]. Meunier et al. conducted an environmental impact assessment of producing methanol from CO_2 feedstock and found that regeneration of Monoethanolamine (MEA), which was used to capture CO_2 from point sources, was significant in most impact categories [98]. Additionally, operational challenges associated with MEA could lead to additional environmental burdens. Therefore, the energy required for upstream and downstream activities, such as CO_2 capture and regeneration of used solvents and catalysts, may lead to high energy demands.

Although the comparative ex-ante LCA for the novel and reference systems was designed with a cradle-to-gate boundary, it became apparent that the background activity labeled "market for carbon dioxide, in chemical industry" used for modeling the novel technology did not accurately reflect a carbon capture process. Specifically, the environmental impact assessment of this background activity showed no characterization results, as CO_2 was treated as a by-product in ammonia production, resulting in a zero allocation factor in the Ecoinvent v3.8 database. This omission highlights the need to explicitly model the CO_2 capture process through the carbon capture unit rather than relying on generalized background market data [99]. Moreover, different flue-gas-specific carbon capture technologies can influence the characterization results for modeling the technology systems under study [99,100]. De Kleijne et al. highlighted that modeling of CO_2 utilization technologies for ex-ante LCA studies is influenced by the modeling choices made for the inclusion of the carbon capture technology pathway, the source of CO_2, the source of energy employed, and the lifetime consideration of the product (cradle-to-grave as a system boundary) [64]. In the context of this study, characterization results presented for LCA of post-combustion carbon capture technologies study conducted by Zhang et al. were used to represent the post-combustion carbon capture unit in the novel technology system [101]. Therefore, the system boundary in this section was expanded to include a power-based plant carbon capture process described in Zhang et al.'s study [101]. A brief overview of the MEA-solvent and gas separation membrane carbon capture systems is described in *SI*-S4.

3. Results and discussion

3.1. LCA results

The characterization results were compared per impact category, as

shown in Fig. 5. From the figure, the novel technology system has the most significant impact on AC (acidification), FETP (freshwater ecotoxicity), METP (marine ecotoxicity), TETP (terrestrial ecotoxicity), HTP (human toxicity), MMR (material resources, metals/minerals), and POFP (photochemical oxidant formation) when compared to the conventional reference system. Therefore, the conventional technology is 43.5% (AC), 46% (FETP), 88.8% (METP), 1.8% (TETP), 52.2% (HTP), 87.9% (MMR), and 14.1% (POFP) lower in impacts when compared to the novel technology system. Although the systems were modeled similarly, the contribution of material inputs in the electrolysis process profoundly affects the impact categories mentioned above. Since the novel technology system uses metal anode and cathode to conduct electrolysis, it has the highest impact in the material resources category compared to the conventional technology system. This could be due to the upstream impacts of mining and cathode and anode metal production. The detailed LCA results are shown in Table S2.

High values observed in the FETP and METP might be contributed using electrolytes in the novel technology system. The values of both systems in EP (eutrophication), TETP, and POFP are close. With respect to EP, both systems have a large amount of effluents entering the wastewater treatment plants, which contain a large number of macronutrients. As a result, both systems exhibit commensurate values. Also, the gases and volatile organic compounds that are a part of the background processes in both systems could contribute to comparable values in the POFP impact category. Comparable values in the TETP category could be associated with the effluents and background processes which may impact terrestrial ecosystems.

Unsurprisingly, the conventional technology system is higher in the CC (climate change) (41.9%) and the ODP (ozone depletion) (67.2%) impact category because natural gas is a crucial raw material in producing MWCNTs using the CVD method. Additionally, the conventional technology system consumes a large amount of electricity as amorphous carbon formed in MWCNTs production must be eliminated (also see *SM2* and *SM9* for the sheets for electricity). On the other hand, amorphous carbon as a byproduct of MWCNTs production is absent in the novel technology system. Moreover, hydrogen and natural gas recycling was modeled in the conventional technology system. The gas recycling system requires a high aggregate of electricity input, which is not a specification in the novel technology system. The contribution analysis further explains the reasoning behind the range of characterization values observed across the impact categories. To ensure the consistency of both systems analyzed in this study, the consistency checklist was used [94], as shown in Table S3.

3.2. Contribution analysis

The top seven process contributions are illustrated for each impact category. In the AC category, for both systems, copper anode, copper cathode, and electricity contributed to the majority of emissions. The impact of the copper cathode and anode in the novel system could be due to the direct consumption of brass electrodes in the electrolysis reaction, while the conventional system required higher electricity input for purification and gas recycling processes. In the CC category, high and medium-voltage electricity are major contributors to emissions, as both systems require substantial electricity to produce and purify MWCNTs. Additionally, the electrolysis process in the novel system released CO_2 and O_2 as waste emissions, contributing to this category. In the ecotoxicity impact category, processes affecting freshwater, marine, and terrestrial ecosystems were identified.

For the FETP impact, dehusked coconut, used in producing dodecanol from coconut oil for surfactants, had a significant impact. Barium carbonate production also affected freshwater ecosystems due to its persistence in the environment. For METP, barium carbonate was again a major contributor in the novel system, while high-voltage electricity was significant in the conventional system due to extensive purification steps. For TETP, dehusked coconut had the largest impact, mainly due to deforestation for coconut oil production. In the NRER ((non-renewable energy resources) category, electricity from natural gas and hard coal was the largest contributor, particularly in the conventional systems that required more energy for purification. Ethylene, used to produce ethylene oxide for non-ionic surfactants, also contributed significantly. EP impacts were mainly due to wastewater and non-ionic surfactants. Sludge from wastewater treatment contributed to EP when disposed of in landfills or used as fertilizer. Dehusked coconut, part of the non-ionic

