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ABSTRACT

Graphene transparent electrode (GTE) has been attracting much attention due to fascinating physical properties. However, the extensive deployment of copper foil within GTE production has imparted substantial environmental burden. This paper is a cradle-to-gate life cycle assessment (LCA) study to investigate the environmental impacts of roll-to-roll produced chemical vapor deposition (CVD) GTE and the environmental potential of recycling copper foil for cleaner production. Four production scenarios are developed to promote the lab-to-fab progress, including lab scenario, industry baseline scenario, industry recycling scenario and microwave plasma chemical CVD scenario. The functional unit is set as 1 m^2 of the GTE production and the life cycle inventories of different scenarios are explored. Results show that the copper foil is a major contributor in baseline scenario in the category of primary energy consumption and global warming. The impacts of GTE production in industry recycling scenario vary from 0.01 to 0.18 of the values in industry baseline scenario. Therefore, copper foil recycling shows environmental potential for GTE production. If all building integrated photovoltaics transition to employing perovskite solar cells with GTE produced in copper recycling scenario, the potential reduction in CO_2 emissions is estimated at 141.2 million kilograms per year. The findings serve as a roadmap for the industry, highlighting key areas where improvements can be made to upscale production while minimizing environmental impact. This paper provides insights into the major environmental contributors in the GTE production, guiding the upscaling routes for cleaner GTE production in the future.

1. Introduction

The discovery of graphene is a ground-breaking progress for the world due to its unique electrical properties and high strength (Novoselov et al., 2004). Graphene provides more possibilities for the development of the material field, especially as graphene transparent electrodes (GTE) of flexible solar cells (Chen et al., 2019). Now, Indium Tin Oxide (ITO) is the most successful front transparent electrodes because of its high transparency and low resistance (Arvidsson et al., 2016). Nevertheless, Indium is a scarce material, which is not enough to produce solar cells at a terawatt level (Kim et al., 2022; Li et al., 2023; Li and Zanelli, 2021). As a result, mass produces solar cells need to find other solutions. Additionally, the cumulative energy requirement of ITO material and production accounts for a big share of solar cell production (Espinosa et al., 2011). Graphene with the same properties is a promising substitute for ITO. Flexible organic solar cells and flexible perovskite solar cells using the GTE exhibit a high power conversion efficiency, reaching 15.2% and 16.8%, respectively (Koo et al., 2020; Yoon et al., 2017). Bae et al. incorporated GTE into a fully functional touch-screen panel device that can withstand high strain (Bae et al., 2010). These research results all show that GTE has a vast potential for replacing ITO in the near future.

Chemical vapor deposition (CVD) on copper foil is the most reliable method for graphene production (Zhang et al., 2020). Due to its prominent controllability, superior uniformity, and satisfactory scalability, CVD can grow graphene with high quality and large area on flexible substrates (Xin and Li, 2018). CVD has two main technology routes, thermal chemical vapor deposition (TCVD) and microwave plasma chemical vapor deposition (MPCVD). With the successful achievement of graphene production by CVD method in the research, requirements of commercial applications with a high-throughput production method at large scale and low cost become the focus. Meanwhile, CVD is also an energy-intensive process, which means that high cumulative energy for graphene production is required (Kushnir and Sanden, 2008).

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It was shown that the direct energy use for the production of graphene foils in the baseline scenario based on theorical calculation is 22 kJ/cm^2, which is lower compared to 44 kJ/cm^2 in the ITO baseline (Arvidsson et al., 2016). However, in the given sensitivity analysis, the direct energy use is 160 kJ/cm^2 in high scenario, which is further higher than that of ITO production in high scenario. Furthermore, Cossutta et al. (2017) presented that the primary energy consumption ranges from 1.7 kJ/cm^2 in continuous fab-scale production to 199 kJ/cm^2 in batch lab-scale production with a different carbon source. In the decarbonized energy production scenario, the global warm potential of graphene production can be decreased from 8.91*10^{-3} kg CO$_2$eq/g to 4.39*10^{-4} kg CO$_2$eq/g in different CVD routes. These results all show that the production route and scale are essential for the control of the environmental performance in the production.

Therefore, substantial effort of researchers is being devoted to understanding how to translate the lab-scale graphene production to cost-effective and energy-effective production that is suitable for commercialization. The roll-to-roll (R2R) method has emerged to achieve mass rapid continuous production with low primary energy requirement. Several laboratory R2R systems for graphene production have been proposed in the academic literature. As for TCVD method, Bae et al. firstly provided a roll-to-roll method being capable of producing predominantly monolayer 30-inch graphene films with sheet resistances of 125 Ω/sq and optical transmittance of 97.4% (Bae et al., 2010). Then, a concentric tube system for R2R TCVD production was presented, which employed a bench-scale prototype machine to fabricate graphene at varying speeds (Polsen et al., 2015). After many improvements being made, a scalable route was proposed for the rapid and continuous production of large-area graphene by a multizone R2R TCVD reactor (Kidambi et al., 2018). As for MWPCVD method, a R2R system was established, which is promising to produce continuous and large area graphene films (Yamada et al., 2012, 2013). In this system, heating graphene by microwave plasma replaces the TCVD heating method, decreasing the required graphene growth temperature from 1000 °C to less than 400 °C. TCVD method and MWPCVD method use the same theory to produce the graphene electrode. But MWPCVD employs microwave plasma to enhance the decomposition of precursor gases, allowing for lower temperatures and shorter processing times compared to TCVD. TCVD typically operates at higher temperatures, usually above 900 °C, to facilitate the decomposition of precursors. MWPCVD operates at lower temperatures, often below 700 °C, due to the efficient energy transfer from microwave plasma, which reduces thermal stress on substrates.

Simultaneously, copper foil, serving as a catalyst, stands as the prevailing substrate used for the synthesis of GTE (Pizzocchero et al., 2022). The extensive deployment of copper foil within GTE manufacturing has engendered notable ecological ramifications, thus imparting substantial environmental burden and financial outlay. This predicament profoundly intersects with the overarching goal of attaining sustainable production practices. To address this exigency, R2R production emerges as a pivotal mechanism, poised to effectuate the large-scale recycling of copper foil within industrial settings (Wang et al., 2011).

Life cycle assessment (LCA) offers a comprehensive method for evaluating the environmental impacts associated with every stage of a product's life cycle (Long et al., 2023). By scrutinizing materials and manufacturing processes involved in production, LCA delineates each contributor's share in various environmental profiles. Widely utilized, LCA has been instrumental in assessing the environmental performance of various solar cell components (Li et al., 2022). In prior LCA studies on PV components, key indicators such as primary energy demand and greenhouse gas (GHG) emissions have been commonly employed to gauge the sustainability of graphene transparent electrode production (Yuan et al., 2023). Arvidsson et al. (2016) evaluated the life cycle resource demands associated with producing a graphene electrode area via CVD and contrasted it with ITO production. Their assessment focused on energy and scarce metals as key resources (Li et al., 2024a,

2024b, 2024c, 2024d). The findings indicate that graphene layers may exhibit lower life cycle energy consumption compared to ITO layers, with a notable 3–10 times reduction observed in the best-case scenario. Concerning the utilization of scarce metals, the study highlights that the use of indium in ITO manufacturing presents more significant challenges than the use of copper in graphene production. However, it suggests that copper may pose a potential resource constraint in the distant future. Cossutta et al. (2017) outlined a LCA examining three distinct graphene production methods. Their cradle-to-gate LCA delved into the environmental implications of these production processes. The findings underscored that nearly all processes witnessed environmental benefits with scale-up activities. Among the routes explored, chemical oxidation followed by thermal reduction emerged as the least environmentally impactful material route. Chaosukho et al. (2024) undertook a laboratory-scale optimization of perovskite solar cells, exploring gold and carbon electrodes. Employing primary data, they conducted a cradle-to-gate LCA. The analysis demonstrates that shifting from gold electrodes to carbon electrodes, produced through a low-temperature process, not only sustains comparable power conversion efficiency but also improves stability. This transition leads to substantial reductions in environmental impacts across all categories, notably an 86% decrease in global warming potential per 1 kWh of energy generated.

