Life cycle assessment of bio-based hard carbon for sodium-ion batteries across different production scales

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ABSTRACT

This paper aims to address research gaps surrounding the environmental impact of Hard Carbon (HC) production by conducting a Life Cycle Assessment (LCA) based on data from two laboratories with differing backgrounds and scenarios. HC is commonly used as anode material for sodium-ion batteries, a potentially sustainable and cost-efficient alternative for lithium-ion batteries. The study identifies environmentally sustainable routes for HC synthesis by comparing various biomass and synthesis pathways. The study reveals that the energy consumption of the pyrolysis process is the primary contributor to the environmental footprint of lab-scale HC production. A prospective LCA is performed by upscaling the laboratory processes to pilot- and industrial scale based on expert judgement and assumptions on energy and material balance. The results show that the environmental profile of HC can be significantly improved when the production scale is expanded. At large production scales, HC shows great potential to be used as a counterpart to graphite in future battery systems. However, direct emissions, such as methane, and the depletion of materials, such as argon and acid, become more critical to the environmental footprint, highlighting the need for energy recovery, emission treatment strategies, and more efficient use of materials. This work provides a framework for future LCA studies of HC, highlighting the limitations of simplified upscaling. It also provides a foundation for developing sustainable energy storage systems, thereby contributing to more informed decision-making in HC industrial production.

1. Introduction

Batteries are considered to be a key technology for the energy and mobility transition, with a high share of renewable energy provision. [1] Lithium-ion batteries (LIBs) are currently the most commonly used energy storage technology, but face many problems in respect of sustainability aspects, such as the usage of critical materials from politically unstable countries, and related supply risk, material availability, and cost issue. [2] Sodium-ion batteries (SIBs) have been proposed as a promising alternative to LIBs in many applications, such as stationary storage and electric vehicles. [3,4] SIBs share the same cell structure

with LIBs, with main components of anode, cathode, separator, electrolyte, etc. However, SIBs have the important advantage of not using critical raw materials in those components such as cobalt, nickel and lithium in cathode, and graphite in anode. Therefore, SIBs have potential to alleviate the strain of environmental impacts [5,6], supply risks and costs [7,8].

However, the commonly used graphite anode from LIBs cannot be combined stably with the sodium-ion, which makes this anode type unsuitable for SIBs. [5] Hard Carbon (HC), a non-graphitizable carbonaceous material, emerges as a viable anode material for SIBs, due to its good balance between performance, cost and sustainability and huge

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potential for large-scale commercialization. [5,9] HC is now commonly used in various commercial prototype SIBs [10].

HC can be derived from fossil-based precursors such as resin or petrol coke, but also from various types of biomass. Bio-based HC offers a supply-risk-free alternative. Despite HĆs huge potential, there is a lack of comparative sustainability studies between different bio-waste precursors and processing routes, which hinders the identification of key factors for minimizing the environmental burden of HC production. Furthermore, a study considering different scales and throughput was not offered by previous work, due to lack of data availability. At the stage prior to large-scale commercialization, it is urgent to fill the data gap and crucial to conduct a holistic study that can identify the drivers in the production chain of HC.

Addressing these gaps, this study introduces an novel, comprehensive life cycle assessment (LCA) study of HC production, examining environmental impacts across different scales, from laboratory to industrial, and suggesting improvement strategies. By incorporating diverse data sources and investigating into the effects of processes, this LCA study not only fills primary data gaps but also improves the understanding of upscaling effect on environmental performance and provide a consistent comparison with other LCA studies.

This study, comparing HC synthesis in two labs with several electricity mixes and emission scenarios. It addresses uncertainties in data collection and system modeling (e.g., efficiency of equipment), analyses multiple biomass types and synthesis pathways, and provide a foundation for industrial-scale implementation feasibility. The research further explores upscaling from laboratory to pilot- and industrial production. A prospective LCA integrates upscale methodologies from previous studies to predict the environmental footprint with higher throughput, and demonstrates the upscaling effect of carbon-based anode materials. A follow-up comparison with the previous LCAs of HC and graphite establishes the benchmarks of HĆ carbon footprint against existing anode materials from laboratory to industrial scale, using consistent background data and methodology for fair comparison.

This LCA study represents a novel approach to the field of HC, demonstrating the correlation between environmental performance, process design and production scale,. Furthermore, it provides a detailed blueprint for advancing HC production along an environmental sustainability roadmap.

2. Literature review

Recent research has identified that the HC is in the early market development phase, positioned between Technology Readiness Level (TRL) 6–7 with significant growth potential. [9] At this stage, there is a high degree of flexibility to design the system, offering opportunities to mitigate environmental impacts before it escalates to a higher level. Conducting LCA at this stage enables the prospective identification of potential environmental concerns and hotspots related to HC production. [11] Studies have investigated various manufacturing pathways for HC [12–14] and assessed their environmental footprints. [15–20].

A key process in manufacturing HC from organic material feedstock is carbonization (or pyrolysis), which involves decomposing precursor materials at temperature above 1000 °C in an inert atmosphere. Carbonization is often complemented by pre-treatment of precursors (e. g., grinding, washing, drying) and post-treatment (e.g., ball milling, sieving, scrubbing) to enhance the quality of final product and alleviate emission. [5] Additional treatments, such as acid treatment, hydrothermal carbonization (HTC), and stepwise pyrolysis have been explored to improve the electrochemical performance of HC in SIBs. [13,14,17,21,22] Strategies to reduce the energy consumption in HC production include increasing carbon yield, lowering carbonization temperature, and shortening reaction time. [17,23,24] As many manufacturing processes of HC are similar to those of graphite, such as pyrolysis and de-ashing, it is suggested that leveraging existing expertise and equipment from graphite production could accelerate HĆs

commercial viability. [9].

Peters et al. conducted the first LCA of HC anode materials in SIBs, indicating that the production of HC material from sugar exhibits the highest environmental impacts among various precursors. [25] Conversely, HC from biowaste and petroleum coke demonstrated lower impacts, attributed to lower impacts from upstream feedstock and higher yield percentage, respectively. [18] This research underscores the complexities of utilizing bio-based materials, which may not always be practical or cost-effective. [7,8] Indeed, bio-based materials such as resin and sugar could be burden-intensive as the upstream supply chain for production could have high impacts, for instance associated to intensive energy consumption. However, the series of studies modeled the HC synthesis mainly based on secondary data and emphasized the need for experimental and industrial data from the most probable production pathways, and the need to identify processes and precursors associated with low impacts.

Titirici et al. explored alternative HC production methods, such as HTC, as a way to curtail the energy use and greenhouse gas emissions. [16,17] Follow-up research by Trotta et al. emphasized the importance of investigating the upscaling effects on the environment impacts and optimizing process routes to minimize the impacts. [15] Other preliminary LCA studies based on primary data have been conducted to investigate the effects of processes and precursors, but only with a limited extent of LCA methodology implementation. A comprehensive comparison of these effects is not offered. [13,26].

Graphite as a conventional anode material and other candidate for SIB anode materials are good references and baseline for identifying the environmental performance of HC. The commercial LCA database ecoinvent 3.8 contains a dataset for battery grade natural and synthetic graphite based on literature and approximates the industrial baseline for graphite production. [27] Detailed LCI proposed by Engels et al. for natural graphite based on primary data from manufacturers are remodeled as a solid reference for comparison. [19] Surovtseva et al. offer a combined LCI for synthetic graphite for commercialized scale but based on public literature data. [20] This study draw upon those extensive datasets, including the commercial LCA database ecoinvent 3.8 and scientific papers, and compare these with new data from advanced HC manufacturing processes to establish a robust environmental baseline. There are also LCIs for graphite manufactured by new technologies or for other emerging anode materials for SIBs, such as synthetic graphite via electrochemical graphitization provided by Kulkarni et al. [28] and Fe2O3 anode materials for SIBs proposed by Malara et al. [29] These technologies are at a very early stage of development. Thus, the LCA results of these emerging materials are in most cases excessively high and are not included in the comparison of this study.

A significant gap in current research is the lack of a deep investigation into primary driver of HĆs environmental burden comparing different precursors, process strategies and parameters in the HC production chain, inconsistency of data. [8] Most studies rely on secondary data or limited data from a single laboratory. Secondary data can lead to uncertainties and inaccuracies due to the differences between actual production and inconsistent assumptions based on literature. The primary data from single source might lead to potential uncertainties caused by infrastructure and measurement protocols. There is a need for experimental and industrial data, and robust LCA methodologies for accurate and standardized comparisons. [15].