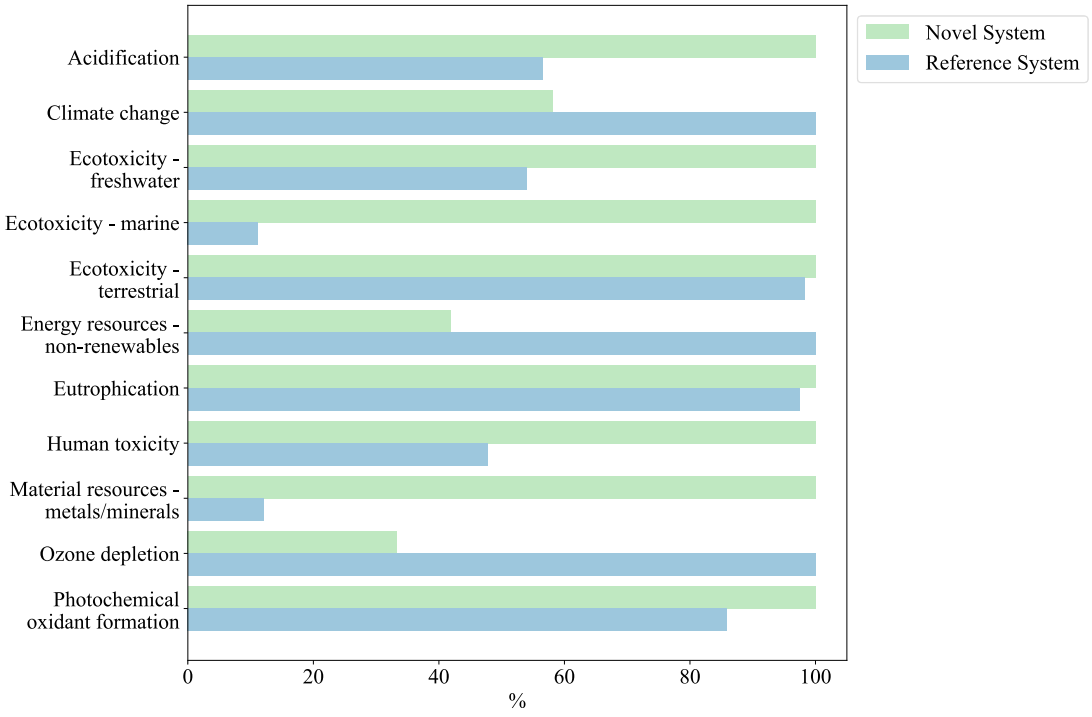

Fig. 5. Characterization results compared to highest value per impact category.

surfactant market, also led to EP due to land use. HTP impacts were driven by non-ionic surfactants and materials such as barium carbonate and copper anode in the novel system, which are considered toxic at high concentrations [123].

MMR impacts were primarily due to copper concentrate used in air compression devices and nickel concentrate for catalyst preparation in the conventional pathway. ODP impacts were largely caused by natural gas use and transportation, essential in both systems for MWCNT production and electricity generation in Belgium. POFP was influenced by the production of dodecanol and the use of copper anode and cathode, with precursor pollutants emitted during brass production and refining processes. For detailed illustrations of the process flows contributing to each impact category, refer to Fig. S3 to S6.

3.3. Sensitivity analysis

3.3.1. Change in electricity source

According to the characterization results, producing electricity to meet the energy demands of novel and conventional technology systems was a significant contributor in most impact categories. Any changes in the effects of the electricity-generating system would thus have a significant impact on the overall environmental profile of MWCNTs produced using either the novel or conventional technology pathway. As a result, in this sensitivity analysis, a change in the source of electricity was used. Although the electricity mix of Belgium is dominated by nuclear energy [102], the background market activity for Belgium's electricity mix in the Ecoinvent v3.8 database assumes an energy mix dominated by natural gas and imports from countries such as Great Britain, France, Netherlands, and Luxembourg [92]. Therefore, earlier assessments used power based on Belgium's energy mix, which operates on a majority share of fossil fuel sources such as natural gas and coal. This sensitivity analysis employed electricity produced from an onshore wind turbine. The changes in each impact category are presented in Fig. 6.

The shift from a fossil-based to a renewable-based electricity source in both systems results in a significant decrease in impacts, as observed in Fig. 6. High percentage reductions are noticed in CC, NRER, and ODP impact categories. This shows the significant impact of electricity production in terms of its emissions in these categories. Reasonable reductions can be noted in AC, METP, EP, and POFP, as the upstream processing of fossil-based electricity carries a significant environmental load. Although negligible differences are noticeable in FETP, TETP, and HTP, there is an increase in impacts in the MMR category. This could be related to increased material resources needed to construct infrastructures such as wind turbines and the high-voltage transmission networks.

3.3.2. Change in materiality requirement

The reason for changing the materiality requirement in the froth floatation process is two-fold. The environmental impacts of the background market activity of the non-ionic surfactant are notable in impact categories such as FETP, TETP, NRER, EP, and HTP. Additionally, in retrospect, when the froth floatation process recommended by Lertrojanachusit et al. was upscaled to suit that of the novel and conventional system [49], the ratio of the amount of surfactant to that of the MWCNTs of 90% purity yield was observed to be 0.92:1. This ratio is considered high when compared to that of industrial processes, where the amount of surfactants applied for the froth floatation process is supplied at grams per ton of solids [103]. Therefore, the calculated scaled-up value of the surfactant amount prescribed by Lertrojanachusit et al. is inconsistent relative to industry standards where the froth floatation processes were used [49]. Therefore, a 75% reduction in the surfactant amount was applied to understand the sensitiveness of the environmental impacts of the novel and conventional technology systems. However, the amount of deionized water required for the froth floatation steps of the novel and conventional technology systems remained the same. The characterization results of the 75% reduction in surfactant amount and the changes in each impact category are presented in Fig. 7.

The highest decrease in impact is observed in FETP, TETP, HTP, and POFP. This is due to the decreased precursor components required to manufacture non-ionic surfactants – dodecanol and ethylene oxide. Additionally, the 75% reduction in the surfactant amount in both the novel and conventional systems led to a decrease in TETP due to the reduced land needed for coconut oil production, a key precursor for dodecanol. In the novel system, barium carbonate, used as an

