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Comparative cradle-to-gate life cycle assessment of hydrothermal zinc oxide nanowire synthesis methods

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Zinc oxide nanowires (ZnO NWs) are promising materials for applications in sensors, transistors, and energy harvesting devices, owing to their unique structural and electronic properties. Despite advances in synthesis techniques, their environmental impacts remain an important consideration for sustainable nanomaterial development. In this study, we introduce a novel hydrothermal synthesis route inspired by Fehling's reaction, enabling the growth of ZnO NWs at low temperature and atmospheric pressure using bio-based and low-cost reagents such as glucose. To assess the environmental footprint of this novel method, a comparative life cycle assessment (LCA) methodology was employed using the OpenLCA software. The new route was benchmarked against a conventional sol–gel/chemical bath deposition synthesis which yields NWs of similar morphology. Results show that the Fehling-inspired method significantly reduces environmental impacts—by one to two orders of magnitude—across key categories such as climate change, ozone depletion, and human toxicity. In both methods, the silicon wafer substrate, electricity use, and hazardous waste treatment emerged as the dominant contributors to overall impacts, while chemical inputs had relatively minor effects, reinforcing the green chemistry potential of the proposed process. Sensitivity analyses explored several strategies for further impact reduction, including testing the influence of substrate materials, energy optimization, and regionalization. This work underscores the value of LCA as a tool for early-stage process evaluation and highlights practical opportunities for improving the sustainability of nanomaterial synthesis.

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1. This study quantitatively assesses through life-cycle assessment (LCA) the difference in environmental impacts between two hydrothermal zinc oxide nanowire (ZnO NW) synthesis methods and explores several pathways for emissions reductions through sensitivity analysis.
2. A novel in-solution ZnO NW growth method has fewer environmental impacts, including a 25% reduction in carbon emissions, as compared to the well-studied sol–gel/chemical bath deposition method. Emissions could be further reduced up to 75% by eliminating the need for a silicon wafer substrate and focusing on the development of a free-standing membrane.
3. Our paper focuses on exploring substrate-free methodologies to reduce the emissions from the silicon wafer, but a detailed analysis into the feasibility and potential carbon savings with substrate reuse or recycling is also interesting and relevant. Further research could also work to include the "use" and "end-of-life" life cycle stages for a complete analysis.

Introduction

The synthesis and application of nanomaterials are among the most transformative innovations of the 21st century, driving advancements in electronics,[1] energy,[2,3] healthcare,[4] and environmental technologies.[5] Among nanomaterials, zinc oxide (ZnO) stands out as a multifunctional material due to its non-toxicity, low cost, and natural abundance[6,7] coupled with its excellent electrical,[8] mechanical,[9] and optical properties.[10] As a wide bandgap (~3.37 eV) n-type semiconductor, ZnO is particularly well suited for diverse applications such as electron transport layer,[8,11] photocatalysis,[12] and sensors,[13] owing to its high surface activity and abundant oxygen vacancies.

In recent years, nanostructured ZnO architectures—including 0D nanoparticles, 1D nanowires (NWs), 2D nanosheets, and 3D hierarchical frameworks—have gained attention for their enhanced physicochemical properties and application-

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specific advantages.[14] Among these, ZnO NWs are especially promising for technologies such as gas sensing,[15] piezoelectric devices,[16] and photocatalysis[17] due to their high aspect ratio, large specific surface area, and crystallographic alignment.

The performance of ZnO NWs is strongly influenced by their synthesis method, which determines structural quality, defect density, and morphology.[6,18] Common low-temperature approaches include the sol–gel technique,[19] prized for its simplicity and scalability, and the hydrothermal method,[20] which yields higher crystalline quality and size control. Hybrid methods are also prevalent, where sol–gel-derived seed layers are combined with hydrothermal growth to guide ZnO NW formation.[21,22] However, despite advances in this field, sustainability concerns surrounding solvent use, energy input, and reagent toxicity remain largely underexplored in ZnO NW synthesis.[23–25]

In this context, the development and evaluation of low environmental impact approaches to ZnO NW synthesis are urgently needed. In our recent work,[26] we presented a simple hydrothermal method using Fehling's reaction—a classical aqueous redox system—for the controlled synthesis of cuprous oxide (Cu_2O) NWs under mild conditions using glucose as a bio-based and renewable reducing agent. Motivated by its operational simplicity and green profile, we adapted this method for ZnO NW synthesis. To the best of our knowledge, this represents an unexplored pathway, aside from a few studies involving Cu_2O–ZnO hybrid nanostructures.[27]

To assess the environmental relevance of this novel synthesis route, we used the Life Cycle Assessment (LCA) tool, a comprehensive and science-based methodology that evaluates environmental impacts across the entire production chain, from raw material extraction to final product formation (*cradle-to-gate*).[28,29] LCA has become increasingly vital in nanomanufacturing, providing robust metrics on energy consumption, greenhouse gas emissions, water usage, and human and ecological toxicity. This is particularly critical in light of the fact that the global materials sector accounts for ~23% of anthropogenic carbon dioxide emissions,[30] and nanomaterials production is projected to grow exponentially through 2050.[31]

In this study, we conduct a comparative LCA of two ZnO NW synthesis routes:

(i) the "conventional" sol–gel/chemical bath deposition (CBD) method, and

(ii) a novel Fehling-based in-solution growth method.

By quantifying material and energy flows for each, this work identifies key environmental hotspots and establishes the sustainability trade-offs between conventional and alternative synthesis routes.