From the above studies, LCA method is feasible for the environmental performance assessment of electrodes. However, the R2R manufacturing process as an emerging technology has never considered. Moreover, it can be found that the benefits of copper recycling have never been assessed. To demonstrate the environmental performance of the production and environmental potential of copper recycling, conducting a LCA study for the graphene production via R2R CVD method is extremely urgent. We seek to propose a life cycle assessment for the production of graphene and copper recycling, which is critical for the beneficial use of graphene electrodes and provides the potential to substitute for ITO electrodes to realize sustainable production.

In this work, a prospective LCA study is conducted to investigate the environmental impacts of R2R produced CVD GTE and the environmental potential of recycling copper foil for sustainable production. We firstly extract and organize the process of TCVD lab scenario and MWPCVD scenario from previous literature. Based on that, the TCVD industry baseline scenario and recycling scenario are designed by this work. A cradle-to-grave LCA for four scenarios mentioned above is performed to investigate the environmental impacts. Primary energy consumption and global warming are calculated, and 9 impact categories are investigated using CML-AI method. These results are not only compared with existing transparent electrode technologies to place GTE in the context of proven technologies, but also with those of graphene production in previous literature to show the accuracy of the results.

This paper is organized as follows. Section 2 presents the procedure of 4 graphene production routes with the material and equipment. In section 3, the goal and scope definition, life cycle inventory and life cycle impact categories are provided according to the standard procedure of an LCA. Section 4 investigates the life cycle impact assessment, including CML-IA baseline, primary energy consumption and IPCC 2013 GWP 100a. The results are compared with those of previous work and ITO production. Finally, insights are provided into the future of graphene production.

2. Material and methods

In this section, the four production scenarios are introduced first. Then, the LCA method and related data are provided in Section 2.2.

2.1. Production scenarios

This work considers four production scenarios, including TCVD lab, TCVD industry baseline, TCVD industry recycling and MWPCVD.

TCVD lab scenario: is to produce the GTE by TCVD method in lab

scale.

TCVD industry baseline scenario: is to produce the GTE by TCVD method in industry scale but does not consider the recycling of the materials (especially recycling copper foils).

TCVD industry recycling scenario: is to produce the GTE by TCVD method in industry scale and considers the recycling of the materials.

MWPCVD scenario: is to produce the GTE by MWPCVD method in industry scale.

This section only shows the production process in TCVD industry baseline scenario, which we are developing based on our research and the previous literature (Bae et al., 2010; Kidambi et al., 2018; Polsen et al., 2015). Other scenarios are shown in the supplementary Information. The TCVD industry scenario are shown in the Fig. 1 to give an overall picture for the production process. The manufacturing of GTE consists of the following 3 steps: preclean of copper foil, chemical vapor deposition and transfer of graphene. For TCVD industry scenario, the detailed process is provided as follows:

2.1.1. Step 1: preclean of copper foil

The width of every graphene foil line is set as 305 mm to match the width of organic solar cell (Espinosa et al., 2011). The success of 30-inch graphene films for transparent electrodes via R2R production proves the feasibility of the width (Bae et al., 2010). Based on the production experiences, there are three graphene electrodes processed simultaneously in one production line (Espinosa et al., 2012). The processing length in one oven is set as 10 m. The unwinder has a brake works to keep the appropriate tension. It is known that the copper foil is covered by oxide layers, requiring to be precleaned before the graphene synthesis. The preclean of copper foil includes 3 steps.

1. Etch organic residues.
2. Rinse the impurities.
3. Dry the foil.

In the rapid mass fabrication, the processing should be continuous to increase the efficiency and meet the requirement of follow-up processes. The standstill processing for annealing and growing graphene is an essential procedure. It is reasonable to set the processing speed of current graphene production as 20 m/h slightly exceeding the speed 15 m/h reported by (Polsen et al., 2015) 5 years ago. This speed also meets the requirement of OPV manufacture whose processing speed is 18 m/h (Espinosa et al., 2011).

2.1.2. Step 2: chemical vapor deposition process

The CVD process includes two steps:

1. Annealing of copper foil.
2. Graphene growth.

In the industrial production, there are two ovens each with a 10 m heating line for annealing and graphene growth respectively, which is set based on the testbed of Clean Energy Institute of UW(Zweibel, 2010). Graphene lines are separated by the quartz panel to ensure the copper foil can be heated evenly. The line is along the center of two quartz panels. The quartz panels also create an enclosed path for H_2 and CH_4 passing through. The flow rate of H_2 and CH_4 is not changed as 0.2 cm/min and 0.6 cm/min. Recycling the mixed gas can reduce the raw material, but currently there is no cost-effective and energy-effective method to separate the mixed gas. Therefore, this work does not consider the gas recycling.

2.1.3. Step 3: graphene transfer

After growth, the graphene layer grown on copper foil needs to be transferred to the substrate. The transfer via R2R method is provided with 5 steps:

1. Adhere to polymer supports.
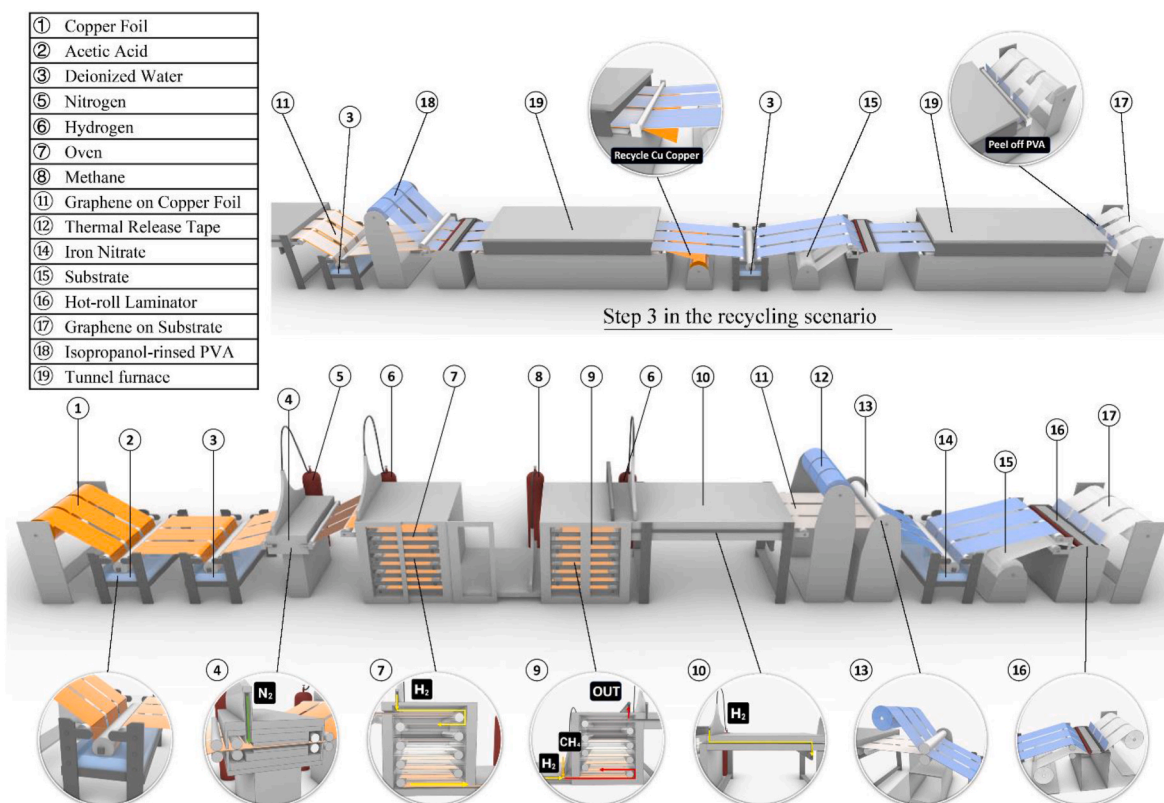
Fig. 1. The TCVD industry baseline scenario is on the bottom of the figure; the transfer processes in TCVD recycling scenario is on the right top of the figure. Note that this figure only shows processes influencing the environmental impacts of the graphene production, but not the specific processing details in practice.

2. Etch away copper.
3. Rinse the impurities.
4. Transfer printing to a target substrate.
5. Remove the polymer supports.