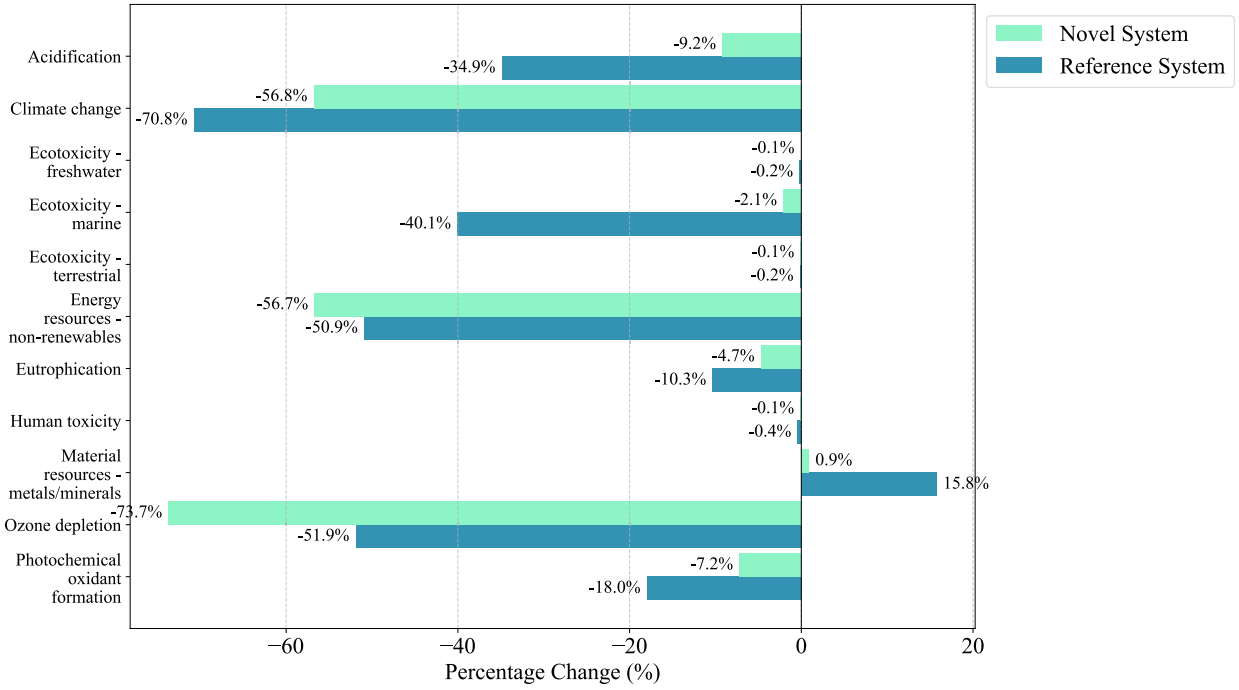

Fig. 6. Percentage change compared to baseline characterization observed for the change in electricity source.

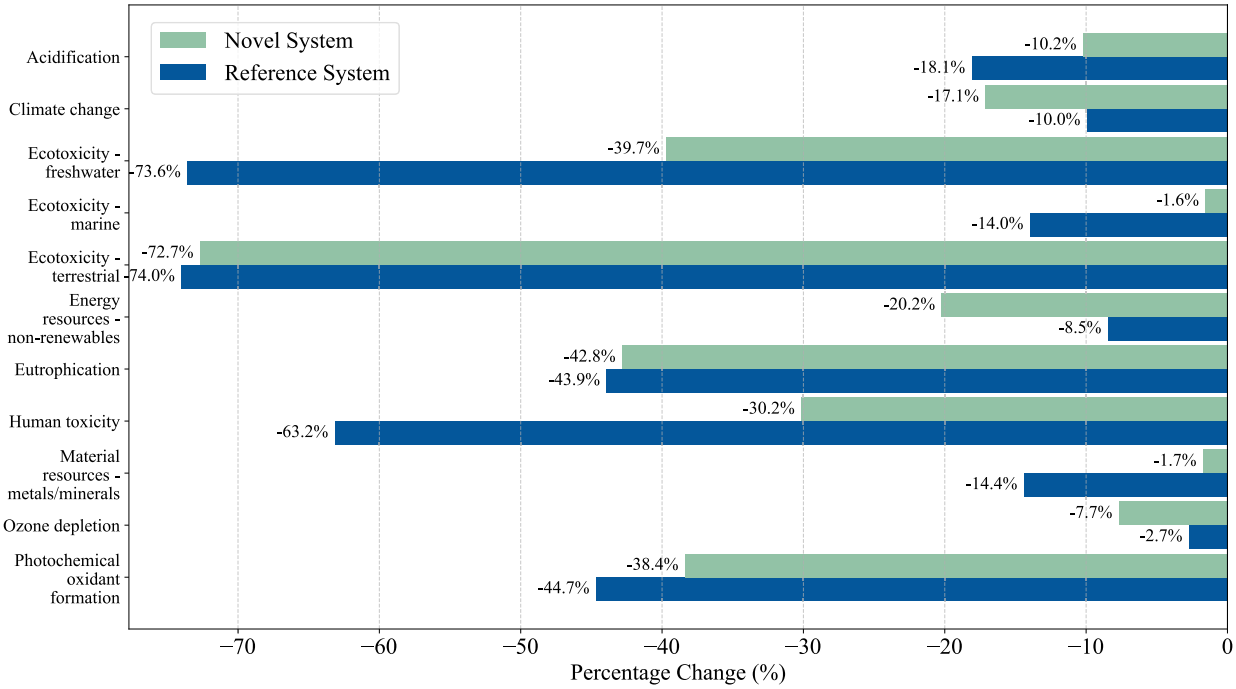

Fig. 7. Percentage change compared to baseline characterization observed for the change in surfactant amount.

electrolyte, notably impacted FETP due to its low solubility in water, maintaining a significant impact, marked by the 39.72% decrease shown in Fig. 7. The reduction in surfactant amount also led to a decrease in ethylene production, resulting in a 10.23% and 18.11% decrease in the AC category. This also explains the percentage decreases in CC, NRER, HTP, ODP, and POFP impact categories.

3.3.3. *Change in the source of CO_2 input feedstock*

The source of CO_2 as an input feedstock influenced the environmental consequences of CO_2 utilization technologies [64]. In the baseline scenario, the CO_2 input feedstock (market for carbon dioxide background activity) was sourced from ammonia production through a steam methane reformation process. Steam methane reformation is the most common, efficient, and cost-effective technology for producing ammonia [104]. However, because it employs fossil fuel as an input feedstock, it negatively influences the environment. According to Singh et al., the global warming potential of ammonia production via steam methane reformation is the second greatest (3.03 kg CO_2-eq per kg of ammonia generated) when compared to coal gasification (3.85 kg CO_2-eq per kg of ammonia produced) [104].