Experimental methodology

ZnO NW synthesis *via* sol–gel/chemical bath deposition

ZnO NW synthesis *via* the sol–gel/CBD approach is a widely used and well-studied two-step process. First, a ZnO sol–gel is prepared based on a previously published protocol by vigorously mixing zinc acetate dihydrate (ZAD), ethanolamine, and 1-butanol for 3 hours at room temperature.[32] In this mixture, ethanolamine is used to aid the dilution of zinc in a liquid solvent and provide the necessary basic pH conditions. 1-Butanol is used as a solvent due to its high boiling point, high dielectric constant, and non-toxicity. Once fully mixed, 300 μL of the formed sol–gel is deposited onto a clean silicon wafer substrate *via* spincoating. The sample is then annealed at 540 °C for one hour, leading to the crystallization of the ZnO seed layer.

Once the sol–gel has been deposited and the seed layer has been crystallized, ZnO NWs are synthesized using the CBD method. A 50 mM aqueous solution of zinc nitrate hexahydrate (ZNH) and hexamethylenetetramine (HMTA) is mixed. ZNH provides the zinc salt and HMTA is the alkaline reagent. In this chemical reaction, the HMTA undergoes a decomposition reaction in water to produce ammonia, which then undergoes a hydroxyl supply reaction to form hydroxide ions. These free hydroxide ions then react with the zinc salts from the ZNH to form the ZnO NWs.[19] The mixture is heated to 90 °C at which time, the silicon wafer substrate with the ZnO seed layer is affixed to a sample holder at a 45° angle and placed facing down in the solution for a given duration. As the growth proceeds, the NW length increases proportionally to the growth duration until the precursors are depleted, which takes roughly 3 hours.[33] In this study, we consider ZnO NWs which undergo a 3-hour growth duration. Scanning electron microscopy (SEM) images of the synthesized ZnO NWs are provided in SI (Fig. S1).

ZnO NW synthesis inspired by Fehling's reaction

ZnO NW synthesis in-solution inspired by Fehling's reaction begins with the preparation of three distinct stock solutions:

Metal oxide precursor solution. A 0.3 mM aqueous solution of ZNH (Aldrich) was prepared as the ZnO precursor. ZNH was chosen for its high solubility and ability to provide a consistent supply of zinc ions during the reaction, which is essential for the controlled growth of ZnO nanostructures.[26]

Alkaline buffer solution. A second solution was prepared by dissolving sodium hydroxide (14 mM, Aldrich) and sodium tartrate dibasic dihydrate (8 mM, Aldrich) in deionized water. Sodium hydroxide serves as a strong base to create the basic pH conditions necessary for ZnO formation, while sodium tartrate dibasic dihydrate acts as a chelating agent to regulate the release of zinc ions, preventing premature precipitation and ensuring uniform particle growth.[26]

Reducing agent solution. The third solution consists of D(+)-glucose dissolved in deionized water. While D(+)-glucose functions as the reducing agent in the classical Fehling's reaction, its specific role in the nucleation and growth of ZnO NWs remains unclear and is still under investigation in our group. However, it should be noted that this is outside the scope of this study and does not impact the results below.

Fehling reaction procedure. For the solution-phase synthesis *via* Fehling's reaction, 10 mL of the ZNH solution is combined with 10 mL of the alkaline buffer solution. Next, 20 mL of the

D(+)-glucose solution is added and the resulting solution is transferred to a 50 mL glass bottle and sealed. The bottle is then placed in an oven at 95 °C for 90 minutes, during which time the reaction takes place and ZnO NWs are formed directly in solution.

Transfer on Si wafer. The ZnO NW suspension is first vacuum-filtered onto a nitrocellulose membrane (Merck Millipore, mixed cellulose esters (MCE) membrane, 0.1 μm pore size & 47 mm diameter). The NWs are then subsequently transferred onto a silicon wafer substrate *via* a wet process involving 1,4-dichlorobenzene, followed by dissolution of the membrane in acetone.[34] SEM images of the synthesized NWs are provided in SI (Fig. S1).

Life cycle assessment

LCA is a comprehensive analysis methodology to assess the environmental impact of a product or production system. It provides insights into the life cycle stages and specific inputs which have the greatest environmental impact, allowing the mitigation of environmental risks through process or product optimization. According to ISO Standard ISO 14044:2006, performing a comprehensive LCA involves four major steps, as shown in Fig. 1: (1) defining goal and scope, (2) inventory analysis, (3) impact assessment, and (4) interpretation.[35,36]

Goal and scope. The first step in conducting an LCA is defining the goal and scope of the study, which includes determining the purpose, system boundaries, functional unit, and limitations of the study.

The goal of this study is to compare the environmental impacts of two ZnO NW synthesis methods: (1) sol–gel/CBD, and (2) in-solution growth inspired by Fehling's reaction. Further, this study will identify the key contributors to environmental concerns of each synthesis method, providing a foundation for future research to target these critical areas and develop strategies that effectively minimize environmental impacts.

The functional unit for this study is 2 mg of ZnO NWs deposited onto a silicon wafer substrate with an area of 9 cm^2. The mass of NWs produced *via* each methodology was determined both experimentally and estimated using literature references from the research team. A full discussion of this process is provided in SI (section S1). While the ZnO NWs produced *via* the sol–gel/CBD route exhibit a vertically aligned architecture (perpendicular to the substrate surface) and those from the in-solution Fehling-inspired method form a horizontal nanonet structure (parallel to the substrate surface), both synthesis approaches yield NWs of comparable morphology, composition, and functional purpose. Further, existing literature shows that while NW orientation can affect device integration, the characteristics and performance of the NWs remain comparable. For example, Tsivion *et al.* demonstrated that horizontally grown ZnO NWs exhibit properties comparable to vertically oriented NWs.[37] Similarly, Nikoobakht *et al.* showed that horizontally oriented ZnO NWs perform comparably to vertical NWs in terms of optical and electrical characteristics.[38] As such, the difference in orientation does not inhibit a meaningful life cycle comparison since the LCA is normalized to the mass of functional material rather than its structural form.