Furthermore, existing LCAs often overlook crucial emissions data (such as methane, biogenic CO_2, and SO_2) and made broad assumption about electricity consumption, leading to high uncertainties in impact assessment. However, these aspects have been identified as sensitive factors in LCAs, where slight variations of emission and energy consumption can significantly alter environmental impact results. The electricity mix (a part of background system) used to manufacture HC can significantly impact the environmental impact and has also not been discussed. [33].

Many methodologies have been proposed to upscale laboratory data

for prospective LCA studies of higher manufacturing scales and maturities of whole cells or batteries. Erakca et al. discussed how the influence of production scale on the environmental impacts of LIB cell production had been demonstrated with the strong correlation of specific energy demand and production scale. [30,31] However, similar research demonstrating the upscale approach does not exist for HC materials. There has been no research on the pilot- and industrial-scale production of HC. LCA based solely on laboratory-scale data only provides a preliminary direction and cannot represent large-scale commercialization and fabrication. Also, most graphite LCA studies are based on industrial scale. In order to offer a fair comparison between HC and graphite, prospective results associated with the environmental impact of HC pilot- and industrial production are urgently needed. By integrating methodologies designed to predict the environmental footprint at higher throughputs, this research not only contextualizes the scaling effects on hard carbon anode materials but also sets new benchmarks against existing anode materials from laboratory to industrial scale.

Unlike previous studies, this research critically examines HC production from various bio-waste precursors across different scales. This study seeks to bridge these gaps by offering a comparison with previous LCAs, to establish an environment baseline for HC production and providing a detailed insight into sustainable advancement of HC as anode materials for SIBs. This work stands out by incorporating a systematic approach to upscale from laboratory data to pilot and industrial production, offering a unique perspective on the environmental impacts at each stage of scaling. The holistic approach proposed by this work is crucial for transitioning HC from a promising laboratory innovation to a commercially viable and environmentally sustainable solution.

3. Method and materials

According to ISO 14040/14044, the four main stages of an LCA include goal and scope definition, inventory analysis, impact assessment, and interpretation. [32,33].

3.1. Goal and scope definition

The primary objective of the present LCA study is to identify the environmental hotspots of HC production produced from different bio-waste and to evaluate sustainability potentials of HC for large-scale applications in comparison with graphite. The LCA starts at a laboratory perspective by leveraging primary data from two laboratories, aiming to explore various biomass precursors and synthesis pathways.

This initial phase lays the groundwork for a prospective pilot and industrial-scale LCA. The upscale model, built upon laboratory data and supplemented with values from literature, facilitates a balanced comparison with commercial anode materials. The presented results examine the key factors influencing the environmental impact during the escalation of production scale and maturity.

This is a cradle-to-gate LCA study, i.e. the use phase and the end-of-life treatment are excluded from the scope of this study, as shown in Fig. 1. The foreground system consists of the synthesis process based on primary data, while the supply chain of raw materials and energy are modeled in the background. Data from the database ecoinvent 3.8 is used for the background system. The functional unit of this LCA study was primarily set to 1 kg of HC. However, aligning with literature practices, the results can also be converted into a secondary functional unit, such as 1 mAh, to take the electrochemical performance of HC into consideration and provide a different perspective on the findings. [13,15,26].

3.2. Data source and assumptions

Primary data, including process routes for the synthesis of HC, material flows and energy consumption, were collected directly in two laboratories, respectively in China (A) and Germany (B). Studies related to both measurements have been published in [14] and [13]. The main precursors presented in the two publiaitions are bamboo and hazelnut. However, additional biomass, listed in Table 1, were tested using the same process design for comparison in these two laboratories. The experimental primary data for all listed raw materials are provided to conduct the laboratory LCA. More details about the electrochemical domain can be found in these two publications. The primary data obtained through measurements and calculations are the main components of the foreground system.

To ensure the consistency and comparability across different scenarios, assumptions and extrapolation are employed. Energy consumption from the two laboratories is recalibrated based on the maximum capacity of the infrastructure, considering that laboratory facilities do not always operate at peak efficiency. [30,31] Additionally, energy allocation in multi-output processes in the laboratory, such as a drying oven and fume hood used in acid soaking, is determined through a combination of assumptions and expert judgment by laboratory scientists. For example, drying oven is determined by the heating time, space required, and maximum operating space of the oven. Fume hood is for different chemicals simultaneously. Thus the ratio of energy is also determined by the space suggested by scientists. In the contribution
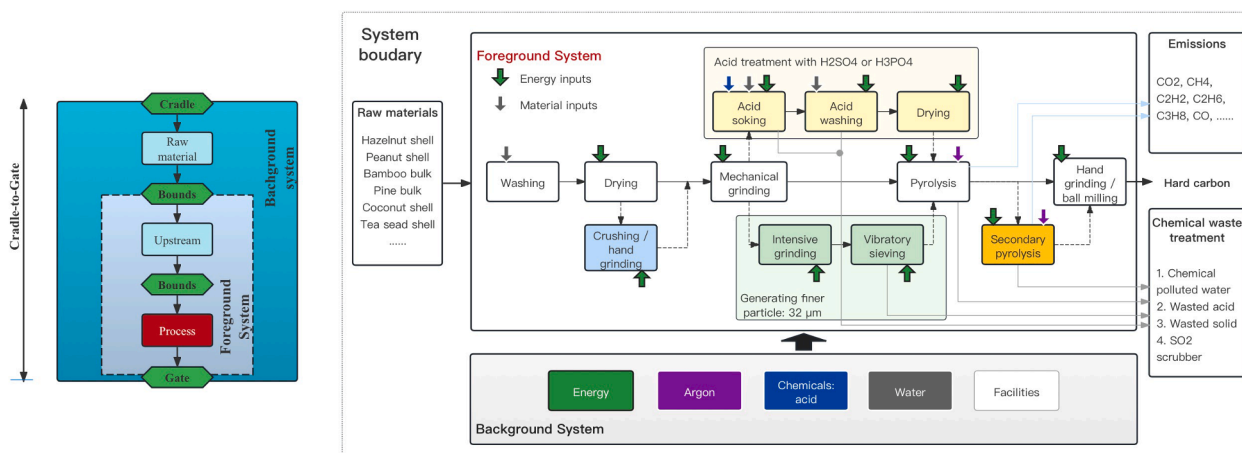

Fig. 1. System boundary of lab-scale synthesis from laboratories A and B. The diagram on the left represents the cradle-to-gate boundary of the lab-scale LCA for hard carbon synthesis, which encompasses the supply chain of raw materials and other input materials. The diagram on the right depicts the laboratory synthesis process route for hard carbon, with data from two laboratories. The main production steps are identical, with some additional processing steps listed alongside. The supply chain of the input substances is taken from the background system (modified and adapted from [34,35]).

Table 1
Description, parameters, and properties of HC synthesis in laboratories A and B.