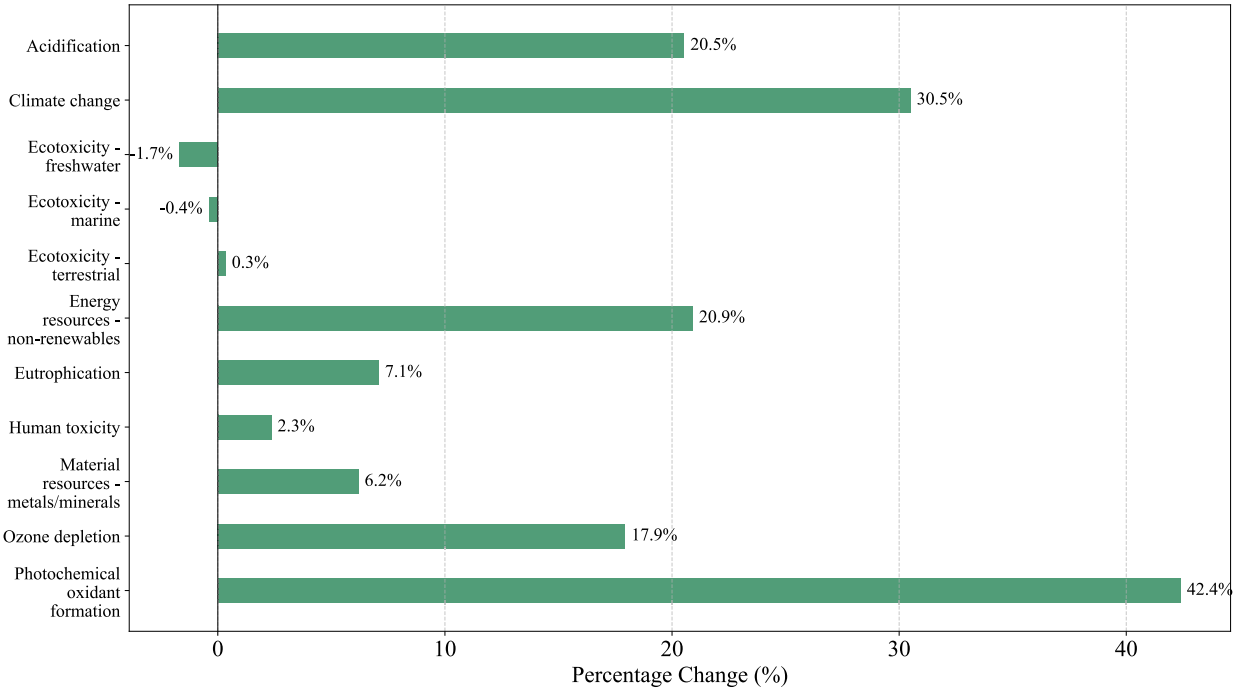

Fig. 8. Percentage change compared to baseline characterization observed for the change in CO_2 source.

The sensitivity study was performed to investigate the shift in environmental impacts caused by a "green" CO_2 source. In the context of this study, a "green" CO_2 source suggests the use of a renewable source of CO_2 production. Therefore, biogas, a renewable CO_2 source with a high content of CO_2, was chosen to represent the "green" CO_2 input feedstock. The changes in each impact category are presented in Fig. 8.

Changes in ecotoxicity-based impact categories are minimal as the use of surfactants remains the primary contributor to impacts on FETP, METP, and TETP. Conversely, the highest increase in impact (42%) in the POFP category, demonstrated in Fig. 8, was due to a combination of upstream activities related to biogas production and the higher electricity required for the flameless combustion of the biogas process. This also led to increases in CC (30%), AC (21%), and EP (7%), driven by the upstream activities of biogas production involving the anaerobic digestion of manure, used vegetable oil, bio waste, and sewage sludge. The increase in electricity consumption compared to the baseline is approximately 4.6 times. Consequently, the additional upstream activities required to meet this increased electricity demand resulted in higher impacts across several categories, including NRER (21%), HTP (2%), MMR (6%), and ODP (18%).

However, the Natural gas composition available in high-pressure (high calorific) industrial gas networks of Belgium is primarily composed of Methane [105]. Methane was the primary molecule undergoing thermal dehydrogenation in the conventional technology pathway, this was followed by the 5-step purification process – oxidation, acid treatment, froth floatation, washing, and drying. The designed 5-step purification process under consideration in this study was primarily selected to remove impurities and byproducts of the chemical vapor deposition process that uses natural gas from industrial gas networks in Belgium. As a result, a change in the composition of methane-rich-gas necessitates changing the operating conditions of the purification process or the selection of alternative purification processes altogether. To evaluate the sensitivity of the model in this ex-ante LCA study, biogas and biomethane were initially considered as alternatives to natural gas. Therefore, in this part, the sensitivity analysis was not carried out for the conventional technology pathway regarding change in the source of CO_2 input feedstock.

3.4. Carbon capture unit

The characterization result of the CC impact category of MEA-based solvent CO_2 capture technology from Zhang et al. was used to update the characterization results of this study [101]. Table 3 shows the CO_2 capture and emissions from an MEA-based capture unit linked with the MSCC-ET system. The CO_2 capture rate by MEA was calculated as 0.93 kg CO_2-eq/kWh based on the annual carbon capture capacity of the MEA associated with a power plant. However, considering the emissions from the MEA itself (as shown in Table 3), it can be determined that for every 1 kg CO_2 captured, 0.0131 kg CO_2 is emitted. This coincides with Von Der Assen et al. highlighting that a CO_2 capture unit will not capture (–) 1 kg CO_2-eq for every kg CO_2-eq emitted [106]. Given the reaction required 726.5 ton of CO_2 sourced directly from the MEA unit, a total of 716.6 ton of CO_2 emission mitigation can be realized. Therefore, the combined net CO_2-eq emissions for the MEA and MSCC-ET system were reduced to 1073 ton CO_2-eq, representing a 40% reduction. This outcome highlights the advantages of integrating MEA capture with advanced processes to reduce CO_2 emissions and mitigate the CC impacts. However, this trend does not extend to other impact categories such as FETP, TETP, HTP, ODP, and EP [99]. This is associated with the increased consumption of electricity to operate the capture units. The MEA-based CO_2 capture unit shows a decrease in PMFP and AC impact categories due to the co-capture of SO_2, NO_x, and particulate matter with the solvent absorption method [101]. A similar calculation was performed for a two-stage membrane capture unit, and the details can be found in *SM2*.

Table 3
Calculations of CO_2-eq captured by MEA-based capture unit based on data obtained from Zhang et al. [101].

Parameter	Value	Unit	Note
Annual CO_2 capture from a power plant	$3.9*10^9$	kg CO_2	Total CO_2 captured based on a coal-based electricity generation system
Annual operational hours	7500	hours	Operational hours of the coal-based electricity generation system
Annual total net electricity output	$4.19*10^9$	kWh	For installing an MEA-based CO_2 capture unit
Annual MEA emission	$5.12*10^7$	kg CO_2-eq	MEA annual electricity consumption 66 MWe; MEA emission factor 103.5 kg/MWh.
CO_2 captured by MEA	0.93	(captured) kg CO_2-eq/kWh	
CO_2 emissions of MEA	0.0122	(emitted) kg CO_2-eq/kWh	Based on the emissions from the MEA CCU itself
CO_2 capture-emission relationships	0.0131	kg CO_2-eq	For every 1 kg CO_2 captured, 0.0131 kg CO_2 emitted
Total CO_2 emission mitigation due to MEA regarding CO_2 sources	−716,625.7	kg CO_2-eq	726,165 kg CO_2 is required to be captured for the novel system

4. Limitations and prospects

In this study, sensitivity analysis was performed to determine the impact changes from a lower surfactant value in the froth floatation step. However, even with the reduced values of surfactant associated with sensitivity analysis, the ratio of surfactant to MWCNTs is 0.23:1, which is higher than the surfactant amount consumed in industrial processes, particularly in mineral processing industries, where the froth floatation process is widely employed.