This is a cradle-to-gate LCA, meaning the assessment considers the environmental impacts from raw material extraction up to the point at which the final product (*i.e.*, a silicon wafer with ZnO NWs on one face) has been fabricated at the laboratory scale. The use phase and final disposal of the ZnO NWs are out of scope for this study since the focus of this paper is the impacts of the synthesis methods and not the NWs themselves.

Limitations and uncertainty. This study presents a cradle-to-gate LCA based on laboratory-scale synthesis routes, which introduces several inherent limitations. Laboratory processes often rely on small-scale setups, research-grade substances, and non-optimized energy use, which do not necessarily reflect the efficiencies of large-scale manufacturing. Further, energy consumption data were derived from laboratory equipment usage, which can differ substantially from industrial energy profiles. Additionally, not all chemicals and nanomaterial-specific inputs used in this study were readily available in the considered LCA database. In such cases, approximations were made based on the best available information, which may not fully capture the environmental profile of the actual materials. Although efforts were made to account for these limitations, uncertainties remain in the resulting impact assessment.

To account for these uncertainties and ensure a robust comparison of the environmental performance of the two ZnO NW synthesis methods, uncertainty and data quality were explicitly characterized and propagated through the model using a Monte Carlo simulation. Uncertainty was characterized directly within OpenLCA at the flow level by assigning lognormal distributions. Data quality indicators were incorporated at the flow level following the EcoInvent data quality system matrix to capture variability arising from data reliability and geographical representativeness. These uncertainty and data quality parameters were propagated through the life cycle model using a Monte Carlo simulation with 1000 iterations. The number of iterations was deemed sufficient based on a

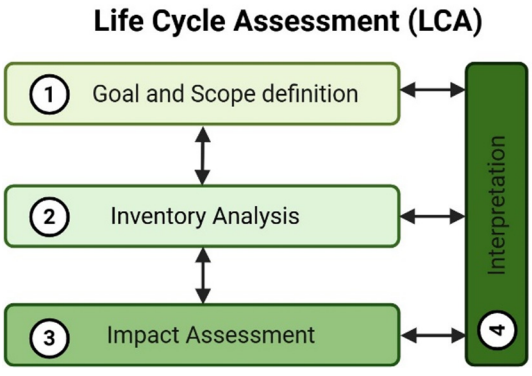

Fig. 1 The four stages of a life cycle assessment.

convergence analysis, which showed that the mean, standard deviation, and 5th and 95th percentile values for 15 of the 16 impact categories changed by less than 2% beyond 500 iterations, indicating statistical stability and robustness of the results.

Inventory analysis. The next step is to create and analyze the inventory of all material and energy inputs. There are two kinds of data used for the inventory analysis: (1) primary data, and (2) secondary data. The primary data are measurements taken in the lab including the quantity of chemicals used, machine specifications, and electricity requirements of equipment. The secondary data are collected from literature.

OpenLCA is a free, open-source software widely used for conducting high-quality, professional LCAs. For this study, the EcoInvent 3.10 database was used to source data on material and energy inputs associated with the NW synthesis process. However, certain chemicals—such as ZNH, ZAD, sodium tartrate dibasic dihydrate, and HMTA—were not available in the database. To address this limitation, literature sources (primarily industrial patents) were consulted to reconstruct synthesis routes for these compounds. Using the main chemical reactions reported, stoichiometric relationships were used to estimate the mass of precursor or base chemicals required to produce one mole of the target compounds. These precursor chemicals, which are present in the EcoInvent database, were then modelled in OpenLCA to approximate the environmental impacts of the unavailable substances. The breakdown of each chemical's synthesis route is shown in Fig. 2, and full details of the reaction pathways, mass calculations, and supporting literature references are provided in SI (section S2). The selected geographic provider for energy data is France, as this is where the experiments were carried out, while material data uses the geographic provider RER (representing Europe in the EcoInvent database); where RER data was unavailable, the global (GLO) provider was used as an alternative.

Inventory of sol–gel/CBD synthesis method. As described above, the process of synthesizing ZnO NWs *via* the sol–gel/CBD approach involves first fabricating a ZnO sol–gel, then spreading this sol–gel on a clean silicon wafer, and finally growing the NWs directly on the substrate in a chemical bath. The full list of material and energy inputs that make up the sol–gel/CBD synthesis method is outlined in Table 1. The energy consumption data presented here for laboratory equipment was estimated from the nominal power rating of each device and its operating time, following established practice for laboratory-scale LCIs.[39,40] This proxy method, while conservative, is widely applied when direct metering data are unavailable and is consistent with recommended approaches for early-stage chemical process assessment. Resulting uncertainty from this estimation method is considered in the Monte Carlo analysis presented in Results and Discussion. As mentioned previously, the breakdown of chemicals ZAD, ZNH, and HMTA into their base components can be found in SI. Finally, the residual growth solution remaining after synthesis was modelled as a hazardous waste stream to account for potential unreacted precursors and nanomaterials. A representative hazardous waste treatment process from the EcoInvent database was used as an approximation to capture this environmental burden and provide a reasonable representation of this waste within the system boundary.

Inventory of in-solution growth inspired by Fehling's reaction. The preparation of ZnO NWs *via* in-solution growth inspired by Fehling's reaction involves first fabricating three mother solutions: (1) metal oxide precursor, (2) alkaline buffer, and (C) reducing agent. These solutions are then mixed and placed in an oven at 95 °C for 90 minutes. At this time, the solution is vacuum filtrated and the NWs are transferred to a silicon wafer substrate. For the energy consumption data, the same estimation methodology was employed as described above. Additionally, a hazardous waste stream is considered in the output of this process to account for potential unreacted precursors and nanomaterials in the leftover growth solution that is thrown away. The full list of material and energy inputs that make up this process is outlined in Table 2.