	Laboratory A		Laboratory B
Basic information			
Raw material	Bamboo bulk, pine bulk, coconut shell, tea seed		Hazelnut shell, peanut shell
Process route	Direct pyrolysis: raw materials are directly put into the tube furnace for two-step pyrolysis Mechanical grinding: raw materials are crushed by mechanical grinder into small particles Acid treatment: raw materials are treated by sulfuric acid for 6 h and then washed before two-step pyrolysis		Direct pyrolysis: raw materials are washed by deionized water and dried, then put into the tube furnace for one-step pyrolysisIntensive grinding: after mechanical grinding, raw materials are ground with intensive grinder and sieved to get smaller particles (<32 μm or 1 mm) Acid treatment: raw materials are treated by phosphoric acid for 2 weeks and then washed with deionized water before pyrolysis
Scenarios and abbreviation	• bamboo_direct: direct pyrolysis with bamboo bulk • bamboo_at: pyrolysis with acid treatment with bamboo bulk • pine_at: pyrolysis with acid treatment with pine bulk • coconut_at: pyrolysis with acid treatment with coconut shell • teaseed_at: pyrolysis with acid treatment with tea seed		• hz_ww_1mm: direct pyrolysis with water washing, grinding below 1 mm, hazelnut shell • hz_ww_32μm: direct pyrolysis with water washing, intensive grinding below 32 μm, hazelnut shell • hz_at_1mm: pyrolysis with acid treatment, grinding below 1 mm, hazelnut shell • peanut_at: pyrolysis with acid treatment, hand crushing (due to low hardness of peanut shell), peanut shell
Acid treatment			
Acid agent	Sulfuric acid		Phosphoric acid
Treatment time	6 h		2 weeks
Pyrolysis			
Pyrolysis type	Two-step fast pyrolysis		One-step slow pyrolysis
	Primary pyrolysis	Secondary pyrolysis	
Max. operation capacity of furnace (g/batch)	10	10	15
Temperature (°C)	500	1300	1100
Heating rate (°C/min)	10	10	1
Holding time (hour)	2	2	1
Cooling rate (°C/min)	10	10	3.13
Inert gas	Argon		
Flow rate (mL/min)	200–1000 (200 mL/min applies for base case) [39]		
Measurement			
Location	China		Germany
Energy consumption	Primary data directly measured		Primary data based on power and time
Scaling of energy	Based on maximal operation capacity		Based on maximal operation capacity
Reference	[14]		[13]

analysis, data from both laboratories are modeled using the same background system, i.e. German energy mix and supply chain (If German supply chain is not available, average EU supply chain is used). Then the scenario analysis was conducted to investigate different locations, including Germany (DE) and China (CN). Additionally, a scenario

was developed to examine the potential of a pure renewable energy supply, with 50 % wind and 50 % PV. These scenarios were designed to demonstrate the importance of location and energy supply selection. The grid mix data was sourced from Ecoinvent 3.8, which has considered both transmission losses and electricity generation specifics. However, it is important to note that these modeling assumptions introduce a degree of uncertainty, which is further investigated through scenario analysis to understand the inherent uncertainties of these assumptions.

Transportation are not considered in this laboratory LCA. The goal of this study is to compare the effects of process design and inherent differences of biomass on the environmental impacts. Therefore, the transportation is not a necessary consideration, especially when it involves a high degree of uncertainty and only makes minor contribution to the overall impacts.

3.3. Assessment method

The ILCD (International Life Cycle Data System) 2011 midpoint is used as base Life Cycle Impact Assessment method (LCIA). This method is recommended by the European Commission for LCA and is widely recognized by previous battery-related LCA studies. [36] The calculation is conducted in the software openLCA V11.1. [37] To conduct the impact assessment, LCI data were converted into midpoint indicators using characterization factors provided by the ILCD 2011 methodology. This step involves aggregating emissions and resource consumption data from the inventory analysis and calculating the contribution of each process to the selected impact categories. The results are then analysed to identify the most significant environmental impacts and hotspots in the life cycle of HC. Comprehensive environmental impacts of all other impact categories from the ILCD method are shown in S3.1. Among them, the impact categories selected for contribution analysis focus on global warming potential (GWP), acidification potential (AP), and resource depletion potential (RDP), as these three categories have been verified to represent the most significant impacts and always been discussed in previous hard carbon LCAs. [16,18].

Scenario analysis is conducted on several variables to evaluate the sensitivity of input material and robustness of results. The variables considered in this analysis included process parameters (e.g., volume of inert gas amount, carbon yield of pyrolysis), type of acid, throughput of pyrolysis equipment, and assumptions related to geographical supply chain (including electricity mix and raw material supply). The goal of this scenario analysis is to assess the sensitivity of the process parameters and approaches to the overall environmental impacts of HC. For example, a scenario analysis of inert gas was conducted, in which the flow rate was linearized based on theoretical considerations. In the scenario analysis of acid type, we proceeded to consider the theoretical possibility of a change in the type of acid, while maintaining the quantity and other material balance constants.

3.4. Life cycle inventory modeling

Various biomasses are considered for the laboratory-scale manufacture. The biomass materials need to have a suitable structure for HC, easy to collect on a large scale, and low cost. The biomass investigated in this study has been selected by the researchers and validated in laboratory to provide stable performance for hard carbon. The laboratory synthesis of HC can be divided into three main stages: pre-treatment, pyrolysis, and post-treatment. In this way, a typical route for HC synthesis mainly consists of pre-treatment (washing–drying–mechanical grinding)–pyrolysis–post-treatment (ball milling, scrubbing). Based on this main route, acid treatment, intensive grinding, and secondary pyrolysis are optional steps, as shown in Fig. 1. Noted that the grinding conducted at the end of the process route is distinct from the grinding conducted prior to pyrolysis. Based on expert interview, the hand grinding and ball milling are essential for achieving the desired particle size and uniformity of battery-grade anode active materials. [38]

Although the specific process route varies up to raw material selection and equipment functionality, the overarching framework remains consistent across different scenarios in this study.

The aim of both laboratories is to identify the most environmentally sustainable approach and material for the production of HC. All scenarios of process design in laboratory A and B were strategically designed to isolate and compare the effects of different treatment methods on a single raw material. This approach was employed prior to any subsequent extensions of such methods to other materials. For example, in laboratory A, bamboo was initially investigated to compare the environmental impacts of direct pyrolysis and acid treatment. This controlled comparison ensured that any differences observed in environmental impact could be attributed solely to the treatment method, rather than to variations in the raw materials. Different raw materials were then subjected to the same process route and conditions to assess the influence of raw materials on environmental impact.

Cooperating with scientists Laboratory A and B, the material flow data and energy consumption on a laboratory-scale was measured directly on-site during the experiments. There are two ways to measure energy consumption: direct measurement of the energy consumed during a certain time in the format of kWh; or measuring the power (W) and then calculating the total energy consumption ($\sum$W*h) based on the time (h), Energy use by unmeasurable equipment is calculated using maximum power and actual running time. The energy consumption is recalculated based on the maximum acceptable feeding capacity of the equipment, when the equipment is fed according to the actual needs of scientists in laboratory activities and does not run at its maximum power. The HC synthesis system is therefore modeled with a bottom-up approach, providing the material flows and energy consumption of each process.

Data collection in two laboratories enables a comparison of different routes and a discussion of laboratory-specific LCA from a methodological perspective. However, it can also lead to discrepancies in data collection, as the two laboratories apply different equipment (including measurement and synthesis equipment) and have different background (including energy-mix and supply chain, room environment, and experimental routines). Table 1 illustrates the detailed parameters and properties in laboratories A and B in order to present the details of and differences between the two sources.

A comprehensive inventory of the material and energy flows associated with the laboratory-scale production of HC is provided in S1.1.

3.5. Upscaling to pilot- and industrial scale

A simplified scale-up and predictive LCA based on the laboratory results was performed in order to get a general idea of the sustainability level of HC on the map of anode materials and the scale effect. The best case demonstrated in the laboratory scale (i.e. "bamboo with H2SO4 treatment") is upscaled to two different levels: pilot scale (based on 100 kg/batch), which is currently technically feasible and has been executed by commercial manufacturers, and hypothetical industrial scale (based on 1000 kg/batch), which is expected to be achieved in future HC plants. This information has been provided by developers of HC industry.

Pilot trials have been conducted by many commercial HC manufacturers. Previous studies have suggested that energy consumption is likely not linearly related from laboratory to higher scale. [40] The process route and parameters (e.g., time, temperature) of laboratory scale are used to extrapolate the energy consumption of pilot scale. Moreover, the energy efficiency and equipment size vary from the pilot plant setting. Based on expert interviews [9], most of the current HC pilot equipment has been converted from past graphite equipment, and pyrolysis indeed is seen as a low temperature graphitization. [5,9] Therefore, energy consumption for graphite pilot production equipment can be adapted to generate the energy balance for pilot-scale LCA of HC. [41] The material balance (e.g., reactants, products) is proportionally upscaled according to throughput (from 10 g to 100 kg/batch) and then modified based on the empirical values of equipment efficiency. This equipment-based upscaling of energy and material balance enables a process-to-process reflection of the real situation.