Because the production route has not been mature. This study makes some reasonable assumption in the design and calculation. They are listed as follows:

1. The speed of the conveyor does not lose in the whole production process.
2. In the recycling scenario, the 95% of copper foil can be recycled.
3. The amounts of input N_2 and deionized water are set based on the experience in the lab.
4. The chemical reaction is assumed to be sufficient without any loss. The uncertainty analysis is conducted to analyse the uncertainty caused by the amount of the input material.
5. All devices are supported by electricity.
6. The purity of the input materials is same with the experimental design.

2.2. Life cycle assessment

2.2.1. Goal and scope definition

Life Cycle Assessment (LCA) approach provides a highlighted foundation for carrying out environmental impacts. This work conducts an LCA study according to International Organization for Standardization 14040:2006 and 14044:2006 sections. Particularly, all data collected and used are operated in SimaPro v.8.3 from Ecoinvent database v.3.8 and previous literatures.

The goal of this work is to assess a potential life cycle impacts for GTE production via R2R CVD method. In this work, four production scenarios of GTE production have been provided in the last section. The functional unit is set as 1 m^2 of the GTE production. The scope of this LCA study is shown in Fig. 2.

Raw material's collection, fabrication, use and disposal are four main parts in the life cycle of a product or process. Based on the ISO standard indications, the determination of the system boundary is essential for the assessment of the product or process (Li et al., 2020). This work is a cradle-to-gate LCA and the inclusive processes have been shown in Supplementary Information. As can be seen, raw material extraction and graphene production are included in this work. The use stage is not considered because there is little environmental impact while graphene working in the electronic components. Due to research about graphene disposal, the disposal is not considered, either. Moreover, the energy requirement from transport of materials to the production plant is neglected because it only accounts for a little share of the whole energy requirement. This assumption is used in previous LCA research for graphene production (Arvidsson et al., 2016; Cossutta et al., 2017).

The retrospective approach is taken to account for environmental impacts in terms of the laboratory production, because the technology

routes for graphene production have been certain (Zhu et al., 2019). As for the industrial production, the processes are currently under development with many uncertain factors, the prospective approach is taken aiming at the evolution of system development over time. Furthermore, this is a consequential LCA study due to the comparison of differences in the production methods and scales.

2.2.2. Life cycle inventory analysis

Inventory analysis plays an important role in an LCA research. Based on the mentioned system boundary, the LCI of every scenario is collected into two categories: material inventory and energy inventory, shown in supplementary information. As for the Ecoinvent dataset v.3.8, it was released in 2021. This study uses the data collected in national scale of European regions, shown as "RER" in the dataset. The database is a comprehensive repository encompassing a wide range of sectors, containing over 20,000 datasets that model various human activities and processes.

In terms of material inventory, the amounts of raw materials and emission during production per functional unit are included. The functional unit is set as 1 m^2 of the GTE production. In the mass production, it is not easy to directly get the results for 1 m^2. For convenience, the embodied energy of 10 m processing graphene is firstly calculated in TCVD lab scenario, TCVD industry baseline scenario and MWPCVD scenario. In terms of TCVD industry recycling scenario, 20 m processing graphene is calculated firstly because of the processing length in one oven. Then, the results will be calculated to show functional unit amounts. The material inventory of 1 m^2 GTE production is exhibited in Table 1 and Table 2. The specific process of data acquisition and estimation is shown in section S5.

We use 3 steps to show the allocation. The first step is to define the scope of the LCA study shown in Fig. 2. Next, the flows of the production in the four scenarios are shown in Figs. S1, S2, S3, and S4. In the production flow, the input and output materials and production conditions can be found. And then, Tables 1 and 2 clearly show the input and output materials and electricity consumption of 1 m^2 GTE production. In this way, the allocation is shown in the paper.

2.2.3. Life cycle impact assessment

Life cycle impact assessment is the approach to convert LCI data to

Fig. 2. Scope of this LCA study.

Table 1
Material inventory for 1m^b GTE production in four scenarios.

Material	TCVD lab	TCVD industry	TCVD recycling	MWPCVD	Unit
Step 1: Preclean of copper foil					
Cu (36 μm) [a]	322.56			295.7	g
Cu (25 μm) [a]		224.00	0.23		g
N_2 [b,c]	68.90	40.98	40.98	70.86	g
CH_3COOH [a]	0.31	0.21	0.22	0.28	g
Deionized water [b]	130	130	130	130	g
Step 2: Chemical vapor deposition					
H_2 [a,c]	0.53	0.01	0.005	0.05	g
CH_4 [a,c]	67.48	0.28	0.14	0.41	g
Step 3: Graphene transfer					
$Fe(NO_3)_3$ [a]	2456.10	1705.60		2251.4	g
Polyester [a]	138	138		138	g
Acrylic [a]	1	1		1	m^2
Deionized water [b]	130	130	130	130	g
PVA [a]			31.2		g
Emission					
CO_2 [a]	16.52	16.52		16.52	g
C_2H_4 [a]	12.98	12.98		12.98	g

*The data is collected from different sources.
[a] - theoretical calculation.
[b] - estimates.
[c] - actual measurements.

Table 2
Electricity consumptions during R2R processing of 1m[b] GTE production in four scenarios.

Processing	TCVD lab	TCVD industry	TCVD recycling	MWPCVD	Unit
Step 1: Preclean of copper foil					
Delivery [a]	755.9	983.6	491.8	81.6	Wh
Step 2: Chemical vapor deposition					
Oven [a,b]	680315.0	7082.0	3541.0		Wh
Flow controller [a]	15307.1	4.5	2.3	18.4	Wh
Pressure controller [a]	6866.1	19.7	9.9		Wh
Microwave generator [a]				16320	Wh
Rotary pump [a]				122.4	Wh
Step 3: Graphene transfer					
Laminator [a]	7086.6	29.5	49.20	153.0	Wh
Unwinder [a]	944.9			36.7	Wh
Tunnel furnace [a,c]			29.5		Wh
Total	711269.3	8119.0	4124	16737.4	Wh

*The data is collected from different sources.
[a] - theoretical calculation.
[b] - estimates.
[c] - actual measurements.

several indicators, aiming at evaluating the contributors of material and energy consumption to a set of potential environmental impacts. In this work, CML-IA baseline method, primary energy demand and IPCC 2013 GWP 100a are applied to show the environmental profile of GTE production by R2R CVD method.

CML-IA baseline method introduces a serious of impact categories for the evaluation of the environmental impacts. This paper analyses 9 impact categories by this method, including abiotic depletion (AD), ozone layer depletion (OLD), human toxicity (HT), fresh water aquatic ecotox (FWAE), marine aquatic ecotoxicity (MAE), terrestrial ecotoxicity (TE), photochemical oxidation (PO), acidification (AC) and eutrophication (EU). The CML-IA baseline refers to a LCA methodology proposed by Leiden University. This methodology adopts a problem-oriented midpoint approach and offers a comprehensive list of impact assessments for mandatory impact categories commonly utilized in LCAs. The baseline indicators are recommended for streamlined studies, facilitating simplified assessments of environmental impacts. As a result, the CML-IA baseline method stands out as one of the optimal choices for evaluating the environmental impacts associated with various life phases. This method offers aggregated results, providing a comprehensive overview of environmental impacts across different stages, thereby facilitating the upscaling details of the production.

Primary energy consumption and global warming are two most remarkable environmental consequences. Primary energy consumption refers to the supply or direct use at the source of energy that has not been transformed or converted to any forms of secondary energy. The global warming is the total greenhouse gas emissions that are generated by the production. The investigation of global warming is based on IPCC with a timeframe of 100 years. They are two common environment indicators to directly show the energy consumption and greenhouse gas emissions, which researchers concern most. Both primary energy consumption and global warming potential enable comparisons between production by different methods and different production scales. By using these indicators, researchers can identify opportunities for finding a more suitable production route to reduce environmental impacts associated with graphene electrode production.

In this work, both thermal energy share and electrical energy share in the production of GTE are transformed to the equivalent primary energy consumption at the final-result stage. The conversion efficiency is referred to the technology mix of the electricity supply system that is variable between different period and different regions, even in one country. In this work, the electricity is the average electricity mix in

Europe. In terms of thermal energy consumption, the heat is supported from an industrial natural-gas plant. The heat conversion coefficient and electricity conversion coefficient are 2.38 MJ primary energy/MJ heat and 7.92 MJ primary energy/kW h electricity, respectively.