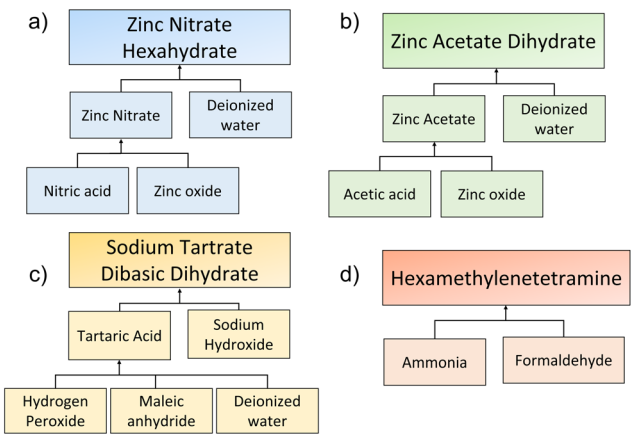

Fig. 2 Chemical break-down of (a) zinc nitrate hexahydrate, (b) zinc acetate dihydrate, (c) sodium tartrate dibasic dihydrate, and (d) hexamethylenetetramine into their base components.

Results and discussion

Impact assessment

The obtained life cycle inventories were assessed using the Environmental Footprint-EF 3.1 method of impact assessment for LCA. This method was selected because it was developed by the Joint Research Centre of the European Commission and as such, it is widely used for quantifying the environmental impacts in the European Union. Further, it is a midpoint-focused impact assessment method, which is recommended for use in scientific publications as it provides higher confidence and lower uncertainty as compared to endpoint-focused methods. In the EF 3.1 method, there are 16 assessed impact categories: (1) acidification (AC), (2) climate change (CC), (3) ecotoxicity-freshwater (ET-F), (4) energy resources: non-renewable (ER-NR), (5) eutrophication: freshwater (EU-F), (6) eutrophication: marine (EU-M), (7) eutrophication: terrestrial

Table 1 Inventory for sol–gel/CBD growth route

Input flow	Amount	Unit	Provider	Description
Electricity, low voltage	0.05	kWh	Market for electricity, low voltage | electricity, low voltage | cutoff, S – FR	Electricity to stir sol–gel for 3 hours at ambient temperature
Electricity, low voltage	1.20	kWh	Market for electricity, low voltage | electricity, low voltage | cutoff, S – FR	Electricity to anneal the seed layer for 1 hour at 540 °C
Electricity, low voltage	3.06	kWh	Market for electricity, low voltage | electricity, low voltage | cutoff, S – FR	Electricity for 3-hour NW growth on a hotplate at 90 °C
Water, deionized	190	g	Market for water, deionized | water, deionized | cutoff, S – Europe without Switzerland	Growth solution
Zinc nitrate hexahydrate	5.00	g	See SI	Growth solution
HMTA	5.00	g	See SI	Growth solution
Zinc acetate dihydrate	0.07	g	See SI	Sol–gel fabrication
1-Butanol	0.24	g	Market for 1-butanol | 1-butanol | cutoff, S – GLO	Sol–gel fabrication
Monoethanolamine	0.02	g	Market for monoethanolamine | monoethanolamine | cutoff, S – GLO	Sol–gel fabrication
Single-Si wafer, for electronics	9.00	cm^2	Market for single-Si wafer, for electronics | single-Si wafer, for electronics | cutoff, U – GLO	Substrate

Output flow	Amount	Unit	Provider	Description
Hazardous waste for incineration	200	mL	Market for hazardous waste, for incineration | hazardous waste, for incineration | cutoff, S – Europe without Switzerland	Leftover growth solution

Table 2 Inventory for in-solution growth route inspired by Fehling's reaction

Input flow	Amount	Unit	Provider	Description
Electricity, low voltage	1.06	kWh	Market for electricity, low voltage | electricity, low voltage | cutoff, S – FR	Electricity used during NW growth to power oven at 95 °C for 90-minutes
Electricity, low voltage	0.40	kWh	Market for electricity, low voltage | electricity, low voltage | cutoff, S – FR	Electricity used to power pump during NW filtration
Water, deionized	53.3	g	Market for water, deionized | water, deionized | cutoff, S – Europe without Switzerland	Sum of deionized water used to prepare the Fehling A, Fehling B, and glucose solutions
Sodium tartrate dibasic dihydrate	0.005	g	See SI	Fehling B
Glucose	0.00	g	Market for glucose | glucose | cutoff, S – GLO	Reducing agent
Zinc nitrate hexahydrate	0.001	g	See SI	Fehling A
Sodium hydroxide	0.03	g	Market for sodium hydroxide, without water, in 50% solution state | sodium hydroxide, without water, in 50% solution state | cutoff, S – RER	Fehling B
Nitrocellulose filter	1.00	g	Market for cellulose fibre | cellulose fibre | cutoff, U – RoW	Used during NW filtration
Acetone	15.6	g	Market for acetone, liquid | acetone, liquid | cutoff, S – RER	Used to transfer NWs to substrate
o-Dichlorobenzene	1.00	g	Market for *o*-dichlorobenzene | *o*-dichlorobenzene | cutoff, S – RoW	Used to transfer NWs to substrate
Single-Si wafer, for electronics	9.00	cm^2	Market for single-Si wafer, for electronics | single-Si wafer, for electronics | cutoff, U – GLO	Substrate

Output flow	Amount	Unit	Provider	Description
Hazardous waste for incineration	200	mL	Market for hazardous waste, for incineration | hazardous waste, for incineration | cutoff, S – Europe without Switzerland	Leftover reaction solution

(EU-T), (8) human toxicity: carcinogenic (HT-C), (9) human toxicity: non-carcinogenic (HT-NC), (10) ionizing radiation: human health (IR-HH), (11) land use (LU), (12) materials resources: metals/minerals (MR-MM), (13) ozone depletion (OD), (14) particulate matter formation (PMF), (15) photochemical oxidant formation: human health (PCF), and (16) water use (WU).