However, there is currently limited information on large-scale production equipment or processes of HC. This means no such equipment-based energy expansion for pilot-scale is available for industrial scale. This study employed an alternative thermodynamic and chemical reaction-based energy estimation method to estimate energy consumption and scale-up laboratory experiment to industrial scale. Before continuous production, it is expected that HC production will be first batch setup in the future. Therefore, the prospective LCA of industrial scale is based on a reaction setup of 1000 kg/batch, with 20 % material saving due to due to efficiency. [42] The energy balance for each process is calculated with corresponding theoretical energy exchange equations. [42–44] This scenario excludes direct emissions treatment and consequential recycled energy.

Energy recovery by combustion of exhaust gas from pyrolysis, also called pyrogas, is expected to be applied in future industrial plants, followed by flue gas treatment. To explore the potential emission reduction in the future, a hypothetical industrial "best case" scenario is formulated with the following assumptions:

- Complete emissions treatment (no additional carbon footprint)
- Renewable energy (50 % PV and 50 % wind)
- Utilization of flue gas-derived energy for 50 % drying
- 68 % material recovery rate [42]

It is assumed that flue gases and bio-oils from the pyrolysis furnace are treated through complete combustion. A conservative assumption suggests that the energy recovered by the combustion of flue gas can cover 50 % energy requirement of drying process in the acid treatment. The industrial-scale production of hard carbon is an emerging technology that is still in need of detailed and recent data on material recovery rates. To ensure consistency across the upscaling, the 68 % recovery rate is adopted from the primary data source [42], which represents the most relevant average value currently available. It is noted that this model is an ideal scenario with simple energy balance calculation and conservative assumptions in order to demonstrate the potential for environmental impact reduction through simple optimization approaches in the future. It is recommended that future research conduct a more detailed analysis.

The objective of this study is to examine the general upscaling effect of HC, rather than to identify a specific supply chain or absolute value. Therefore, the transportation and co-product are not included in this study. It is crucial to recognize the significance of their contributions in future research, especially in the context of industrial scale considerations. Average material efficiency rate and infrastructure data are based on average value from literature. Since this method is based on theoretical calculations, large uncertainties are taken into consideration. Considering the goal and scope of this study to provide a preliminary estimation of a future situation, it allows us to roughly draw a baseline of the industrial scale for an initial comparison with other commercially available anode materials. Further details of calculation and assumption for the pilot- and industrial scale are available in S2.1 and S2.2. Life cycle inventories are found in S1.2.

3.6. Harmonization of other LCA studies for comparison

Findings from this study are benchmarked against previous LCA studies and existing commercial datasets of other anode materials, including HC and graphite, focusing on carbon footprint and energy consumption as key indicator, as these impact factors of the greatest interest. This comparison aims to validate results of this study and assess the impact of different production scales. It assists to verify the sustainability level of HC in current anode systems in order to identify its environmental mitigation potential and enhance the sustainability of

SIBs.

To ensure a consistent and fair comparison, this study reconstructs models from selected LCA studies using an identical assessment methodology and background data. [15–20,27] The results are reproduced based on LCI data provided in the literature, while consistent background data (Germany / European dataset), the modeling approach (bottom-up modeling), and impact assessment method (ILCD 2011 midpoint+) of LCA are used across different research. Potential methodological variances are minimized by adopting an identical framework encompassing the same geographical and temporal scope, emissions, and waste treatment methods, and LCIA approach. This harmonization ensures that differences in the environmental impact between HC and graphite are attributed solely to the materials, rather than discrepancies in assessment methodologies. By comparing results from this study with the results from adopted models, the analysis highlights the areas where HC shows superior environmental performance.

3.7. Limitations

This study is subject to several limitations. One significant limitation is the unaddressed uncertainties associated with upscaling. Due to lack of data, these uncertainties cannot be eliminated at current stage. Some assumptions were drawn from the reference that may be outdated, such as the material recovery rate, due to data availability. It is recommended that further research is conducted with updated data to potentially validate or revise the assumed value taken in this research. Moreover, the current upscale model, based on laboratory-scale data, lacks optimization of parameters and processes at a higher scale. It still has distance from the realities of actual mass production. Therefore, further research with a more reliable upscale method of HC is needed. Another limitation pertains to the lack of modeling for continuous processing, which, according to Elginoz et al. [43], could significantly differ in energy and material flow from batch processing. Finally, this study does not encompass the environmental impact of the biomass supply chain and transportation, as this aspect was beyond the study's scope. Future research could delve into the consequences and potential alternatives of using biomass waste, to gain a comprehensive understanding of HC production and its environmental footprint.

4. Results and discussion

4.1. Contribution analysis of lab-scale production

4.1.1. Energy consumption analysis

Fig. 2 presents the energy consumption breakdown of lab-scale HC

synthesis. The results highlight that the pyrolysis process plays dominant role (up to 98 %) in the energy consumption. Also, the equipment efficiency and the preparation methods employed in the laboratories have a significant impact. For example, despite recalibrated to maximum capacity, there is still a significant gap between laboratory A and B, stemming from the efficiency of the employed equipment, especially pyrolysis furnace. Furthermore, Laboratory A implemented a two-step pyrolysis process incorporating a high-temperature secondary stage (1300–1800 °C). While this short duration and the higher temperature stage consumed more energy compared to the primary pyrolysis (300 °C–650 °C), it reduces even more energy of the primary pyrolysis process and increase the overall HC yield, therefore leading to energy savings for the entire system. This indicates the potential benefits of optimizing the pyrolysis process for enhanced sustainability performance. Secondary pyrolysis also represents a potential method for enhancing the electrochemical performance of hard carbon. [14] However, possible barriers hindering the widespread adoption of secondary pyrolysis and the potential adverse effects on the HC produced should be further investigated in future research.

In laboratory B, an additional acid treatment step enables the removal of impurities from the biomass raw material and increases the carbon content in the precursor, increasing the yield of the consequential pyrolysis process and reducing pyrolysis' energy consumption (bamboo reduced the overall energy demand by 5.9 % and hazelnut shells by 16.4 %). Comparatively, the added energy consumption of acid treatment is significantly less than the saved consumption from the pyrolysis process. In addition, in order to generate 32 μm hazelnut particles, intensive grinding and vibratory sieving are conducted, which lead to substantial material losses (approximately 80 %). Therefore, the quantity of dried raw material required for 32 μm is greater than that required for 1 mm. This results in a notable increase in the energy consumption required for drying. In contrast, the energy associated with additional intensive grinding and vibratory sieving is relatively insignificant.

Different biomasses feedstocks exhibit varying yields even with identical treatment approaches, parameters, and conditions for processes like acid treatment or pyrolysis. These disparities arise from their internal structure and material composition. These yield variations impact both energy consumption and environmental footprint. The selection of biomass type can influence energy consumption by affecting the yield. Interestingly, the difference between raw materials type (biomasss) is not as significant as the difference between the process routes. This is likely due to the fact that in our study, all biomass raw materials were treated as bio-waste, and therefore, their inherent environmental burden was excluded.
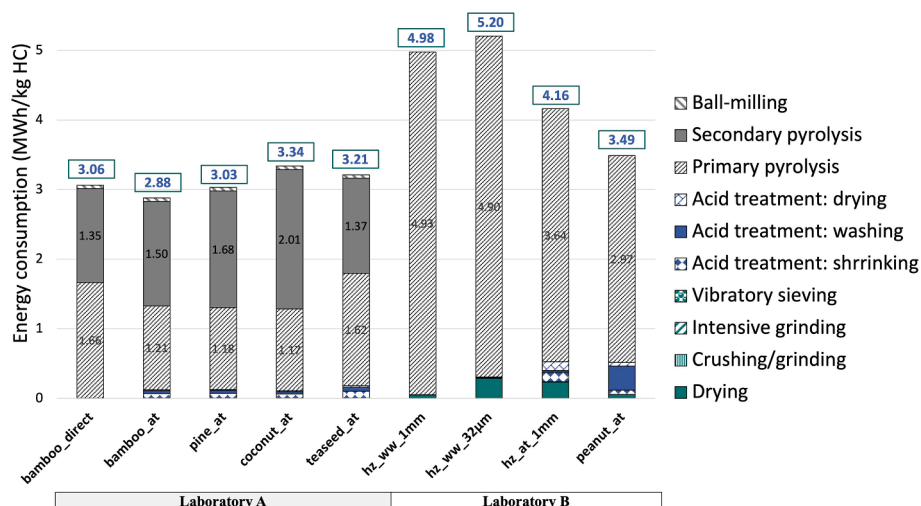

Fig. 2. Energy consumption (MWh/kg) based on maximum capacity from laboratories A and B.