2.2.4. Uncertainty and sensitivity analysis

It is obvious that the accuracies of LCA results are fully influenced by the input parameters, however, whose reliability are not enough. In terms of primary energy consumption and global warming results, there must be fluctuations since the technologies are not sufficiently mature and have not reached industrial manufacture. Moreover, it is a significant challenge to get accurate input materials which are estimated based on previous research experience. As a result, this work uses the probability distribution to the mentioned input material amounts from the experience of previous researches. And how much these uncertain parameters can influence on the primary energy consumption and global warming will also be explored in the simulation software Simapro. Except insolation, the distributions of other parameters all follow the lognormal distribution. And insolation follows a normal distribution. This work sets 500000 trials for the Monte Carlo simulation. Sensitivity analyses for the simulation results are also conducted to identify the influence extent of input materials on the environmental performance in the production.

3. Results

3.1. Primary energy consumption and global warming

Among the impact categories, primary energy consumption and global warming attract enormous attention in the LCA study of GTE. According to the LCIs abstracted in the last section, their results with the contribution share of every material and processing step are calculated and shown in Fig. 3 (a) and (b), respectively. In the figures, other materials include acetic acid, nitrogen, deionized water, hydrogen and methane. This work makes some assumption about the amount of acetic acid, nitrogen and deionized water that are uncertain. Fig. 3 (a) and (b) show that these three materials have little influence on the primary energy consumption and global warming. Therefore, the results of this work are reliable.

As can be seen in Fig. 3 (a) and (b), no matter whether primary energy consumption and global warming, the impacts of GTE in lab-scale production via the R2R TCVD method are obviously higher than those in industrial production. In TCVD lab scenario, the electricity consumption of step 2 is the most important contributor to both impact indicators because of low utilization efficiency of the oven. The oven with 1200 W working power can only produce 63.5 m^2 graphene in 1 h, resulting in that the share of direct process energy in step 2 accounts for 93.49% (1564.4 MJ) and 78.96% (43.8 kg CO_2-eq) in terms of primary energy consumption and global warming, respectively. When looking at the share of the primary energy used for materials, the biggest share iron nitrate is responsible for only 2.38% (39.3 MJ) of the entire energy, followed by the copper foil and the thermal release tape, 1.85% (30.5 MJ) and 0.98% (16.2 MJ), respectively. The global warming shows a similar distribution as primary energy consumption. Therefore, in lab-scale, while comparing the impacts related to the electricity and the impacts related to materials, one can conclude that the latter can be rather negligible in the lab-scale production.

While employing one production method (TCVD), it is distinct that the scaled-up production from lab to fab saves the processing electricity consumption remarkably, reducing embedded primary energy from 1564.4 MJ down to 17.9 MJ in baseline scenario and to 9.1 MJ in recycling scenario. In terms of material inventory, iron nitrate dominates the two impacts while using the thermal release tape for graphene transfer. The reason for a big share residing in the iron nitrate is because nitric acid as its production raw material shows unfavourable environmental performance. Furthermore, the copper foil is also a major
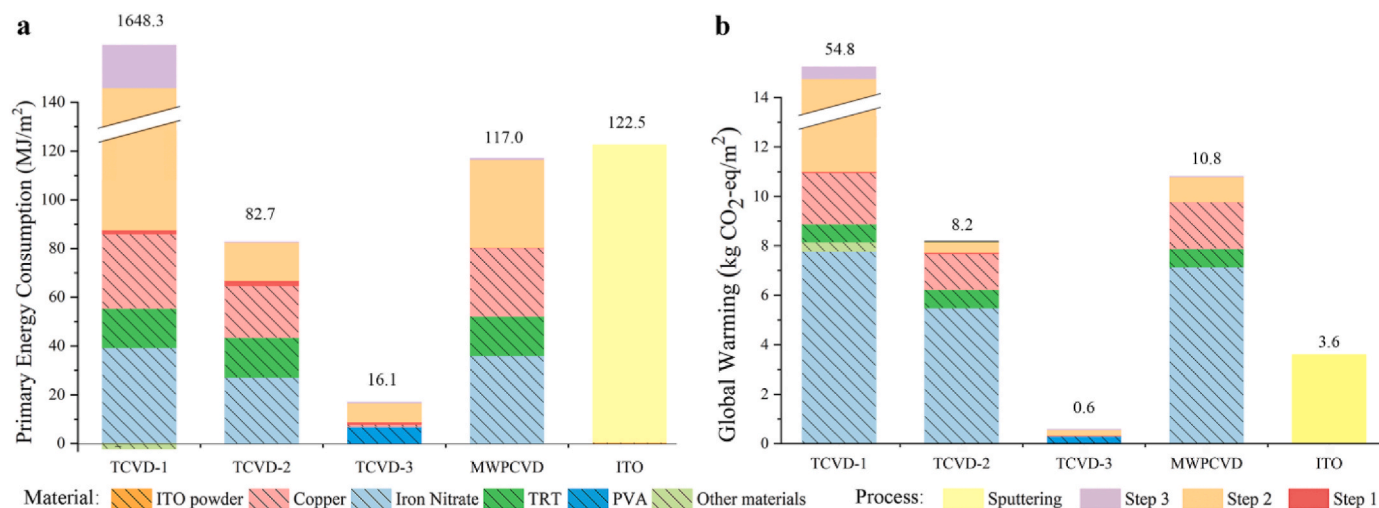
Fig. 3. (a) Distributions of the primary energy consumption for graphene production. (b) distributions of the global warming for graphene production.

contributor, whose primary energy consumption share is over 25% in graphene baseline scenario. It means the transfer method using thermal release tape is a block to decrease the energy consumption and GHG emission. As the substitute, the transfer method with PVA and laminator really has a considerably positive environmental effect because of iron nitrate free and copper foil recycled. Finally, it is noticed that the GTE in the recycling scenario consumes and emits only approximately 1% of the primary energy and GHG embedded in GTE produced in laboratory. It shows that the improvement of production procedure makes an extraordinary achievement.

From the results, it is also found that recycling copper foil shows environmental benefits in terms of primary energy consumption and global warming. In the recycling scenario, 66.6 MJ primary energy can be saved compared to the industrial baseline scenario. The primary energy consumption in the recycling scenario is only 19.5% of the industrial baseline scenario. In terms of global warming, it is low to 0.7 kg CO_2-eq by copper recycling and processing optimization. Although, the CO_2-eq emission of 1 m^2 ITO production is only 3.6 kg. The GTE production in the recycling scenario is only 19.4% of ITO production. The market analysis conducted indicates a significant milestone for the global photovoltaic sector in 2022, wherein the cumulative deployment of Building Integrated Photovoltaic applications reached an unprecedented 80 GW. If the power conversion efficiency of the PV panel is 20%. It means a 200 W PV panel requires 1 m^2 electrode. Using 1 m^2 GTE as alternative of ITO can save 3.6 kg CO_2-eq emission. Should all BIPV systems transition to employing perovskite solar cells integrated with GTE, the potential reduction in CO_2 emissions is estimated at 141.2 million kg.

As for MWPCVD scenario, its value of primary energy consumption and GHG emission are at the same level as those in TCVD baseline scenario. Furthermore, it is obvious that GTE produced by MWPCVD method and by TCVD method in baseline scenario have the same distributions in the results of global warming and primary energy consumption. Actually, using the mentioned two production methods with the same transfer method, not only are these two impact indicators the same, but other impacts also have the identical distributions. The reason is that different production methods in same scale consume the same raw materials with the similar percentage and the energy supply is only electricity.