Interpretation

Comparative LCA. In this study, LCA methodology is first employed to understand the differences in environmental impacts between the two studied ZnO NW synthesis methods. Table 3 below shows the raw data output comparing the sol–gel/CBD process to the in-solution process inspired by Fehling's reaction for each of the 16 impact categories. We find that the sol–gel/CBD route consistently exhibits the highest environmental impact across all assessed categories, indicating that this synthesis method is overall less environmentally favorable compared to the in-solution route. This is seen graphically in Fig. 3, in which a bar chart compares the relative impacts from each methodology. In this graph, the highest impact for each category is set to 100% and the result of the other method is displayed in relation to this result. The error bars represent the relative standard deviation for each data point, determined using a Monte Carlo simulation performed in OpenLCA. The statistical analysis indicates that the difference in environmental performance between the two synthesis routes is statistically significant for 14 of the 16 assessed impact categories, demonstrating that the observed differences remain significant even when underlying data uncertainty is considered. The only categories for which the difference was not statistically significant are freshwater eutrophication and water use.

A closer examination of individual impact categories reveals varying degrees of difference between the two methods. For instance, in the water use category, the Fehling-inspired route results in approximately 18% fewer impacts compared to the sol–gel/CBD process, suggesting relatively similar performance. Further, the Monte Carlo analysis suggests that this difference is within the standard deviation, meaning it is not statistically significant. In contrast, categories such as ionizing radiation and material resources (metals/minerals) show much larger disparities, with the in-solution route generating only about 40% of the impacts seen in the sol–gel/CBD route, highlighting more significant environmental advantages in those areas. To understand further why some impact categories show much larger differences than others, we can perform a contribution analysis.

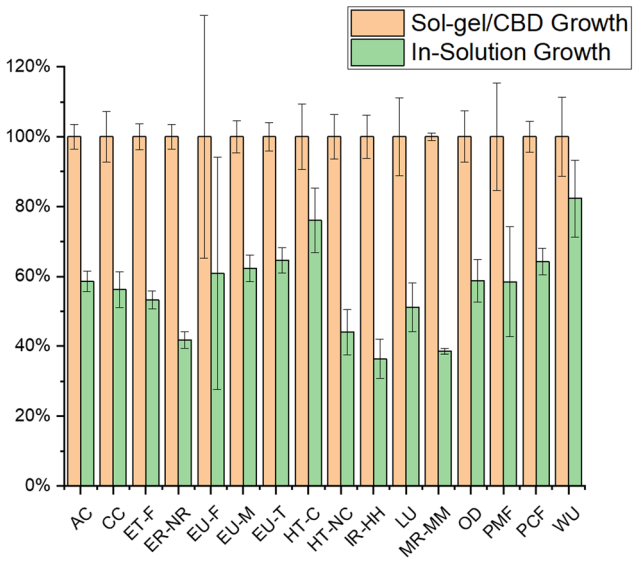

Fig. 3 Comparative life cycle impact analysis of ZnO NW growth *via* the sol–gel/CBD approach and the in-solution method inspired by Fehling's reaction.

Table 3 Comparative LCA results

Impact category	Unit	Sol–gel/CBD route	In-solution route
AC	mol H^+ eq.	4.59×10^{-3}	2.69×10^{-3}
CC	kg CO_2 eq.	1.19	0.667
ET-f	CTUe	11.8	6.28
ER-NR	MJ, net calorific value	54.6	22.8
EU-F	kg P eq.	4.60×10^{-4}	2.80×10^{-4}
EU-M	kg N eq.	1.01×10^{-3}	6.30×10^{-4}
EU-T	mol N eq.	9.28×10^{-3}	6.00×10^{-3}
HT-C	CTUh	4.80×10^{-9}	3.65×10^{-9}
HT-NC	CTUh	1.93×10^{-8}	8.53×10^{-9}
IR-HH	kBq U-235 eq.	2.19	0.799
LU	Dimensionless	4.23	2.17
MR-MM	kg Sb eq.	1.96×10^{-5}	7.57×10^{-6}
OD	kg CFC-11 eq.	2.56×10^{-8}	1.50×10^{-8}
PMF	Disease incidence	4.31×10^{-8}	2.52×10^{-8}
POF	kg NMVOC eq.	3.11×10^{-3}	2.00×10^{-3}
WU	m^3 world eq. deprived	2.18	1.79

Contribution analysis. A contribution analysis in LCA identifies the main drivers of environmental impacts for each impact category. This analysis is essential in any LCA, as it helps prioritize areas for impact reduction by highlighting the most significant contributors. Fig. 4a shows the top contributing factors for the sol–gel/CBD method. The data shows that over 99% of the impacts come from just three inputs across all 16 impact categories: (1) the silicon wafer substrate, (2) electricity consumption, and (3) the treatment of hazardous waste. Electricity consumption is the single largest contributor in most categories (12 of 16). A similar pattern emerges in the contribution analysis for the in-solution NW growth method, shown in Fig. 4b. The results demonstrate that the environmental footprint is once again dominated by impacts from the silicon wafer substrate, electricity consumption, and the treatment of hazardous waste, with minor impacts stemming from the use of acetone and dichlorobenzene. Specifically, the silicon wafer is the largest contributor in 10 of the 16 categories, while electricity has a greater influence in the categories of energy resources (non-renewable), human toxicity (non-carcinogenic), ionizing radiation, land use, and material resources (metals/minerals).