4.1.2. Environmental impact analysis

Fig. 3a–c showcases the environmental impacts of HC production in laboratories A and B using identical background data (German energy mix, European supply chains of raw materials). Fig. 3a reveals that lab Á secondary pyrolysis, despite contributing to 23–35 % GWP, significantly reduces the primary pyrolysis GWP by over 50 % through increased HC yield and reduced the temperature/time of the first pyrolysis. The shorten pyrolysis time also means the reduction of corresponding inert gas consumption. The GWP of the pyrolysis process are strongly correlated with the pyrolysis temperature and time. Inert gas supply emerges as the third major contributor, strongly linked to pyrolysis time and heating rate. Longer acid treatment times, such as for peanut shell, increase GWP (up to 5.1 % more GWP impact) due to higher electricity consumed in treatment facilities. Similarly, preparing finer particles of hazelnut shell involves an additional intensive grinding, contributing an additional 4.4 % to GWP.

As shown in Fig. 3b and c, the highest contribution to the Acidification Potential (AP) and Resource Depletion Potential (RDP) is also the energy demanded for both pyrolysis processes. Unlike GWP, the use of phosphoric acid in laboratoy B causes significantly more AP and RDP impacts, and the inert gas used in laboratoy B also contributes notably in. In contrast, laboratoy A uses surfuric acid with less environmental burden than the phosphoric acid supply chain. Moreover, laboratoy A minimizes the argon consumption due to the shorter heating time required by the more efficient equipment (faster heating rate). Section

4.2 presents further scenario analysis exploring the distinct impacts of acid types and inert gas amount on various environmental categories.

Note that the low energy efficiency and material utilization inhere in the experimental-laboratory-scale foreground data, coupling with industrial-scale background data (ecoinvent 3.8) for modeling, can amplify background system impact and diminish foreground data differences. This emphasizes the importance of matching the foreground and background system scales when performing LCA. The subsequent upscaling analysis in this study provides further insights into this phenomenon.

4.2. Scenario analysis

The scenario analysis results are presented by the most important impact categories identified in the contribution analysis in order to assess the influence of key parameters on environmental performance. Fig. 2 highlights energy consumption as the primary driver of HC production. A dedicated scenario (Section 4.2.1) explores the potential impact of shifting production location and utilizing a pure renewable energy mix, compared to the base case with German E-mix and supply chain.

Previous literature and expert feedback suggest a wide range of accepted inert gas quantities that can be fed into the pyrolysis furnace, which introduces uncertainty. To assess its influence on the results, a sensitivity analysis of inert gas is conducted in Section 4.2.2.
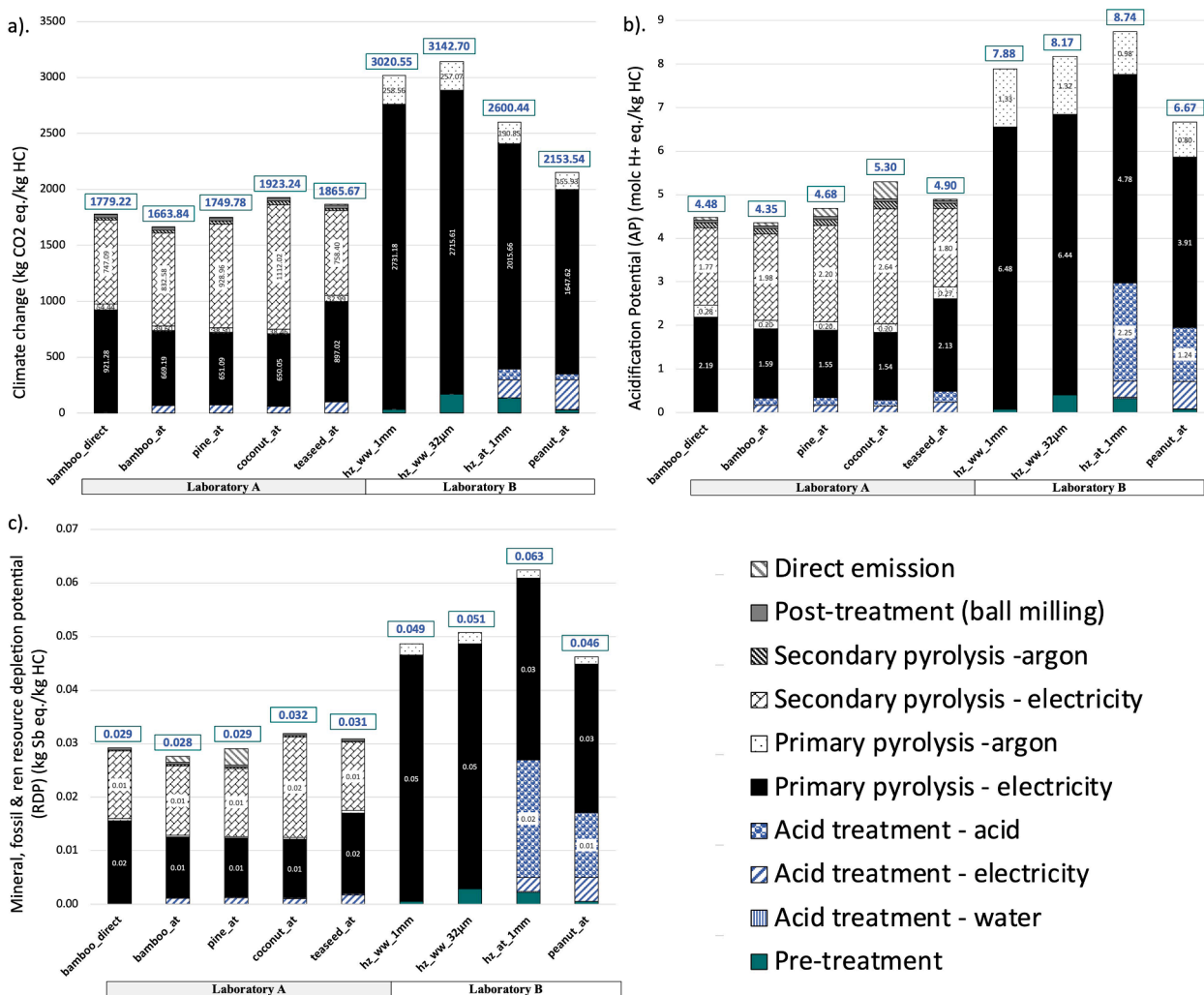

Fig. 3. A).Global warming potential (GWP), b). Acidification potential (AP), c). Resource depletion potential (RDP) of hard carbon synthesis from laboratory scale including laboratories A and B.

The acid used in laboratory B exhibits different contributions to the AP and RDP against laboratory A, which is attributed to the utilization of different acid types. Section 4.2.3 analyzes scenarios involving different acid to elucidate their impacts.

The pyrolysis efficiency, based on carbon yield and furnace throughput, are directly linked to the pyrolysis energy consumption and inert gas amount, representing a important factor for enhancing HC environmental performance. Section 4.2.4 explores scenarios with varying pyrolysis efficiency to showcase the importance and potential for improvement.

4.2.1. Background: Electricity and supply chain

Fig. 4 shows the AP, GWP, and RDP of different background system on HC production and compares Chinese (CN), German (DE), and renewable (REV) electricity mix. The impacts associated with GWP and AP resulting from the German supply chain are significantly lower than those of the Chinese supply chain. In contrast, the RDP is higher in the German scenario, primarily due to the high proportion of photovoltaic (PV) electricity in the grid, as production of PV entails higher embodies depletion of metal resource. The REV E-mix, representing pure renewable electricity mix scenario (50 % PV + 50 % wind) can further evident this influence. However, renewable energy supply can reduce substantial GWP and AP impact of production process, indicating the crucial role of electricity supply in optimizing carbon footprint and acidification potential of HC production. Reduction of GWP and AP of different E-mix outweighs the differences between laboratories, which employs different process routes. It is important to consider the potential benefits of adopting cleaner electricity sources and supply chain as a key strategy for reducing carbon footprint of HC production.