As comparison, the results of ITO transparent electrode are also calculated and provided on the right. The primary energy consumption of industrial-scale GTE production is also lower than that of ITO transparent electrode, though, the LCIs of which is very conservative. In contrast, with respect to the GHG emission in production, ITO transparent electrode only shows a worse performance than GTE in low

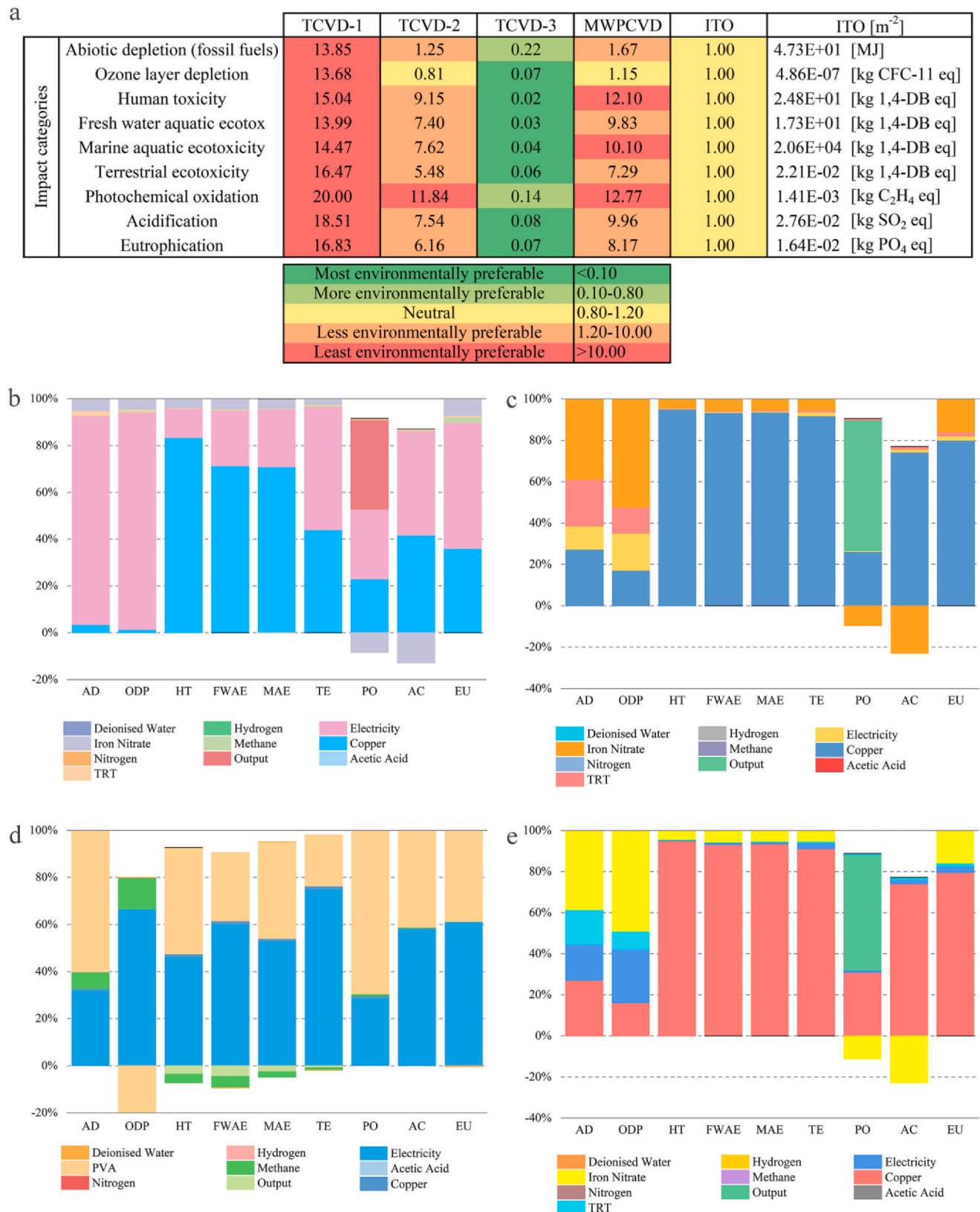energy scenario but has a better performance than other three scenarios.

3.2. CML-IA baseline

Fig. 4 displays comparison of the environmental impacts of ITO transparent electrode to graphene counterparts in different scenarios on a unit area production basis. As expected, lab-scale GTE production in the TCVD method has the highest impacts overall, which are at least 13 times than those of ITO transparent electrode.

Nevertheless, GTE produced by MWPCVD method and by TCVD method in baseline scenario also have further higher impacts, except for abiotic depletion and ozone layer depletion. In Fig. 4 (c) and (e), it can be found that the copper foil shows great influence, contributing more than 90% to human toxicity, fresh water aquatic ecotox, marine aquatic ecotoxicity and terrestrial ecotoxicity. More than 70% share of acidification and eutrophication are because of copper foil. Because the mining and processing of primary copper is an energy-intensive industry with large volumes of pollution and waste. The smelting process also creates a mass of low concentration sulphur dioxide, leading to the increase of these impacts. So, the copper recycling framework within the TCVD industry assumes pivotal significance in advancing the principles of sustainable GTE production. A salient focal point resides in the recycling of copper foil. From the results of recycling scenario, the process of copper foil recycling yields substantial potential to markedly diminish the overarching environmental footprint attributed to GTE manufacturing. Copper mining and processing operations can have significant water usage and can lead to water pollution through the discharge of mine tailings and wastewater. By recycling copper, the demand for water in the copper production process is reduced, contributing to water conservation efforts and reducing the potential for water pollution. Therefore, it can be found that Fresh water aquatic ecotox and Marine aquatic ecotoxicity show a sharp reduction in the recycling scenario. Mining activities associated with copper extraction can cause environmental degradation, including soil erosion, deforestation, and habitat destruction. Recycling copper reduces the demand for new copper production, thereby mitigating these environmental impacts and helping to preserve ecosystems. So, environmental performance in other indicators of CML-IA baseline shows a far improvement.

The main contribution to photochemical oxidation comes from the output of the production. The direct emission in production is C_2H_4 emitting to the atmosphere during the tape thermal releasing process, which forms organic emission impacting the environment. Abiotic depletion (fossil fuels) and ozone layer depletion are impacted most by iron nitrate with approximately 40% share and 50% share, respectively. The use of nitric acid to synthesize iron nitrate is mainly responsible for

a

	TCVD-1	TCVD-2	TCVD-3	MWPCVD	ITO	ITO [m⁻²]
Abiotic depletion (fossil fuels)	13.85	1.25	0.22	1.67	1.00	4.73E+01 [MJ]
Ozone layer depletion	13.68	0.81	0.07	1.15	1.00	4.86E-07 [kg CFC-11 eq]
Human toxicity	15.04	9.15	0.02	12.10	1.00	2.48E+01 [kg 1,4-DB eq]
Fresh water aquatic ecotox	13.99	7.40	0.03	9.83	1.00	1.73E+01 [kg 1,4-DB eq]
Marine aquatic ecotoxicity	14.47	7.62	0.04	10.10	1.00	2.06E+04 [kg 1,4-DB eq]
Terrestrial ecotoxicity	16.47	5.48	0.06	7.29	1.00	2.21E-02 [kg 1,4-DB eq]
Photochemical oxidation	20.00	11.84	0.14	12.77	1.00	1.41E-03 [kg C₂H₄ eq]
Acidification	18.51	7.54	0.08	9.96	1.00	2.76E-02 [kg SO₂ eq]
Eutrophication	16.83	6.16	0.07	8.17	1.00	1.64E-02 [kg PO₄ eq]

Most environmentally preferable	<0.10
More environmentally preferable	0.10-0.80
Neutral	0.80-1.20
Less environmentally preferable	1.20-10.00
Least environmentally preferable	>10.00

Fig. 4. Environmental impacts of 1 m² GTE by CML-IA methodology. (a) Comparison of the environmental impacts of ITO to graphene produced in different scenarios. Yellow coding means that the impact of graphene per m² is similar to that of ITO, which is the reference case. Values coded green indicate better performance, while orange and red indicate worse performance. The actual values of the ITO environmental impacts are shown in the right column. (b) Environmental impacts of 1 m² GTE in TCVD lab scenario. (c) Environmental impacts of 1 m² GTE in TCVD industry baseline scenario. (d) Environmental impacts of 1 m² GTE in TCVD industry recycling scenario. e. Environmental impacts of 1 m² GTE in MWPCVD scenario. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the high impacts. Also, the abiotic depletion (fossil fuels) and ozone layer depletion scores are influenced by the use of copper foil, thermal release tape and electricity consumption, which contribute more than 10%. Comparing these two methods, TCVD baseline scenario shows a slightly better performance in environment impacts than MWPCVD scenario due to less electricity consumption.