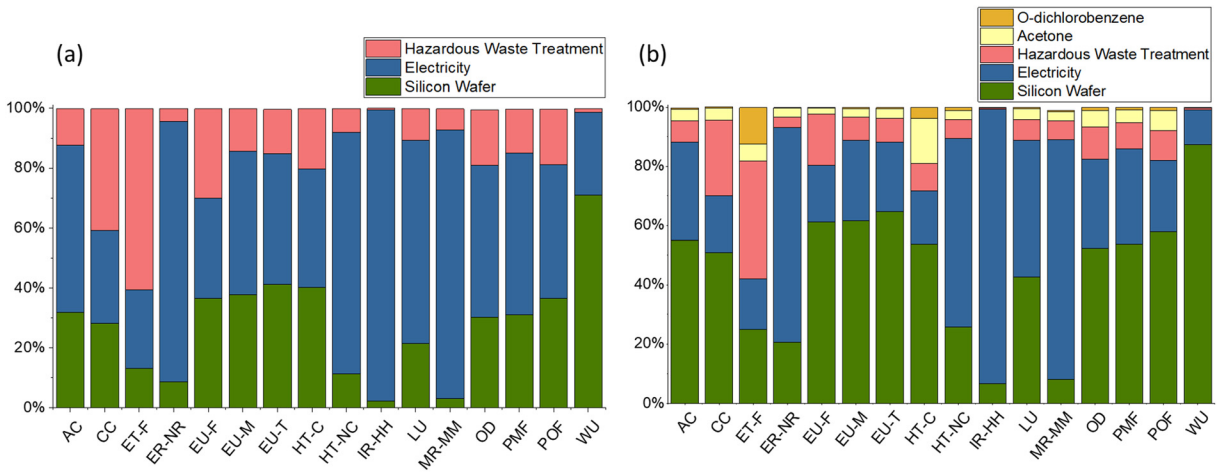

Fig. 4 Contribution tree of ZnO NW synthesis for the (a) sol–gel/CBD methodology, and (b) in-solution methodology inspired by Fehling's reaction.

Both methodologies show that the largest contributor to the impact category of water use is the silicon wafer substrate, as this dominates over 75% of the impacts. This result helps to explain why this impact category does not see a statistically significant change between the two categories when looking at the comparative LCA, as an equal area of silicon wafer is used in both methodologies. The categories in which we see the largest difference in environmental performance between the two synthesis methods (ionizing radiation and material resources (metals/minerals)) are the categories in which we see the largest impacts coming from electricity consumption. This shows that the major environmental advantage of the in-solution growth as compared to the sol–gel/CBD route is the reduced energy consumption. The increased role of electricity in the sol–gel/CBD route likely stems from the energy-intensive steps specific to this process, such as seed layer annealing and temperature-controlled growth conditions.

Overall, these results show, perhaps surprisingly, that the environmental impacts of ZnO NW synthesis are not primarily driven by the chemical precursors. This is explained given the relatively small mass of NWs synthesized compared to the bulk material and energy inputs. These results suggest that improvements in substrate selection, energy efficiency, and recycling of hazardous waste represent the most promising avenues for reducing the environmental burdens of this process. These insights not only highlight the need for greener substrate alternatives or reuse strategies but also stress the importance of optimizing process parameters to minimize electricity use.

Sensitivity analysis: effect of the substrate. In order to most efficiently reduce the environmental footprint of ZnO NW synthesis, we begin by addressing the largest contributors. To do this, a sensitivity analysis is performed. A sensitivity analysis consists of changing one parameter in the system to observe how the results change based on the effect of this sole parameter.

The silicon wafer substrate was found to be a major contributor to environmental impacts for both types of ZnO NW synthesis methods. Silicon wafers are commonly used for lab-scale experimentation due to their thermal and chemical robustness, making them favorable to work with in a wide variety of experimental conditions. This study, however, shows the dangers of depending on such a material for the development of ZnO NW arrays. While other options for substrates do exist, such as wafers based on gallium or germanium, it has been proven in literature that such substrates have an even higher environmental footprint than silicon wafers.[41] As a result, it is interesting to explore the possibility of developing ZnO NW arrays in the form of free-standing membranes, eliminating the need for environmentally costly substrates all together.

While a substrate-free NW membrane has not yet been developed experimentally with ZnO, we have successfully developed free-standing membranes using Cu_2O NWs synthesized in-solution *via* Fehling's reaction. In this sensitivity analysis, we assume that the same process for purifying and filtering the NWs can be applied to ZnO NWs. We note, however, that this assumption represents a simplification, as differences in crystal growth, morphology, and processing behavior between ZnO and Cu_2O may affect the feasibility of developing substrate-free membranes. Therefore, the results of this exploratory analysis are intended to illustrate the potential and promise of this research pathway, and should be interpreted as preliminary estimates while experimental validation is needed in future work.

To develop such a free-standing membrane, after ZnO NW synthesis in-solution, the solution with suspended NWs is centrifuged and cleaned with deionized water and ethanol. The centrifuged and purified solution is then vacuum filtrated. The NWs collected on the filter paper are then dried in ambient conditions. After a period of 1 hour, the membrane is dry and can be removed from the filter paper with tweezers. In eliminating the need for a silicon wafer, more electricity is con-

sumed in the process during centrifugation and filtration, so it is essential to compare across all impact assessment categories to watch for shifting impacts.