4.2.2. Inert gas

Previous studies have discussed the influence of the flow rate and type of inert gas on the properties and electrochemical performance of HC. [45–47] According to consulted developers, the flow rate of inert gases is not strictly controlled in real experiments. The possible gas flow rate can fluctuate in a wide range (+/- 500 % based on current estimation). The flow rate of the inert gas (argon in this study), needs to be investigated in LCA of HC, especially considering future industrial

production. Table 2 shows that a variation between 0.15 % and 2.8 % for environmental effects was observed by increasing the argon flow rate by 10 % (base case assumption of 200 mL/min argon flow). Laboratory B, with longer pyrolysis time and more inert gas consumption, exhibits greater environmental dependence on flow rate, especially for ionizing radiation and water resource depletion. Considering the effect of inert gas flow on electrochemical performance, the overlap due to excessive inert gas might lead to a significant sustainability risk in HC production.

There is no way to measure the argon flow rate in neither laboratory and previous literature suggest a wide range (200–1000 mL/min). [39]. Due to high sensitivity to inert gas flow rate, shifting the minimum flow (200 mL/min, base case) to the maximum flow rate (1000 mL/min) can lead to changes in ionizing radiation of up to 131 % (details in S3.2).

4.2.3. Type of acid

The sensitivity of acid type on AP and RDP is investigated using scenarios with varying acid types while keeping the same amount. Table 3 reveals that phosphoric acid contributes the most to both impact categories among the investigated acids. Therefore, substituting phosphoric acid with sulfuric acid, hydrochloric acid or nitric acid can potentially reduce the impact from acid treatment process. Existing research has discovered the potential impact of acid type on the electrochemical performance of HC used in batteries. [13,21] Future research should consider this potential coupling between environmental and electrochemical performance when evaluating alternative acid types.

4.2.4. Pyrolysis: Carbon yield and throughput

The electricity consumed in the pyrolysis processes is directly correlated with parameters, such as carbon yield and the allowed equipment throughput. The sensitivity of GWP to these key parameters is presented in Fig. 5 (details in S3.3). Based on the base case (carbon yield and throughput measured in laboratory), it is assumed that the carbon yield and throughput can be improved respectively by different percentage, considering future technology development and upscale effect.

Compared to the laboratory A, the improvement in carbon yield offers significant GWP reduction on the laboratory B that employs one-
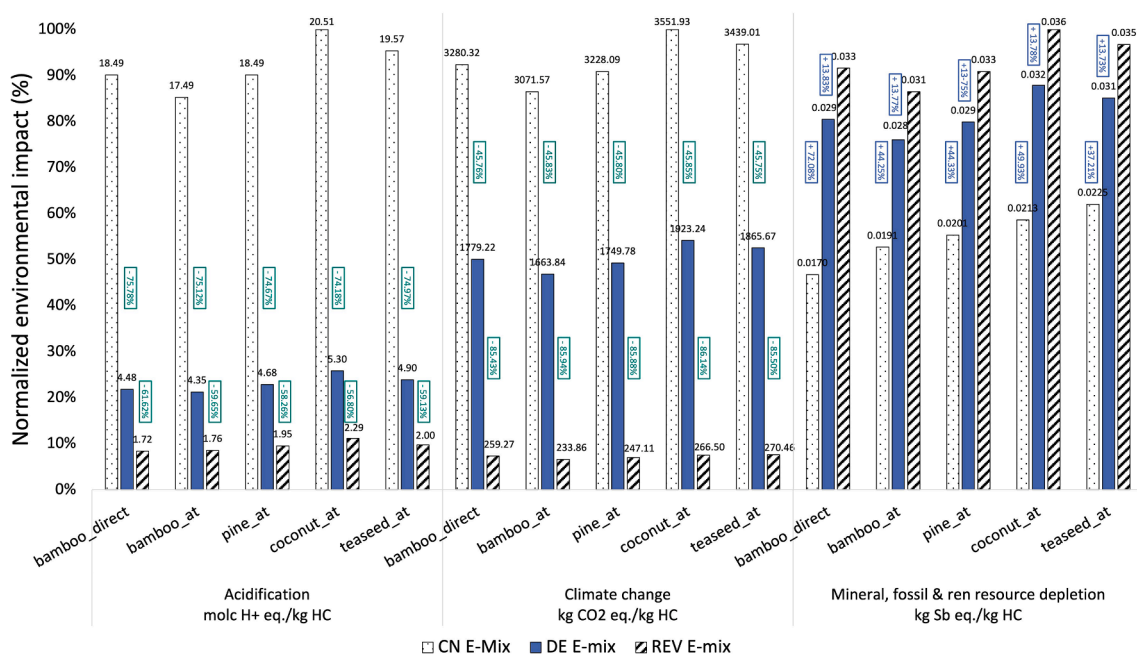

Fig. 4. Environmental impacts (AP, GWP, and RDP) of laboratory B based on Chinese (CN E-mix), German (DE E-mix), and renewable with 50% wind and 50% photovoltic (REV E-mix) electricity. The results are normalized to percentage based on the maximum values (100%) of each impact category. The numbers at the end of each bar show absolute impact results.

Table 2
Sensitivity of argon gas inlet flow rate: increase of environmental impacts with 10% higher argon flow rate (mL/min). The color changes according to the size of the change value (red: better sustainability, green: worse sustainability).

Indicator	bamboo_direct	bamboo_at	pine_at	coconut_at	teaseed_at	hz_ww_1mm	hz_ww_32μm	hz_at_1mm	p_at
Acidification	0.88%	0.76%	0.72%	0.69%	0.79%	1.69%	1.62%	1.12%	1.20%
Climate change	0.43%	0.38%	0.38%	0.37%	0.40%	0.86%	0.82%	0.73%	0.72%
Freshwater ecotoxicity	0.18%	0.16%	0.15%	0.15%	0.16%	0.36%	0.34%	0.28%	0.28%
Freshwater eutrophication	0.30%	0.27%	0.26%	0.26%	0.28%	0.60%	0.58%	0.50%	0.50%
Human toxicity, cancer effects	0.33%	0.30%	0.29%	0.28%	0.31%	0.67%	0.64%	0.42%	0.46%
Human toxicity, non-cancer effects	0.35%	0.31%	0.30%	0.30%	0.32%	0.70%	0.66%	0.52%	0.54%
Ionizing radiation E (interim)	1.58%	1.42%	1.40%	1.37%	1.49%	2.80%	2.70%	2.49%	2.46%
Ionizing radiation HH	1.28%	1.15%	1.13%	1.11%	1.21%	2.33%	2.25%	2.08%	2.05%
Land use	0.21%	0.18%	0.18%	0.18%	0.19%	0.42%	0.40%	0.30%	0.32%
Marine eutrophication	0.55%	0.49%	0.48%	0.47%	0.52%	1.08%	1.04%	0.86%	0.88%
Mineral, fossil & ren resource depletion	0.22%	0.19%	0.19%	0.19%	0.20%	0.44%	0.42%	0.25%	0.28%
Ozone depletion	0.78%	0.70%	0.68%	0.67%	0.73%	1.50%	1.44%	1.13%	1.17%
Particulate matter	0.95%	0.82%	0.79%	0.76%	0.86%	1.80%	1.72%	1.21%	1.29%
Photochemical ozone formation	0.61%	0.63%	0.62%	0.61%	0.67%	1.39%	1.33%	1.08%	1.10%
Terrestrial eutrophication	0.64%	0.59%	0.58%	0.56%	0.62%	1.28%	1.22%	1.03%	1.04%
Water resource depletion	1.31%	1.18%	1.16%	1.14%	1.24%	2.39%	2.30%	2.11%	2.08%

Table 3
Senstivity of acid type: increase/decrease percentage (colour blocks) of a). acidification (ADP) and b). resource depletion potential (RDP) compared to originally-used acid (black blocks with absolute value,) when different kinds of acid are used. The color changes according to the size of the change value (red: better sustainability, green: worse sustainability).

a). Acidification (molc H+ eq./kg HC)	Sulfuric acid	Phosphoric acid	Hydrochloric acid	Nitric acid
Bamboo	4.35	7.47%	-1.94%	-0.46%
Pine	4.68	7.30%	-1.90%	-0.45%
Coconut	5.30	5.46%	-1.42%	-0.34%
Teaseed	4.90	9.77%	-2.54%	-0.61%
Hazelnut	-16.97%	8.74	-21.38%	-18.02%
Peanut	-12.29%	6.67	-15.49%	-13.06%

b). Mineral, fossil & ren resource depletion (kg Sb eq./kg HC)	Sulfuric acid	Phosphoric acid	Hydrochloric acid	Nitric acid
Bamboo	0.0276	16.73%	1.45%	0.94%
Pine	0.0290	16.73%	1.45%	0.94%
Coconut	0.0319	12.87%	1.12%	0.72%
Teaseed	0.0309	21.98%	1.91%	1.23%
Hazelnut	-33.72%	0.0625	-30.80%	-31.83%
Peanut	-25.22%	0.0461	-23.03%	-23.81%

step pyrolysis, due to decreased energy consumption. However, there is limited potential exist to improve the yield during the secondary pyrolysis due to the negligible material loss. Therefore, the assumptions for carbon yield optimization only target the primary pyrolysis, where the mass loss of biomass mainly occurs.