Because the adjusting of graphene transfer process excludes the effects of copper foil, thermal release tape and C_2H_4 emission, the GTE recycling scenario shows an extremely remarkable improvement in terms of environmental impacts. The impacts of graphene recycling scenario vary from 0.02 to 0.22 of the ITO reference values. Also, the impacts of GTE production in industry recycling scenario vary from 0.01 to 0.18 of the values in industry baseline scenario. This indicates the extremely low environmental impacts from this new graphene transfer process with only laminators and PVA. Future effort should be made in promoting the large-scale application of this process replacing the use of thermal release tape and PMMA. The environmental impacts associated with individual processing steps can be determined by assessing the electricity consumption ratio between these steps. The ratio can be obtained in Table 2.

4. Discussion

4.1. Uncertainty analysis and sensitive analysis

Fig. 5 provides the probability distributions of the primary energy consumption and global warming for GTE production in the industry baseline scenario. The fitting curves have been provided by the lognormal distribution with 95% confident regions. Both forecast distributions are in a narrow range and the most possible results are from the highest bars. Forecast distributions are asymmetrical because the influence factors are not linear with environmental indicators. In this section, we use the Monte Carol method to simulate the region that values are possible to happen. Then the values that have 95% probability to be the true values are selected as the final results.

As can be seen, the distributions of both primary energy consumption forecasts and global warming are relatively robust according to the lower rate of standard deviation. The values of the lowest probabilities in the 95% confident regions show the huge potential for the environmental sustainability. Regarding primary energy consumption, the highest value in the 95% confident regions is 91.5 MJ/m^2. It means that GTE production in the industry baseline scenario is capable of consuming 91.5 MJ/m^2 in the worst condition. In the best condition, producing 1 m^2 GTE only consumes 75.8 MJ primary energy. As for global warming, the environmental impact in terms of global warming

for 1 m^2 GTE production is up to 8.8 kg CO_2-eq.

Then, the sensitive analyses for two forecasts can also be seen in Fig. 5 (b). In the results, only the factors whose contributions are more than 10% are shown. The uncertainty of the copper, Iron Nitrate, electricity, TRT lead to the distribution of the primary energy consumption. The negative sign means that the increase of these parameters results in the decrease of the forecast indicator. As can be seen, the copper is the dominant parameter, which contributes 38.9% in these four variables to the primary energy consumption. As for global warming, the copper, also as the most influential parameter, contributes 35.2% among all the input materials and energy. These results show the importance to recycle the copper foil in the production again.

4.2. Comparison with previous studies

The degree of technological maturity and production scale wield substantial influence over the LCA outcomes for emerging technologies. Advancements in technology maturity or increased production scale have the potential to mitigate environmental impacts per unit output by enhancing the material and energy efficiency of the underlying equipment or processes. The Technology Readiness Level (TRL) framework serves as a pivotal metric to gauge the technological maturity within LCA studies. TRLs offer a valuable mechanism for technology developers to monitor the evolution of emerging technologies and their progression into production. At the core of LCA comparisons lies the concept of the functional unit, which facilitates the juxtaposition of various technologies. However, at lower TRLs, the functionality of emerging technologies may not be fully delineated and may evolve with increasing maturity. So, in the section, we only compare the results of GTE production but not ITO. In the context of this study, the production routes of GTE are diverse and remain at the initial stage, all falling within the same TRL. Despite their uniform research boundaries, variations in resource consumption across different studies pose a notable limitation in comparative analysis.

For the graphene transparent electrode, two studies focusing on the LCA field have been published in the past seven years. They both presented energy use calculation of GTE production by CVD in different scenarios. The first one (2016) calculated based on stoichiometric calculation and assuming conditions, which are different from this paper based on actual production processes (Arvidsson et al., 2016). The work provided three scenarios including the scenario with high methane application rate, the scenario with low residence time, and the scenario with high recovery of methane energy. Fig. 6 shows the results in the bar to the left. But the energy results in the first work (2016) were thermal-equivalent amount, which has been transformed to equivalent
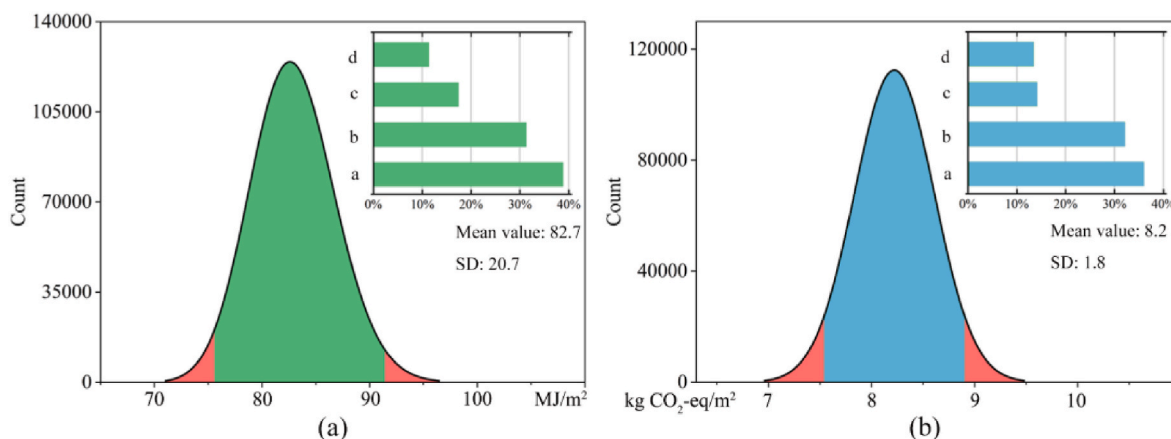

Fig. 5. Uncertainty analysis results for GTE production in the industry baseline scenario. (a) Probability distribution and sensitivity analysis results for primary energy consumption. (b) Probability distribution and sensitivity analysis results for global warming. a represents copper. b represents Iron Nitrate. c represents Electricity. d represents TRT.
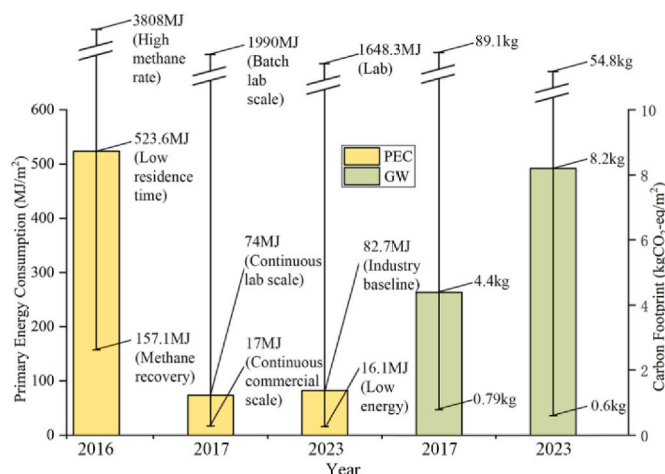
Fig. 6. Comparison with previous work. 2023 is this work. PEC: primary energy consumption. GW: global warming.

primary energy from 157.1 MJ/m^2 to 3808 MJ/m^2 for comparison purposes. No matter which analyzed scenario, the production is in lab scale, resulting in that energy use results are higher compared to our work (2023). However, the primary energy consumption of methane accounts for a big share (68%) in first work (2016), which is extremely different from this work. The reason is that the production method of methane has been transformed, mainly from biogas which is cleaner.

The second work (2017) considered three scenarios, including batch lab scale, continuous lab scale and continuous commercial scale (Cossutta et al., 2017). Their results (2017) are in line with the results of this work; however, this can only prove the energy use trend of GTE production in current production capacity. Actually, recipes of two work are slightly different and our work uses the methane as the main feedstock while theirs is based on hexane. In terms of process, their work (2017) did not consider the transfer process, which means the influence of iron nitrate and acrylic is excluded. Furthermore, their estimated electricity consumption (10 kJ/m^2) is lower than that in our work (4124 kJ/m^2). The reason is that their electricity consumption estimation in annealing process is assumed by 90% heat transfer efficiency and adiabatic conditions while ours is collected from data of oven manufacturer. Therefore, the conclusion can be drawn the actual heat loss in processing is further higher than results by the theorical calculation. As for the global warming, their results vary from 0.79 kg CO$_2$-eq/m^2 to 89.1 kg CO$_2$-eq/m^2. The range of ours is 0.6–54.8 kg CO$_2$-eq/m^2. The results are highly consistent once again despite the existence of these difference. The consistency in different impacts shows the level of GTE production in CVD method.