Curran and Langhorst et al. recommended cross-checking the upscaled datasets with values reported in the literature [86,107]. However, no peer-reviewed ex-ante LCA study was found due to the unique experimental nature of the novel technology system. Moreover, for the reference technology system, LCA studies mostly employ a black box approach, where all the material and energy inputs flow to a small set of foreground activities that do not distinguish thoroughly between different fabrication stages [108,109]. Therefore, before considering the results in other contexts, having an expert analyze the unit process data of all important foreground processes would be preferable.

Guineé et al. highlighted that LCA models are typically linear steady-state models of physical flows. This, by definition, sometimes oversimplifies inherently dynamic processes [94]. An example of the dynamic nature of a process in this study is the recycling of gas streams containing H_2 and Natural gas, leaving the thermal decomposition reaction in the conventional technology system. Moreover, the inclusion of recycling processes lowers environmental burdens experienced. Other recycling processes were not modeled due to the low production threshold taken as a functional unit. For a fabrication process producing 100 tons of 90% yield of MWCNT as a product, the recycling of materials in the purification steps was not considered economically viable and, therefore, sent to the wastewater treatment background activity.

Based on the experiences and findings of this study, we summarize some practical insights and other relevant considerations on nanomaterials in applying ex-ante LCA. These insights could be applied to the industrial-scale production of other advanced carbon nanomaterials such as graphene and carbon fibers, which face similar challenges regarding scalability, resource management, and environmental impact minimization. The process begins by identifying and selecting suitable technologies for comparison, evaluating factors such as TRLs, carbon reduction potential (e.g., expected amount of CO_2 sequestered),

resource efficiency (e.g., raw material sourcing; water usage; electricity requirement, etc.), and basic technical requirements (e.g., CO_2 purity requirement; expected CNT purity; catalyst type; reactor type; operation conditions; compatibility with current industrial processes; feedstock availability, etc.). Other considerations could also involve temporal and geographical scrutiny (e.g., local energy mix; policy context, etc.), aiming for seamless integration into current workflows.

After selecting suitable technologies, an ex-ante LCA is conducted to evaluate the potential environmental impacts at an industrial scale. This involves adapting the classic stages of LCA, as outlined by ISO 14040/44, with appropriate assumptions to address the specific challenges of scaling up technologies in the carbon utilization field. For LCI, multiple methods can be employed such as direct plant data collection, stoichiometry-based mass balance calculations, expert consultations, and engineering process simulations, all designed to accurately reflect the complexity of industrial-level operations [88,89]. During impact assessment and interpretation phase, particular attention can be given to exploring scenario-based CO_2 sourcing (e.g., evaluating the influence of different CO_2 sourcing (i.e., from steel production versus cement factories) on the impacts, carbon intensity variation, geographic source dependency, etc.), and adjusting for multifunctionality (e.g., including multifunctionality in electrolysis to reassess co-products as economic flows rather than emissions, co-product allocation in purification, energy credit for recovered heat, etc.). For instance, in carbon fiber manufacturing, where a by-product like hydrogen could be integrated into the energy balance of the production process, thereby contributing to overall efficiency. These tailored checks can help adapt the LCA to fit the needs of carbon nanomaterial technologies transitioning from the lab to industrial scale.

Finally, based on the LCA findings and combined with engineering insights, actionable recommendations can be made for further refining the technology. This involves rethinking the process design and upgrading equipment to better meet industrial requirements, possibly by modifying or integrating new components into existing systems. In this study, for example, we incorporated carbon capture units to enhance sustainability. Moreover, in CCVD, the integration of gas recycling processes for purification plays a critical role in enhancing sustainability by reducing material demand and improving cost-effectiveness. However, challenges such as electrode degradation, CO_2 supply stability, and effective impurity management persist, especially as the technology scales up and in long-term applications. In addition, previous studies on dynamic scouring in membrane bioreactors have shown the effective reduction of fouling and optimization of system performance [110,111]. Inspired by these findings, the integration of these dynamic model strategies could also provide visions for optimizing and enhancing resource efficiency and minimizing fouling-related issues in catalytic, carbon capture unit, or electrochemical conversion reactors. In the next section, we demonstrate specific recommendations derived from this study.

5. Conclusion and recommendations

The primary motivation for conducting this research study is to extend ex-ante LCA methodology application to chemical engineering practices in the CO_2 utilization technology landscape. The main objective of this study included determining the environmental impacts of the fabrication of MWCNTs by transforming CO_2 as an input feedstock compared to a conventional CNT fabrication process on an industrial scale. To answer this question sufficiently, the study used a parameter-based selection technique to select the preferred novel and conventional technological systems. The environmental impacts of both novel and conventional technology systems were assessed using the ex-ante LCA methodology, focusing on MSCC-ET and CCVD, respectively. We scaled up the chemical processes to fit a functional unit of 100 tons of MWCNTs with a 90% purity yield, using a cradle-to-gate approach. We applied combined methods – expert input, manual calculations, process modeling, recycling integration, and careful parameter scaling – to ensure that the industrial-level model was realistic and based on practical experience. We also conducted multiple sensitivity analyses to test the robustness of the results, ensuring consistency between foreground and background activities using a recent LCI database. Based on the LCA results, recommendations to early-stage technology developers and LCA practitioners are formulated based on the ex-ante LCA performed and the results obtained.

5.1. Relevance of material demand for novel technology system

The contribution analysis reveals the importance of the material demand, particularly in electricity production, electrolysis, and the purification activities of the novel system. Although it is pointed out that the impacts associated with the use of non-renewable energy resources of the novel technology system are 58.1% lower than that of the conventional technology system, this can be attributed to a higher set of activities modeled in the latter compared to the former. Nonetheless, activities such as electrolysis and purification treatments, barium carbonate, brass, iron-nickel–chromium alloys, and surfactants contribute to many impact categories. This is evidenced in the results of the sensitivity analysis, where a 75% decrease in surfactant amount led to an average decrease of 25.7% across all impact categories, with a relatively high decrease in impact categories observed across TETP, FETP, EP, HTP, and POFP compared to the baseline case.

Moreover, the changes in material demand were observed when shifting from a mixture of fossil and renewable-based sources of electricity to a 100% renewable one (onshore wind energy). An average of 19.1% decrease across all impact categories was observed, with a sizeable decrease noted in ODP, CC, and NRER. Predictably, a small increase in impact was noted in the MMR impact category. When the CO_2 source was shifted from CO_2 generated during ammonia production to that of biogas, an average increase of 13.3% was noticed across all impact categories. The highest change in impact is noticeable across POFP, CC, NRER, and AC. This is attributed to a 4.6 times increase in electricity since additional activities such as an air decompression process, heat exchanging unit, and combustion of biogas must be modeled for the supply of CO_2 in the form of biogas.

There were also differences in the number of modeled foreground processes between the novel and conventional systems, particularly in the purification stages. Nevertheless, this initial screening of environmental impacts can hope to provide an early indication of which market processes contribute to select impact categories.