In contrast, the increasing throughput consistently reduces GWP by lowering electricity and resource consumption (e.g. inert gas) across both o laboratory/process pathways. Therefore, enhancing carbon yield of the pyrolysis process and applying more efficient equipment to maximize throughput potential is critical to improve the environmental sustainability of the system in further R&D of HC industrial manufacturing. As demonstrated in this and previous studies, pre-treatment methods such as acid treatment and HTC can improve the carbon yield of pyrolysis and thus increase the environmental performance. [16] However, as carbon yield and throughput increase, the benefits in GWP eventually approaches a plateau. Future research needs to find the optimal point balancing the equipment investment and environmental performance for sustainable hard carbon production at an industrial scale.

4.3. Relationship of scale and LCA result

Fig. 6 highlights the significant environmental benefits of scaling up HC production from laboratory to industrial scale. The GWP impact of HC production reflects a 265-fold reduction from 10 g to 100 kg per batch, and a further 73 % decrease from 100 kg to 1000 kg per batch. While the marginal GWP reduction diminished with upscaling, the GWP impact per kg of HC is optimized with higher throughput and may reach a limit. However, the hypothetical best-case scenario shows that the incorporation of advanced production techniques, including emission treatment, energy recovery, higher material efficiency, and use of renewable energy, can further reduce the GWP of HC.

The results reveal key factors driving the upscaling effect. High GWP in the laboratory setting reflects inefficient equipment and low energy utilization, particularly evident in the pyrolysis step (>90 % of GWP emissions). As production scale increases, more efficient equipment
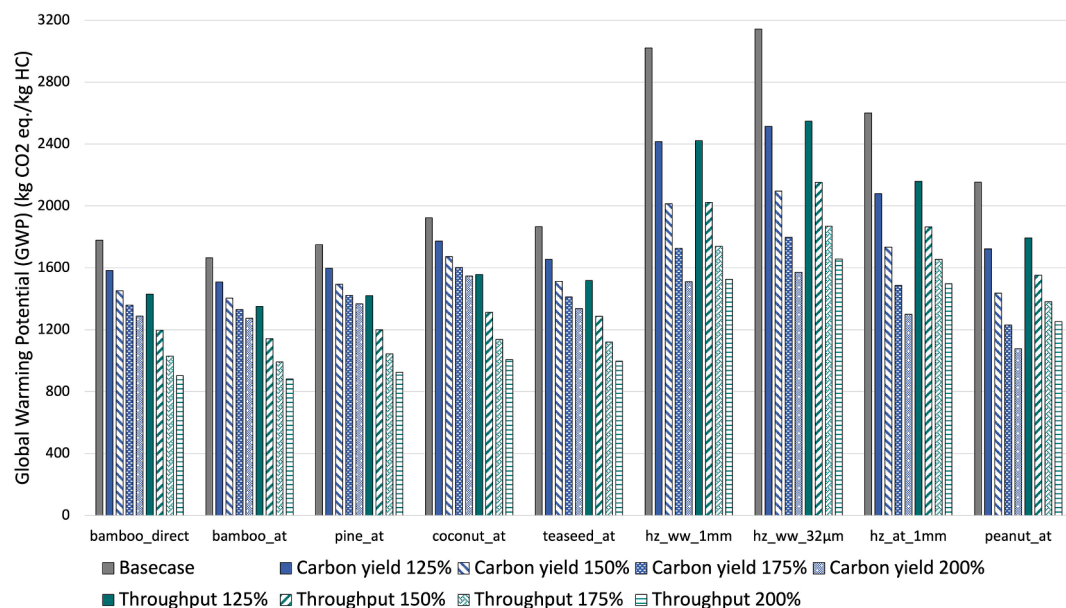
Fig. 5. Sensitivity of carbon yield and equipment throughput of pyrolysis. Change in GWP result (kg CO2 eq/kg HC) by increasing the carbon yield of the primary pyrolysis and the maximum capacity of the pyrolysis equipment (throughput) by 25 %, 50 %, 75 % and 100 %.
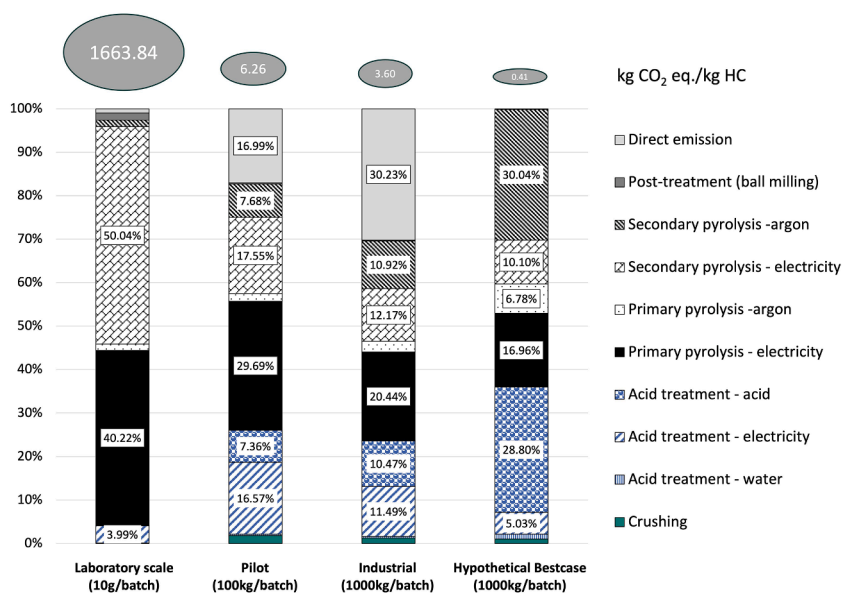

Fig. 6. Contribution of each process to GWP of laboratory-, pilot-, industrial-scale and the hypothetical bestcase industrial production of the process route "bamboo with H2SO4 treatment".

leads to a decreasing ratio of GWP attributed to energy consumption, especially electricity for pyrolysis. Conversely, the ratio of materials increases due to the assumption of limited material savings in the upscaling simulations. Similarly, direct emissions are assumed to increase linearly with scale, leading to an increase in their contribution to the total GWP.

Additional information of the other impact categories can be found in S3.1.

As mentioned in Section 3.6, the results of this study are compared with previous studies. As there is still no LCA of industrial scale HC production, the results are compared only with traditional graphite on industrial scale, to better understand the industrial-scale results and identify the classification of HC environmental performance. The maximum and minimum values of a set of results are selected for presenting different scenarios in one study.

As shown in Fig. 7, the laboratory-scale results of this study are in line with the results of a previous laboratory-scale LCA study of HC. However, notable discrepancies are observed when comparing different production scale. Specifically, it is observed that the preparation of HC exhibits a significant scale effect, although different process route and raw materials are applied. These findings highlight the importance of considering the scale of production when evaluating the environmental performance and sustainability of HC synthesis processes. A comparison between HC and other anode products that have been successfully commercialized on a large scale, such as graphite, needs more information and data for industrial-scale HC. The production of HC involves chemical and thermodynamic reactions, it is better to take into account the factors associated with these reactions during upscaling of HC production.