These works prove that the primary energy consumption and global warming in industrial scale are in the order of magnitude of 10 MJ/m^2 and 0.1 kg CO$_2$-eq/m^2. In current stage, this is reaching the limit of GTE production. Because all materials in the inventory are almost in the lowest amount or recycled. Meanwhile, the processing is optimized to reduce the electricity consumption while the power of devices is fixed. It is not easy to make more improvements in current processes for lower environmental impacts. If pursuing further less environmental impacts, finding a radical change in terms of the production route should become the emphasis of efforts.

4.3. Limitations

In conducting LCA studies, established industrial processes typically provide the foundation for data collection. However, when assessing emerging technologies, reliance on data derived from laboratory-scale processes is often necessary. Despite the utilization of direct and precise process data in LCA, findings based on lab-scale data may not

accurately reflect the environmental impacts upon scaling up to a typical commercial scale. Acquiring adequate inventory data at low TRLs poses a formidable challenge, exacerbated for emerging technologies by factors such as the absence of historical data, confidentiality surrounding industrial processes, and the use of novel materials. The unavailability or protracted gathering process of primary data often necessitates resorting to secondary data as a practical expedient to furnish technology developers with timely decision-making information. Furthermore, novel processes frequently entail the utilization of new materials not prevalent in existing databases, thus limiting the availability of comprehensive raw material data. To mitigate the issue of missing data, the recourse to secondary or proxy data becomes imperative, notwithstanding the attendant challenges. In the present study, uncertainty and sensitivity analyses are conducted to alleviate errors arising from input data and estimations; however, these measures are incapable of fully remedying all discrepancies.

The viability of GTE production utilizing recycled copper foils is substantiated in the lab scenario; however, uncertainties persist regarding its performance in mass production, thereby impacting the reliability of environmental impact assessments. This uncertainty poses a notable limitation to the conclusive findings on environmental impacts. The application of LCA to evaluate GTE production, particularly as an emerging technology at a low TRL, is susceptible to inherent uncertainties inherent in the LCA methodology. Notably, the development of technology in the laboratory setting may introduce biases into LCA results. Therefore, decision-makers must grasp the inherent uncertainties in LCA and their ramifications on decision-making processes to ensure informed decision-making. So now, the technology should focus on the demonstration of viability of GTE production utilizing recycled copper foils in mass production. But so far, the technologies in the industrial baseline scenario are proved for the production. We can also find alternatives for the copper, Iron Nitrate, and TRT.

Copper foil emerges as the primary environmental hotspot in GTE production. Recycling copper foil presents significant potential for environmental benefits; however, its effectiveness in mass production remains a challenge. Additionally, the lifecycle of recycled copper foil is uncertain. Therefore, further experiments are necessary to evaluate the quality of GTEs produced with recycled copper foils. In the perspective of policy, policymakers should support research and development in sustainable production technologies such as microwave plasma chemical CVD. Grant programs and innovation funds can accelerate the transition from lab-scale research to industrial-scale applications, fostering cleaner production methods. Governments can set specific emission reduction targets for the electronics and photovoltaic industries. By incorporating GTEs produced through recycled copper foil into these targets, policymakers can drive the adoption of more sustainable production practices, aligning industry efforts with national and international climate goals.

5. Conclusions

This paper is a prospective LCA study to investigate the environmental impacts of R2R produced CVD GTE and the environmental potential of recycling copper foil for sustainable production. Four production scenarios were developed to promote the lab-to-fab progress, including TCVD lab scenario, TCVD industry baseline scenario, TCVD industry recycling scenario and MWPCVD scenario. The LCA results in this study are compared to the previous literation and have been verified within the reasonable limits.

The scaled-up production from lab to fab is very important for the environmental sustainability of GTE production. The upgrading saves the processing electricity consumption remarkably, reducing embedded primary energy from 1564.4 MJ down to 17.9 MJ in baseline scenario and to 9.1 MJ in recycling scenario. Furthermore, it can be proved that the primary energy consumption and global warming in industrial scale are in the order of magnitude of 10 MJ/m^2 and 0.1 kg CO$_2$-eq/m^2. In

current stage, this is reaching the limit of GTE production.

This study also found the environmental hotspots in the GTE production. From the results, using thermal release tape is a block to decrease the energy consumption and GHG emission. In the recycling scenario, most environmental impacts dramatically drop because of copper foil recycling and iron nitrate free using the transfer method with PVA and laminator. Therefore, it suggests to using PVA and laminator to replace thermal release tape in the future technology development.

From the results, it can be found that the copper foil is a major contributor, whose primary energy consumption share is over 25% in graphene baseline scenario. Through the recycling of copper foil and optimization of processing techniques, in the industry recycling scenario for GTE production, the environmental impacts vary from 0.01 to 0.18 of the values observed in the industry baseline scenario. If all BIPV systems transition to employing perovskite solar cells with GTE in copper recycling scenario, the potential reduction in CO_2 emissions is estimated at 141.2 million kilograms in one year.

This paper provides insights into the major environmental contributors in the GTE production. Copper foil recycling shows an environmental potential for cleaner production of GTE. The findings serve as a roadmap for the industry, highlighting key areas where improvements can be made to upscale production while minimizing environmental impact, particularly by identifying and avoiding the use of materials that contribute significantly to environmental burdens. In the future, we will focus more on the effects of TRL on the LCA study of the scaled-up GTE production. Also, the ongoing development of technology for mass-producing GTE will persist.

Abbreviations

AC	Acidification
AD	Abiotic depletion
CH_4	Methane
C_2H_4	Ethylene
Cu	Copper
CVD	Chemical vapor deposition
EU	Eutrophication
$Fe(NO_3)_3$	Ferric nitrate
FWAE	Fresh water aquatic ecotox
GHG	Greenhouse gas
GTE	Graphene transparent electrode
GW	global warming
H_2	Hydrogen
HT	Human toxicity
ITO	Indium Tin Oxide
MAE	Marine aquatic ecotoxicity
MPCVD	Microwave plasma chemical vapor deposition
N_2	Nitrogen
OLD	Ozone layer depletion
PEC	Primary energy consumption
PMMA	Polymethyl methacrylate
PO	Photochemical oxidation
PVA	Polyvinyl alcohol
R2R	Roll-to-roll
TCVD	Thermal chemical vapor deposition
TE	Terrestrial ecotoxicity

CRediT authorship contribution statement

Qingxiang Li: Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **Malte Ruben Vogt:** Supervision, Investigation. **Haoxu Wang:** Investigation. **Carol Monticelli:** Supervision. **Alessandra Zanelli:** Supervision.

Declaration of competing interest

I confirm that we have mentioned all organizations that funded our research in the Acknowledgements section of my submission, including grant numbers where appropriate. We declare that we have no commercial or associative interest that represents a conflict of interest with other people or organizations that can inappropriately influence our work entitled, "Life Cycle Assessment of Roll-To-Roll Produced Chemical Vapor Deposition Graphene Transparent Electrodes towards Copper Foil Recycling".

Data availability

The authors are unable or have chosen not to specify which data has been used.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2024.143068.

References

Arvidsson, R., Kushnir, D., Molander, S., Sanden, B.A., 2016. Energy and resource use assessment of graphene as a substitute for indium tin oxide in transparent electrodes. J. Clean. Prod. 132, 289–297.

Bae, S., Kim, H., Lee, Y., Xu, X.F., Park, J.S., Zheng, Y., Balakrishnan, J., Lei, T., Kim, H. R., Song, Y.I., Kim, Y.J., Kim, K.S., Ozyilmaz, B., Ahn, J.H., Hong, B.H., Iijima, S., 2010. Roll-to-roll production of 30-inch graphene films for transparent electrodes. Nat. Nanotechnol. 5 (8), 574–578.