5.2. For early-stage technology developers

Based on the results and discussion, three major recommendations are suggested for early-stage technology developers in CCU or carbon nanomaterials. The first two recommendations are highlighted in the process optimization of material inputs, source of CO_2, and electricity sub-section. The third recommendation is detailed in the product lifetime and CO_2 capture sub-section, as shown in Fig. 9. Technology developers in the CCU or carbon nanomaterials can use the recommendations to solve the environmental hotspots and inefficiencies identified at an early stage.

While the novel technology system has lower impacts in the CC impact category compared to that of the chosen reference system, the impacts of the novel technology system in almost all other impact categories are higher. These high impacts, observed in the other impact categories, such as AC, METP, FETP, TETP, EP, HTP, MMR, and POFP, are primarily caused by the use of material inputs such as brass and iron-nickel–chromium alloys as electrodes, barium carbonate as the electrolyte, surfactant, and fossil fuel-based electricity source.

Early-stage technology developers have to note that while moving from a lab scale to a pilot scale or higher has its share of development challenges, one of the key recommendations is to optimize the use of the

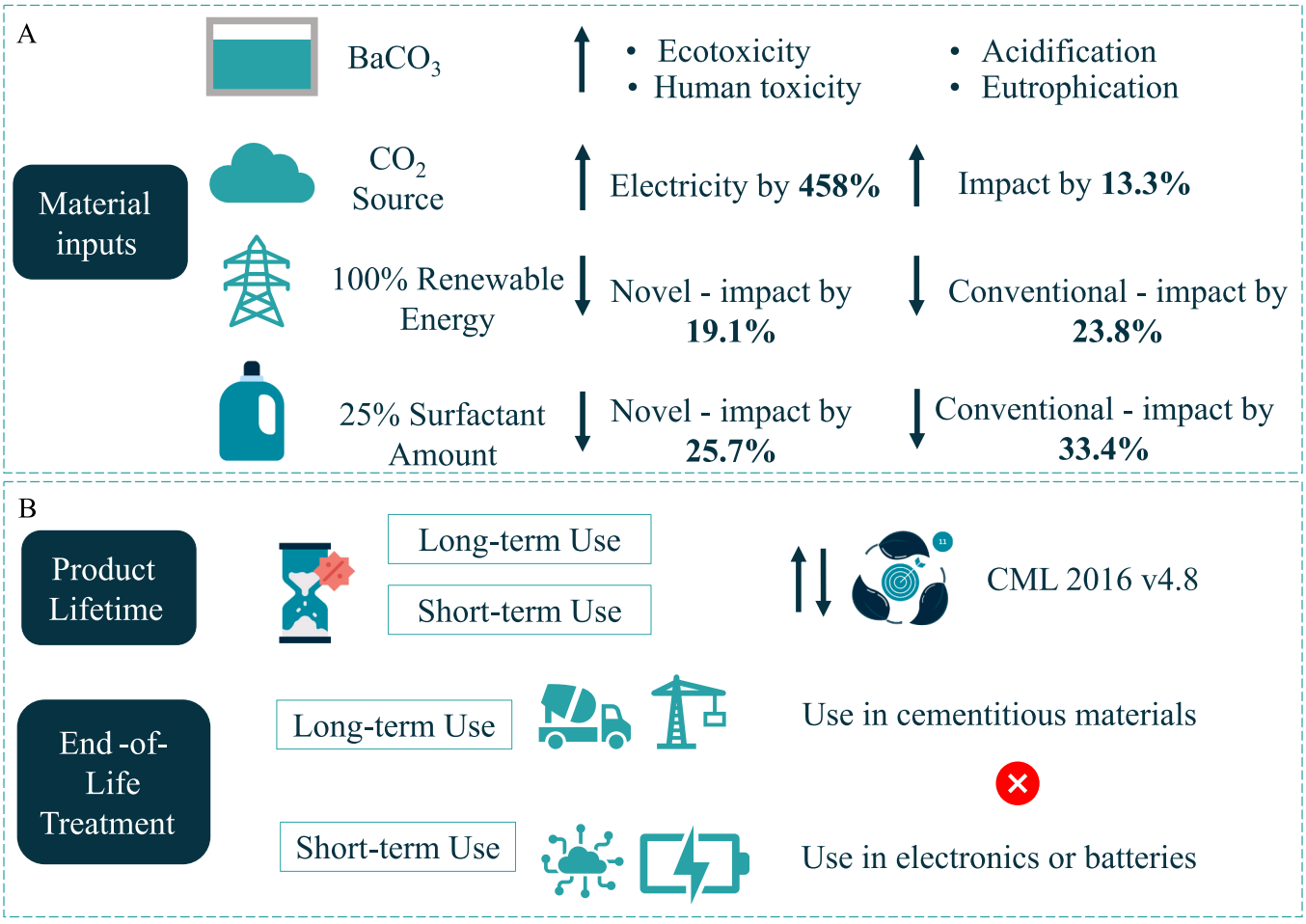

Fig. 9. Recommendations for early-stage technology developers. (A) Material inputs (B) Product lifetime & End-of-life treatment.

above-mentioned material inputs – specifically those as part of the electrolysis and purification steps. For example, in the sensitivity analysis conducted for a decreased use of surfactant and a shift from fossil-based to 100% renewable-based electricity sources, the average percentage decrease experienced in all impact categories is 25.7% and 19.1%, respectively, as indicated in Fig. 9A. While the limitation in the upscaling method used for calculating the surfactant amount in the baseline case is iterated, a cautionary approach is recommended to be followed when moving from a proxy-lab-scale setup to a pilot-scale or industrial-scale operation.

Moreover, careful consideration of the source of CO_2 to be used is also recommended, as shown in Fig. 9A. As part of the sensitivity analysis conducted for operating the novel technology system using biogas as the CO_2 input feedstock instead of CO_2 generated from ammonia production, the electricity requirements increased significantly. Consequently, the overall environmental impacts across all impact categories increased by 13.3%. This is primarily attributed to the increase in foreground processes required to generate a higher purity stream of CO_2 from the biogas source.

The impact of output volume on environmental impacts decreases as the scale of the fabrication plant increases [109]. While this is mostly due to the recycling of feedstock that becomes economically viable at a certain minimum production threshold [109], the environmental impacts of primary material used cannot be overlooked because these effects are experienced at the local level.

While the chosen cradle-to-gate system boundary serves as a convenience to commodity producers, who do not know in which way the product will be used after-sale [106], for a proper accounting of CO_2 to be performed, the product's lifetime must be considered (Fig. 9B). This is primarily attributed to the temporary storage of CO_2. In short-term use case scenarios, the used carbon dioxide is released back into the atmosphere in a short period, thereby leading to higher temporal impacts compared to long-term use case scenarios [64]. Additionally, CO_2 capture unit also influences carbon accounting techniques [101]. We advocate for using a less volatile and more stable solvent or non-solvent capture technology to lower the environmental impacts by decreasing the energy consumption of the CO_2 capture process itself.