The results from this analysis are shown in Fig. 5 and compare the in-solution growth route both with and without a wafer. As we can see, the process of making a free-standing membrane is much more environmentally friendly than the process involving a wafer, seeing drastic decreases in almost every category including climate change and water use. The only impact categories where the no-wafer process exhibits higher impacts than the base scenario are ionizing radiation (human health) and material resources (metals/minerals). This outcome is consistent with the contribution analysis, which identified electricity consumption as the dominant contributor to these categories. Because the substrate-free membrane synthesis requires greater electricity use, these impact categories are consequently more affected. The largest differences seen are in the categories in which the impacts were dominated by the use of the silicon wafer. For example, we saw in the base scenario 87% of the impacts in the water use category came from the silicon wafer substrate. By removing this substrate, the water use category sees an 84% decrease in impacts.

These results highlight the importance of exploring alternative and potentially unconventional research methodologies. The development of a free-standing ZnO NW membrane is interesting for future research and a promising candidate for an environmentally sustainable product. As shown in our results, exploring such a pathway could reduce carbon emissions by up to 55% as compared to the original base scenario. Further work in reducing the effect of the silicon wafer substrate could explore the possibilities of reuse or recycling.

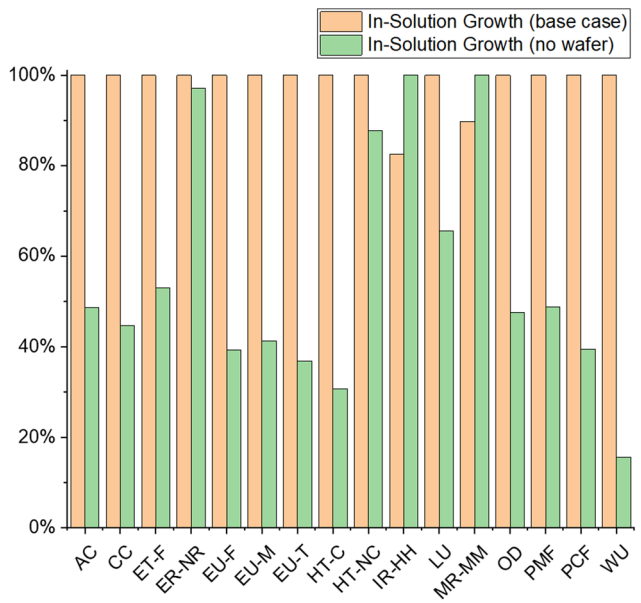

Fig. 5 Sensitivity analysis showing reduction of impacts in the development of a free-standing membrane in comparison to the base scenario.

Sensitivity analysis: electricity consumption. The contribution analysis also identified electricity consumption as a top contributor for both ZnO NW synthesis methods. To this end, a sensitivity analysis is performed to evaluate the reduction in environmental harm through lowering energy consumption. In the sol–gel/CBD route, electricity is consumed during three key stages: sol–gel stirring, seed layer annealing, and NW growth. The durations of the stirring and growth steps have already been optimized for performance and cannot be further reduced without compromising NW quality; however, for the seed layer annealing step, we tested a shorter annealing time of 15 minutes at 540 °C instead of the standard 1 hour. Our analysis confirmed that the seed layer retained its desired structure under the shortened condition, prompting an investigation into how this reduction could influence the process's environmental footprint.

The sensitivity analysis showed that this change led to mild improvements across all impact assessment categories, as outlined below in Table 4. Ionizing radiation decreased by 19%, material resources (metals/minerals) by 18%, and human toxicity (non-carcinogenic) by 16%, highlighting the sensitivity of these categories to electricity use. These results show that minimizing electricity demand, particularly in thermally intensive steps like annealing, can help improve environmental performance. However, the persistence of significant impacts emphasizes that reducing electricity use alone is insufficient to minimize the environmental harm of ZnO NW synthesis; such efforts should be pursued in tandem with other strategies, such as investigating substrate-free synthesis, to achieve greater environmental benefit.

Electricity consumption was also identified as a key contributor in the in-solution growth route. This process has already been optimized for energy efficiency and as such, further reductions in electricity use are currently not feasible without compromising NW quality. Nonetheless, examining the influence of electricity sourcing remains valuable. To this end, a regionalization analysis was conducted. The French electricity grid was used in the base scenario, reflecting the actual conditions of NW growth in our laboratory. However, since semiconductor manufacturing is predominantly concentrated in

Table 4 Results of sensitivity analysis for annealing time with sol–gel/CBD route

Impact assessment category	Unit	1-hour anneal	15 min anneal	Decrease (%)
AC	mol H^+ eq.	4.59×10^{-3}	4.08×10^{-3}	11
CC	kg CO_2 eq.	1.19	1.11	6
HT-C	CTUh	4.80×10^{-9}	4.42×10^{-9}	8
HT-NC	CTUh	1.93×10^{-8}	1.62×10^{-8}	16
IR-HH	kBq U235 eq.	2.19	1.77	19
LU	Dimensionless	4.23	3.66	14
MR-MM	kg Sb eq.	1.96×10^{-5}	1.61×10^{-5}	18
OD	kg CFC-11 eq.	2.56×10^{-8}	2.30×10^{-8}	10
PMF	Disease incidence	4.31×10^{-8}	3.85×10^{-8}	11
POF	kg NMVOC eq.	3.11×10^{-3}	2.83×10^{-3}	9
WU	m^3 world eq. deprived	2.18	2.06	6

the United States, Taiwan, and China, alternative electricity mixes from these regions were considered to assess their impact on the LCA results.