Chaosukho, S., Meeklinhom, S., Rodbuntum, S., Sukgorn, N., Kaewprajak, A., Kumnorkaew, P., Varabuntoonvit, V., 2024. Life cycle assessment of perovskite solar cells with alternative carbon electrode. Environ. Impact Assess. Rev. 106, 107462. https://doi.org/10.1016/j.eiar.2024.107462.

Chen, Y., Yue, Y.Y., Wang, S.R., Zhang, N., Feng, J., Sun, H.B., 2019. Graphene as a transparent and conductive electrode for organic optoelectronic devices. Adv Electron Mater 5 (10).

Cossutta, M., McKechnie, J., Pickering, S.J., 2017. A comparative LCA of different graphene production routes. Green Chem. 19 (24), 5874–5884.

Espinosa, N., Garcia-Valverde, R., Urbina, A., Krebs, F.C., 2011. A life cycle analysis of polymer solar cell modules prepared using roll-to-roll methods under ambient conditions. Sol. Energy Mater. Sol. Cell. 95 (5), 1293–1302.

Espinosa, N., Hosel, M., Angmo, D., Krebs, F.C., 2012. Solar cells with one-day energy payback for the factories of the future. Energy Environ. Sci. 5 (1), 5117–5132.

Kidambi, P.R., Mariappan, D.D., Dee, N.T., Vyatskikh, A., Zhang, S., Karnik, R., Hart, A. J., 2018. A scalable route to nanoporous large-area atomically thin graphene membranes by roll-to-roll chemical vapor deposition and polymer support casting. Acs Appl Mater Inter 10 (12), 10369–10378.

Kim, M., Zhang, Y., Verlinden, P., Hallam, B., 2022. Towards sustainable silicon PV manufacturing at the terawatt level. Presented at the SiliconPV 2021. In: The 11th International Conference on Crystalline Silicon Photovoltaics, Hamelin, Germany / Online, p. 090001. https://doi.org/10.1063/5.0090424.

Koo, D., Jung, S., Seo, J., Jeong, G., Choi, Y., Lee, J., Lee, S.M., Cho, Y., Jeong, M., Lee, J., Oh, J., Yang, C., Park, H., 2020. Flexible organic solar cells over 15% efficiency with polyimide-integrated graphene electrodes. Joule 4 (5), 1021–1034.

Kushnir, D., Sanden, B.A., 2008. Energy requirements of carbon nanoparticle production. J. Ind. Ecol. 12 (3), 360–375.

Li, Q., Chen, Z., Li, X., Brancart, S., Overend, M., 2024a. Vertical perovskite solar cell envelope for the circular economy: a case study using life cycle cost analysis in Europe. J. Clean. Prod. 467, 143017 https://doi.org/10.1016/j.jclepro.2024.143017.

Li, Q., Li, T., Kutlu, A., Zanelli, A., 2024b. Life cycle cost analysis and life cycle assessment of ETFE cushion integrated transparent organic/perovskite solar cells: comparison with PV glazing skylight. J. Build. Eng. 87, 109140 https://doi.org/10.1016/j.jobe.2024.109140.

Li, Q., Monticelli, C., Kutlu, A., Zanelli, A., 2024c. Environmental performance analysis of textile envelope integrated flexible photovoltaic using life cycle assessment approach. J. Build. Eng. 89, 109348 https://doi.org/10.1016/j.jobe.2024.109348.

Li, Q., Monticelli, C., Kutlu, A., Zanelli, A., 2023. Feasibility of textile envelope integrated flexible photovoltaic in Europe: carbon footprint assessment and life cycle cost analysis. J. Clean. Prod. 430, 139716 https://doi.org/10.1016/j.jclepro.2023.139716.

Li, Q., Monticelli, C., Zanelli, A., 2022. Life cycle assessment of organic solar cells and perovskite solar cells with graphene transparent electrodes. Renew. Energy 195, 906–917. https://doi.org/10.1016/j.renene.2022.06.075.

Li, Q., Yang, G., Gao, C., Huang, Y., Zhang, J., Huang, D., Zhao, B., Chen, X., Chen, B.M., 2024d. Single drone-based 3D reconstruction approach to improve public engagement in conservation of heritage buildings: a case of Hakka Tulou. J. Build. Eng. 87, 108954 https://doi.org/10.1016/j.jobe.2024.108954.

Li, Q., Zanelli, A., 2021. A review on fabrication and applications of textile envelope integrated flexible photovoltaic systems. Renew. Sustain. Energy Rev. 139, 110678 https://doi.org/10.1016/j.rser.2020.110678.

Li, Q., Zhu, L., Sun, Y., Lu, L., Yang, Y., 2020. Performance prediction of Building Integrated Photovoltaics under no-shading, shading and masking conditions using a multi-physics model. Energy 213, 118795. https://doi.org/10.1016/j.energy.2020.118795.

Long, L., Li, Q., Gan, Z., Mu, J., Overend, M., Zhang, D., 2023. Life cycle assessment of stone buildings in the Taihang mountains of Hebei province: evolution towards cleaner production and operation. J. Clean. Prod. 399, 136625 https://doi.org/10.1016/j.jclepro.2023.136625.

Novoselov, K.S., Geim, A.K., Morozov, S.V., Jiang, D.-e., Zhang, Y., Dubonos, S.V., Grigorieva, I.V., Firsov, A.A.J.s., 2004. Electric field effect in atomically thin carbon films, 306 (5696), 666–669.

Pizzocchero, F., Jessen, B.S., Gammelgaard, L., Andryieuski, A., Whelan, P.R., Shivayogimath, A., Caridad, J.M., Kling, J., Petrone, N., Tang, P.T.J.A.o., 2022. Chemical Vapor-Deposited Graphene on Ultraflat Copper Foils for van der Waals Hetero-Assembly, 7 (26), 22626–22632.

Polsen, E.S., McNerny, D.Q., Viswanath, B., Pattinson, S.W., Hart, A.J., 2015. High-speed roll-to-roll manufacturing of graphene using a concentric tube CVD reactor. Sci Rep-Uk 5.

Wang, Y., Zheng, Y., Xu, X., Dubuisson, E., Bao, Q., Lu, J., Loh, K.P.J.A.n., 2011. Electrochemical delamination of CVD-grown graphene film: toward the recyclable use of copper catalyst, 5 (12), 9927–9933.

Xin, H., Li, W., 2018. A review on high throughput roll-to-roll manufacturing of chemical vapor deposition graphene. Appl. Phys. Rev. 5 (3).

Yamada, T., Ishihara, M., Hasegawa, M., 2013. Large area coating of graphene at low temperature using a roll-to-roll microwave plasma chemical vapor deposition. Thin Solid Films 532, 89–93.

Yamada, T., Ishihara, M., Kim, J., Hasegawa, M., Iijima, S., 2012. A roll-to-roll microwave plasma chemical vapor deposition process for the production of 294 mm width graphene films at low temperature. Carbon 50 (7), 2615–2619.

Yoon, J., Sung, H., Lee, G., Cho, W., Ahn, N., Jung, H.S., Choi, M., 2017. Superflexible, high-efficiency perovskite solar cells utilizing graphene electrodes: towards future foldable power sources. Energy Environ. Sci. 10 (1), 337–345.

Yuan, K., Li, Q., Ni, W., Zhao, L., Wang, 2023. Graphene stabilized loess: mechanical properties, microstructural evolution and life cycle assessment. J. Clean. Prod. 389, 136081 https://doi.org/10.1016/j.jclepro.2023.136081.

Zhang, J.J., Fan, J.J., Cheng, B., Yu, J.G., Ho, W.K., 2020. Graphene-based materials in planar perovskite solar cells. Sol. RRL 4 (11).

Zhu, L., Li, Q., Chen, M., Cao, K., Sun, 2019. A simplified mathematical model for power output predicting of Building Integrated Photovoltaic under partial shading conditions. Energy Convers. Manag. 180, 831–843. https://doi.org/10.1016/j.enconman.2018.11.036.

Zweibel, K.J.E.p., 2010. Should solar photovoltaics be deployed sooner because of long operating life at low. predictable cost? 38 (11), 7519–7530.