5.3. *For LCA practitioners*

The recommendations identified for LCA practitioners based on the research approach in this paper are divided into two distinct parts. The first part considers recommendations focused on modeling requirements, as illustrated in Fig. 10A, whereas the second part highlights recommendations deemed vital for applying ex-ante LCA methodology to CO_2 utilization technologies, shown in Fig. 10B.

5.3.1. *Model-based recommendations*

1. **Implications of changing the purification treatment processes** – The modeled 4-step and 5-step purification treatment steps of the novel and conventional technology systems, respectively, are known to cause impacts across impact categories such as FETP, METP, TETP, HTP, AC and EP. As a result, it would be recommended to apply other appropriate treatment processes that would not compromise on the comparability and functionality of the end-products for both systems. Therefore, this would also support potential future process designs of novel and conventional technology systems, perhaps

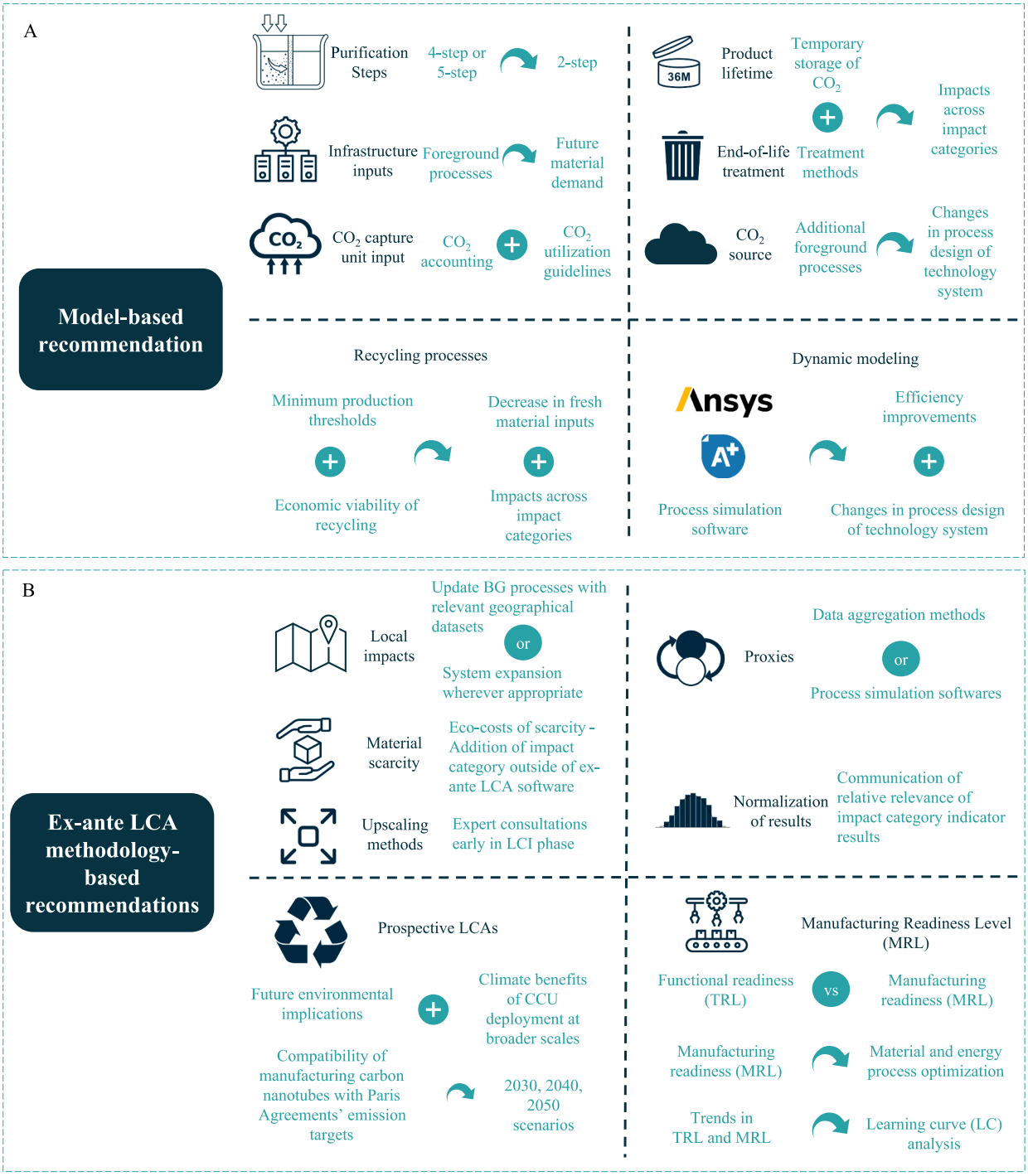

Fig. 10. Recommendations for LCA practitioners (A) Model-based recommendations (B) Ex-ante LCA methodology-based recommendations.

leading to decreased impacts observed in the abovementioned categories.

2. **Inclusion of infrastructure inputs** — Across both systems, the impacts experienced in MMR impact category are infrastructure inputs part of the background activities present in the Ecoinvent v3.8 database. However, the infrastructure demand of the foreground processes was excluded as part of the goal and scope phase, thereby excluding possible inferences that could be derived regarding the material demand of both systems. The inclusion of infrastructure inputs for the foreground processes would help in understanding future material demand, which is specifically relevant to the material scarcity impact category if introduced in the LCIA phase.
3. **Modelling of CO_2 capture unit** — CO_2 capture units should be included to appropriately meet the system boundary specification of cradle-to-gate. In hindsight, substituting the CO_2 capture unit using a market activity from the Ecoinvent database could lead to inconsistent results and, therefore, should serve as a learning opportunity for LCA practitioners.
4. **Modelling lifetime of product and end-of-life treatment** — The temporary storage of CO_2 in long and short-term use cases could lead to higher or lower impacts across various impact categories as mentioned above. Additionally, modeling end-of-life treatment methods for the MWCNTs could provide useful discussion concerning practical treatment methods available. Moreover, modeling the

product's useful lifetime and end-of-life treatment options provides valuable insight and inferences in estimating the overall net emissions across all impact categories.