The results of this regionalization analysis for the in-solution growth route, shown in Fig. 6, demonstrate that the geographic location of the NW synthesis process plays a large role in the resulting environmental impacts. This analysis was conducted for all 16 of the assessed impact assessment categories, but only a selection is shown graphically for clarity. Of the 16 assessed categories, China results in the highest environmental impacts in 10, including the climate change category in which we observe a 243% increase in emissions as compared to France. Keeping ZnO NW production in France results in the highest emissions in the energy resources (nonrenewable), ionizing radiation, material resources (metals/minerals), and water use categories. This result is logical given the large percentage of nuclear power used in the French energy grid as compared to the other assessed countries. Looking at the ionizing radiation category as an example, switching production to China, Taiwan, or the USA could result in 90%, 69%, and 76% fewer emissions, respectively. Using the Taiwanese energy grid resulted in the largest impacts in the freshwater eutrophication and ozone depletion categories, likely due to the country's dependency on fossil fuels and need to import a large portion of its energy resources.

These results also highlight the difficulty in lowering the overall environmental footprint of a product due to the shifting of impacts from one category to another. For example, producing these ZnO NWs in the USA is much better for the climate change category than production in China, resulting in 45% fewer emissions. By moving production to the USA, however, the impacts are increased in the categories of freshwater eutrophication (+39%) and ionizing radiation: human health (+154%). This shifting of impacts highlights the importance of testing environmental impacts across a broad range of impact categories to avoid decreasing impacts in one category at the expense of another. Taking these shifting impacts into consideration is important for understanding the overall footprint of a product.

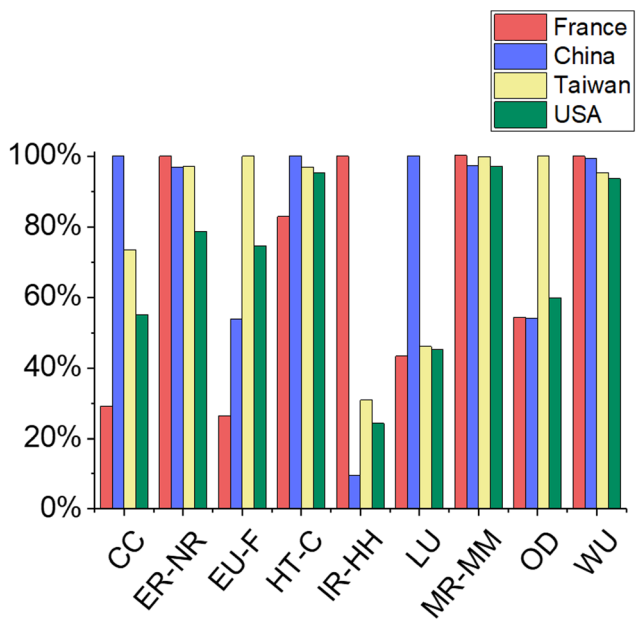

Fig. 6 Comparative impacts of in-solution growth inspired by Fehling's reaction with change of geography in France, China, Taiwan, and the USA for select impact assessment categories.

Conclusions

This study presents a comparative LCA of two hydrothermal synthesis routes for producing ZnO NWs: a conventional sol–gel/CBD method and a novel in-solution growth inspired by Fehling's reaction. Across all assessed environmental impact categories, the novel Fehling-inspired method demonstrated superior performance, including a notable 44% reduction in carbon emissions. A deeper analysis identified the silicon wafer substrate, electricity consumption, and hazardous waste treatment as the primary environmental hotspots in both synthesis routes. To address these contributors, we first investigated the potential environmental benefits of developing a free-standing ZnO NW membrane, which would eliminate the need for a silicon wafer substrate entirely, and showed that carbon reductions of up to 55% are possible. Further, we evaluated the environmental benefits of reducing the sol–gel annealing time to lower energy demand and conducted a regionalization analysis to assess the influence of different electricity grid mixes.

This study offers insight into the environmental implications of ZnO NW synthesis, an area that remains largely underexplored despite the projected surge in nanomaterials production through 2050. By identifying key environmental hotspots and proposing feasible mitigation strategies, this work lays the groundwork for more sustainable ZnO NW fabrication moving forward. Looking ahead, future research should aim to expand the system boundary to include the use phase and end-of-life treatment, while also exploring reuse or recycling opportunities for both the silicon wafer and unreacted precursors to further enhance the sustainability of ZnO NW production. As ZnO NW synthesis transitions from laboratory- to industrial-scale production, building more comprehensive life cycle inventories that incorporate measured waste streams, emissions, and energy data will be essential to accurately reflect the true environmental performance of these processes.

Author contributions

Jamie Silk: validation, formal analysis, investigation, data curation, writing – original draft, visualization. Soline Beitone: validation, formal analysis, investigation, writing – original draft, visualization. Mayrazul Hoque: formal analysis, investigation, writing – review & editing. Céline Ternon: conceptualization, writing – review & editing, supervision. Damien Evrard: con-

ceptualization, methodology, validation, resources, data curation, writing – review & editing, supervision, project administration, funding acquisition. David Riassetto: conceptualization, methodology, resources, writing – review & editing, supervision, project administration, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the supplementary information (SI). Supplementary information: supplementary data 1: raw data from OpenLCA (comparative LCA of base scenario, sensitivity analysis of seed layer anneal time, sensitivity analysis of regionalization, sensitivity analysis of removing silicon wafer); supplementary data 2: contribution analysis raw data – sol–gel/CBD route; supplementary data 3: contribution analysis raw data – Fehling in-solution route; supplementary data 4: Monte Carlo analysis raw data – sol–gel/CBD route; supplementary data 5: Monte Carlo analysis raw data – Fehling in-solution route. See DOI: https://doi.org/10.1039/d5gc03866a.

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