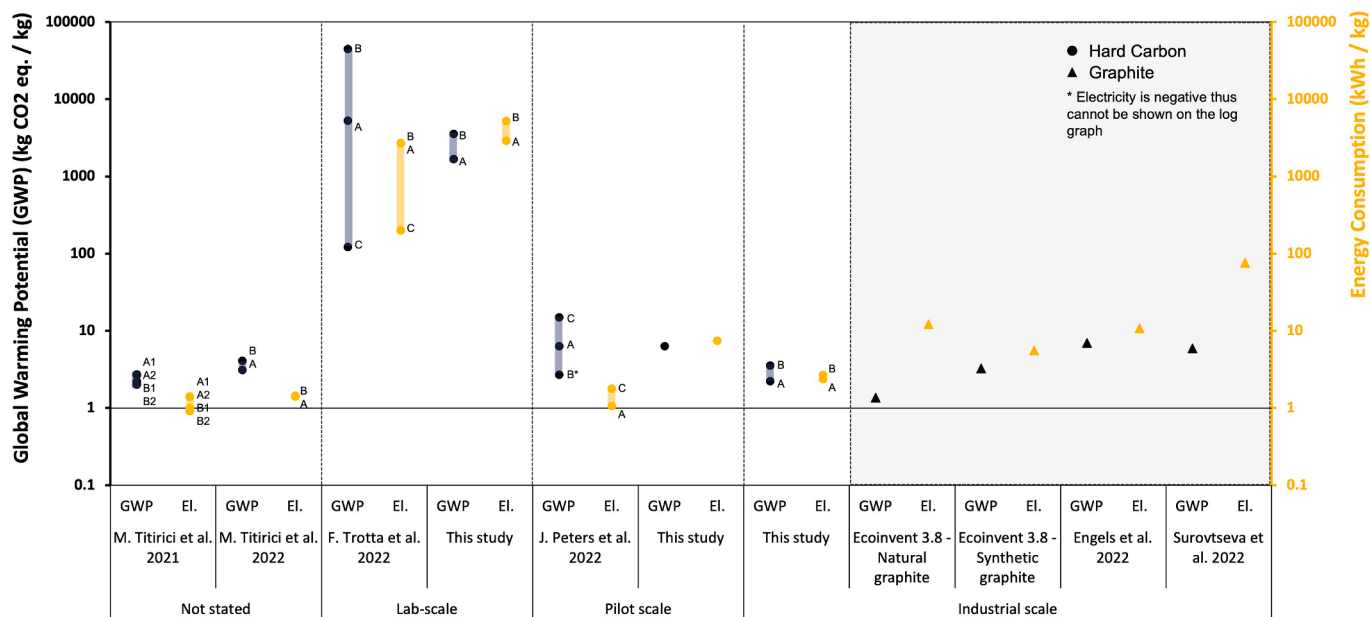It must be also considered that there is a huge difference in

Fig. 7. Comparison of GWP and energy consumption between this and previous LCA studies of HC and a commercial dataset of graphite, with consideration of scale. All results are replicated based on an identical modeling and assessment method for a consistent comparison. Alphabets (e.g. A, B, C) represents different scenarios, more details and explanation of each scanrios are offered in S3.4.

electrochemical performance between different anode materials. The electrochemical performance, i.e. the average gravimetric capacity density (mAh/g), is considered and the resulting GWP value is relative to the average gravimetric capacity density. If the capacity density of anode materials is not provided in the corresponding study, the theoretical energy density is taken. Information on different scenarios and the comparative results based on secondary functional unit mAh is shown in S3.4. In this case, the physicochemical properties of the HC would affect the LCA results. If the properties and technical parameters of HC are improved in manner that leads to, for example, an increase in energy density of HC without additional energy demand, the cell performance should be enhanced. As a result, the environmental performance can be improved based on the functional unit of the energy offered by the cell. However, it is currently not possible to quantify this correlation with sufficient precision, as it is not possible to evaluate the effect of material properties on the cell performance at the material level in a quantitative manner.

It should be noted that the current assumption of pilot- and industrial scale of HC are both batch-based and bottom-up modeled, which could bring inaccuracy. Large-scale production that may occur in the future is ideally through a continuous process route to meet the high demand of HC, informed by industrial technology developers. [48] But there is as yet no continuous production of HC in the existing plants, due to cost and technical threshold. Compared with batch process, continuous process can meet the huge throughput requirements, reduce production time, and increase throughput and efficiency. [42,43,49] Furthermore, the process-to-process upscaling approach establishes a bottom-up modeling of the energy consumption, which might lead to underestimation. [31].

5. Conclusion

This study has made significant progress in understanding the environmental impact of HC production through a simplified upscaling approach. By combining existing upscaling methods from the literature with gathered primary data on a laboratory-scale, the resulting outcomes then compared with the broader literatzre to gain an additional perspective on the environmental impact of future batch production of HC on larger scales.

The laboratory-scale LCA conducted in this study focused on identifying the key factors influencing the environmental footprint per kilogram of HC, utilizing data from two laboratories with distinct manufacturing routes. The objective was to identify the most efficient pathway for HC manufacturing while reducing its potential environmental impacts.

The results revealed that the pyrolysis process constituted the primary contributor to the environmental footprint of HC production, consuming the largest amount of energy and generating the most GWP. The acid treatment process also played a significant role, given the environmental burden such as acidification associated with the acid supply chain. To further investigate environmental hotspots and explore potential avenues for reducing the environmental impacts of HC manufacturing on a hypothetical industrial scale, a sensitivity analysis was performed. This analysis identified several influential factors, including the energy mix, acid type, inert gas amount, carbon yield, pyrolysis process throughput, and production scale. These findings underscore the importance of enhancing the efficiency of pyrolysis processes and considering the sustainability of upstream supply chains, such as electricity and acid, to refine HC manufacturing procedures. In this study, the treatment of bio-waste and the supply chain of precursors were treated as burden-free as these study focuses on different process design. In further research, these aspects should be considered. The use of bio-based precursors might bring the challenges of consequential environmental burden from substitute usage, challenges in consistent supply and quality, and challenges in anode processing.

This work contributes to the field by highlighting the environmental challenges and opportunities in scaling up HC production for SIBs. Unlike graphite, there is a lack of LCA studies focusing on the mass production of HC. This study found that in large-scale production, HC exhibited comparable results to graphite in terms of GWP. Electricity consumption remained the primary impact factor in the large-scale production of HC. However, the proportion of electricity consumption gradually decreased due to scale effect, while the proportion of materials and direct emissions, such as methane, increased. Therefore, addressing the treatment of exhaust gas, which can be incorporated with recovery of energy, and efficient utilization of materials becomes crucial for future industrial-scale production. One potential solution involves energy recovery through the combustion of exhaust gas to be fed back into

the pyrolysis process, combined with carbon storage technology, to simultaneously reduce emissions and energy consumption. However, the economic feasibility and environmental sustainability of such approaches require further exploration, making them a focus of future studies. Future research should focus on enhancing the sustainability of the entire HC supply chain, from bio-waste treatment to advanced anode processing techniques. Addressing these challenges will not only improve the feasibility of large-scale HC production but also position HC as a leading candidate for future sustainable battery material solutions.

In conclusion, this study provides valuable insights into the environmental implications of HC manufacturing and paves the way for future research in this field. It highlights the necessity of improving the efficiency of the pyrolysis process and ensuring a clean energy supply to mitigate the environmental impacts associated with HC production. This work is a step towards sustainable and cost-effective HC production for SIBs, contributing to informed decision-making processes in of sustainable energy storage development.

CRediT authorship contribution statement

Huiting Liu: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Manuel Baumann:** Writing – review & editing, Validation, Supervision, Methodology, Investigation, Data curation, Conceptualization. **Hyein Moon:** Writing – review & editing, Resources, Investigation, Formal analysis, Data curation, Conceptualization. **Xiang Zhang:** Writing – review & editing, Formal analysis, Data curation, Conceptualization. **Xinwei Dou:** Writing – review & editing, Investigation, Formal analysis, Conceptualization. **Maider Zarrabeitia:** Writing – review & editing, Supervision, Resources. **Eleonora Crenna:** Writing – review & editing, Validation, Methodology, Conceptualization. **Roland Hischier:** Writing – review & editing, Validation, Supervision, Resources, Methodology, Conceptualization. **Stefano Passerini:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. **Niklas von der Assen:** Supervision, Resources, Methodology, Conceptualization. **Marcel Weil:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

I have shared my data as supplemental files at the Attach File step

Acknowledgement

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2024.153410.

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