5. **Source of CO_2** — A change in the source of CO_2 employed in the novel technology system has significant impacts due to a notable increase in energy demand, material load, and infrastructure inputs, as additional processes have to be modeled, thereby determining the process design requirements of the system. This is evident in the sensitivity analysis performed with biogas as a renewable source of CO_2 in this study.
6. **Inclusion of recycling processes** — As the production capacity of both systems increases, and minimum production thresholds are met, this increases the economic viability of employing recycling processes, specifically across the purification steps. While this does increase the process design requirements of the systems at large, the inclusion of recycling processes may also lower the impacts across impact categories such as MMR, AC, EP, HTP and ecotoxicity due to a decrease in fresh material inputs. Therefore, useful discussions regarding the environmental impacts of recycling processes can be accommodated.
7. **Dynamic modeling using process simulation software** — While LCA models are not equipped to model dynamic systems [90], process simulation software such as Aspen Plus can provide useful inferences on the effects of efficiencies and efficiency improvements on the process design of the overall system under study.

5.3.2. *Ex-ante LCA methodology-based recommendations*

1. **Updating of geographical scope to include local impacts** — In order to allow for a discussion regarding the location-specific benefits of fabricating CNTs using both systems, further attention has to be provided to the choice of background activities taken into account across common foreground processes such as tap water, deionized water, sulfuric acid, non-ionic surfactant, wastewater market activities, and the steam market activity. Moreover, specific background inputs to the electrolysis process in the novel technology, such as market activities for electrodes and electrolytes, market for hydrogen gas, and catalysts required for the thermal decomposition reaction in the reference system, are either within the region of Europe or considered global. These market activities are taken from the region of Europe or considered global, so location-specific benefits are challenging to infer. If the values for this study are used to extrapolate conclusions to other favorable locations, careful consideration must be taken. A solution is to find specific geographical values and, therefore, provide location-specific context for further discussions. Another solution, although fraught with challenges during the LCI phase, would be to mindfully expand system boundaries to model certain background processes with geographical context-specific datasets taken from manufacturers or peer-reviewed literature sources.
2. **Integration of material scarcity as an impact category** — Instead of the traditional approach of scarcity being defined in terms of absolute depletion, including short-term supply risks outside of the LCA model is beneficial to derive material scarcity impacts. Although the short-term supply risks methodology is currently being investigated, and details of which are much beyond the scope of this study, Vogtländer et al. proposed a novel approach to the problem of short-term material supply shortages based on subsequent sharp price increases that can cause socioeconomic instability [112]. The underlying principle is that each resource has a unique supply chain and price volatility. The eco-costs of scarcity are calculated using the value at risk (VAR), a well-known statistical risk indicator in the finance industry. Additionally, Vogtländer et al. included a list of 42 metal indicators [112]. The values obtained using this method are related to business risks and are relatively easy to understand; however, the researchers recognized that statistics from the past may not be replicated in the future when certain metals experience oversupply to overdemand trends [112]. The method presented by Vogtländer et al. is arguably the simplest to incorporate into future ex-ante LCA studies, particularly for CO_2 utilization approaches that require crucial raw materials [112].
3. **Changes to upscaling methods and unit process data collection** — In order for a process LCA model to provide a good representation of reality, the data sets used should accurately mirror industrial scale levels, especially where economies of scale are an essential parameter in the study's context. It is recommended that foreground processes receive expert consultations early in the process design investigation so that a reiterative approach recommended by Guineé et al. can be used to detect inconsistencies, uncertainty, and variability [94]. Consequently, results obtained from such an exercise would provide a more accurate reflection of industrial processes.
4. **Reducing the use of proxies through system expansion** — Using proxy data sets is the quickest and easiest way to bridge data gaps, but it also has the most uncertainty [113]. On the other hand, data extrapolation approaches may require substantial specialist knowledge and hence be more difficult to apply but may provide more robust results in bridging data gaps. While it is beyond the scope of this study to conduct a full investigation of practical data aggregation methods to solve for the use of proxies in ex-ante LCA studies of CO_2 utilization technologies, data extrapolation approaches for use in LCA studies of bio-based products and services are important [113]. Combined with process simulation software like Aspen Plus, these methodologies can be effective for ex-ante LCA assessments of CO_2 utilization technologies since they consider production characteristics, resulting in a more accurate representation of industrial-scale operations.
5. **Use of normalized results** — In life cycle assessment (LCA) research, a normalization process is commonly used to comprehend impact category data's relative significance [114]. Normalization results may be riddled with errors owing to uncertainty concerning the datasets utilized in LCIA, geographical inconsistencies, and temporal conflicts connected with various sub-systems [114]. Guineé et al., however, proposed normalizing the impact profile because the primary objective of such a method is to transmit information on the relative relevance of the impact category indicators' results [94]. Establishing result normalization as a requirement for future ex-ante LCA studies would assist LCA practitioners and early-stage technology developers in using the LCA study's results to create process design-focused changes, which could have a far-reaching impact on the impact categories.
6. **Performing forward-looking analysis** — Prospective LCAs (p-LCA) can play crucial roles in early research and development by offering environmental information and assisting with scale-up [115]. Prospective LCA refers to assessments of emerging technologies in the early stages of development when there is still potential to employ environmental guidance for important changes [115,116]. In such circumstances, the system modeled is put in a more distant future utilizing scenario modeling principles to reflect technology's potential future environmental implications. Additionally, the merits of CCUs should be compared to their future counterfactuals rather than the current technology base [117]. This is especially important when it comes to using CCU to reduce GHG emissions from the chemical industry, where the climate change mitigation potential of CCU is determined not by the amount of carbon stored in the chemical nor by the amount of CO_2 used but by what the resulting product substitutes [118,119]. Nonetheless, doing forward-looking analyses helps determine the climate benefits of CCU deployment at broader continental or global scales [118,119]. A full outlook on scenario considerations is outside the scope of this study endeavor; nevertheless, Arvidsson et al. provide guidelines on conducting appropriate prospective LCA [116].

7. **Application of Manufacturing Readiness Levels (MRL)** — In addition, it is recommended to use Manufacturing Readiness Level (MRL) instead of using TRL. TRL indicates functional readiness as opposed to manufacturing readiness [120]. Gavankar et al. highlighted that to assess a given technology's maturity and sub-systems, MRL is considered a better yardstick of measurement than TRL [109]. Müller et al. suggested that using CCU on an industrial scale would result in process optimization for both materials and energy efficiency [121]. Economies of scale and established supply chains could also benefit CCU [119]. Furthermore, the researchers emphasized that when CNT fabrication matures from prototype or small-scale to industrial-scale production, a reduction in fabrication energy intensity of two to three orders of magnitude can be envisaged. Furthermore, with sufficient data, the trend noticed with TRLs and MRLs can be documented and validated statistically using more established approaches, such as learning curve analysis [109].

CRediT authorship contribution statement

Justin Z. Lian: Writing – original draft, Conceptualization. **Varsha Balapa:** Writing – review & editing. **Earl Goetheer:** Writing – review & editing. **Stefano Cucurachi:** Writing – review & editing.

Declaration of competing interest

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

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2024.158007.

Data availability

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

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