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The Role of Cycle Life on the Environmental Impact of $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ based Solid-State Batteries

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This study compares the environmental impacts of a lithium-ion battery (LiB), utilizing a lithium iron phosphate cathode, with a solid-state battery (SSB) based on a $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ garnet-structured electrolyte. It uses a hybrid life cycle assessment (LCA), according to two functional units, delivery of 50 MJ of electrical energy and kg of battery, to expand the system boundary. The results of the process LCA indicate that the environmental impact of LiBs is lower than SSBs across most environmental impact categories. Conversely, the input–output upstream global warming potential (GWP) of the LiBs, calculated by hybrid LCA, is higher than that of the SSBs. Sensitivity analysis shows that the SSB cycle life must increase from 100 to 2800 to achieve a GWP impact lower than that of LiBs and therefore outperform LiBs in this environmental impact category. The study, therefore, demonstrates that research into SSBs must be accelerated to achieve a functional and safe battery technology with a reduced impact on the environment.

1. Introduction

Electrochemical energy storage (EES) using battery technologies is ideal thanks to rapid response times, system stability, high efficiency, and low losses.[1,2] Lithium-ion batteries (LiBs) are deployed in a range of applications, from small, portable devices to storage on the grid. A laptop battery has a capacity of approximately 45–100 Wh, electric vehicles (EVs) require a capacity of up to 85 kWh and LiBs on the grid have megawatt capacities.[3] Despite their advantages and extensive uptake, LiBs suffer from state of health reduction caused by the continuous charging and discharging cycles, resulting in dissolution of the electrolyte that releases a flammable liquid which can readily ignite.[4,5] Health and safety concerns have prompted the development of new battery technologies that possess equal or superior operational characteristics. Furthermore, extensive research and technological development are underway with respect to sustainability, performance safety, durability, and cost.[6]

One optimistic line of investigation relates to solid-state battery (SSB) technologies where the liquid electrolyte of the LiB is replaced with a solid alternative, resulting in a reduced risk to health and safety.[7] Additionally, this technology has a higher storage capacity, undergoes reduced operational stress (leading to longer life cycles), operates across a broader temperature range and has reduced susceptibility to shock and vibration.[4] Research in this area has increased with the aim of implementing SSBs in EVs by 2025.[8]

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Life cycle assessment (LCA), a structured framework used to estimate the impact of a product or service on the environment, has been in use for many years[9] and has been used extensively for the determination of the environmental impacts of batteries and other EES systems.[10–14] Hybrid (H)LCA is an adaptation of the original LCA methodology which expands the system boundary by integrating both the process-based LCA and environmental input–output (EIO) LCA methodologies. This is a well-established process and is especially useful in highlighting environmental hotspots of emerging technologies prior to commercialisation.[15,16]

The aim of this study is to compare the environmental impacts of a LiB and an SSB, based on a $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ (LLZTO) garnet structured electrolyte, to determine if the safety improvements associated with SSB technologies is mirrored in their environmental impacts. Previous research[17] has shown that it is the material manufacturing phase of the battery life cycle that has a higher environmental and energy impact when compared to battery assembly; therefore, assessment of the environmental impacts of the LLZTO material structure is of paramount importance to decision-makers. The novelty of this work is two-fold; this is the first comparative assessment of the environmental impacts of a LiB and an SSB containing an electrolyte of LLZTO and as such is the first to report new cradle-to-gate environmental impacts for an LLZTO based SSB. This structure was chosen for consideration due to its stability alongside lithium metal and high current cycling.[18] Furthermore, its reliance on critical materials within the cathode structure is important with respect to their future sustainability. Second, although the LCA methodology has been extensively applied to battery technologies and while this manuscript does not aim to develop this process further, it is the first study to employ the HLCA in a comparative manner to both LiBs and SSBs, thereby expanding the system boundary to avoid truncation errors and provide a more transparent and robust result than process LCA alone.[19]

LiBs are fully commercialised, while the SSB technologies are at a low technology readiness level, and therefore caution must be taken when comparing their environmental impacts. This is due to the unrepresentative nature of the laboratory process when compared with up-scaled industrial processes.[5] Despite this, significant research into laboratory-based product development has shown that the application of HLCA at this stage is key to ensuring that environmental impacts are reduced throughout a supply chain.[19] To ensure that the early intervention for environmental impact mitigation of SSBs is representative, data is sourced from published literature and sensitivity analysis is performed.[20]

This comprehensive, comparative HLCA provides stakeholders with robust data to allow informed decisions to be made with respect to the further development of SSBs alongside LiBs for future battery architectures. As a whole, this data, the comprehensive discussion of the underlying issues that accompany it and the discussion surrounding the implications of processing and material selection offer understanding relating to the future of lithium-based battery technologies.

2. Current State-of-the-Art

2.1. LiBs vs. SSBs

All batteries consist of an anode (positive electrode) and a cathode (negative electrode), separated by a solid or liquid electrolyte. The purpose of the electrolyte is to conduct the ions produced by the chemical reaction between the anode and cathode forcing work to be done through the external circuit. Batteries also contain at least one interconnected electrochemical cell. Each cell provides a current at a given voltage over time (Δt) to depletion. Δt to depletion and output can be increased by increasing either the size of the electrode or the number of parallel connecting cells.[21]

LiBs have dominated the portable battery market for many years, replacing nickel–metal hydride batteries, which has reduced the use of rare earth oxides but has had a negative effect on the consumption of lithium, manganese, and cobalt.[22] The structure of LiBs consists of a positive electrode, usually $LiFePO_4$, on an aluminium substrate and a graphite negative electrode on a copper substrate, separated by a liquid electrolyte of lithium hexafluorophosphate, $LiPF_6$. A polymeric separator is required, along with an aluminium container and a battery management system.[23]

With respect to SSBs, current prospective materials for the anode are lithium metal, Li/V_2O_5 or V_2O_5; the solid-state electrolyte may be constructed from LiPON, $LiBO_2$ or a garnet structure, such as $Li_7La_3Zr_2O_{12}$ (LLZO). Composites of poly(vinylidene fluoride), PVdF, LiTFSI, Ketjen Black, and carbon-coated $LiFePO_4$ have been investigated, along with materials such as $LiMn_2O_4$ and V_2O_5 as potential cathode materials.[4,24]

The garnet framework, LLZO, of an SSB has been shown to have high ionic conductivity (approx. 2×10^{-4} S cm^{-1}) at room temperature and good thermal and chemical stability. The incorporation of Al, Nb, or Ta into the structure reduces the likelihood of the formation of the tetragonal phase which has lower ionic conductivity than the cubic phase. High density, with large grains, is required to optimise electrolyte performance but conventionally sintered LLZO provides both insufficient density and ionic conductivity. 96–98% densities of LLZO have been achieved through hot-pressing though this manufacturing route does not lend itself to mass production and is economically unattractive.[25]

The impact of the use phase of a battery can depend on its application, for example, high energy densities reduce the amount of fuel required for EVs and therefore, the environmental impact is reduced.[5] Though a battery does not contribute direct emissions to its life cycle impact, indirect emissions can arise from energy conversion loss, the energy required to support the weight of the battery and the carbon intensity of the electricity used to charge the battery.[26]

A battery's end of life (EOL) is reached at approximately 70–80% of its original energy capacity.[27] If there are no signs of leakage, internal short circuits or high internal impedance at this stage, reuse is a viable waste management strategy. Reuse necessitates battery disassembly (and subsequent reassembly), testing, additional packaging and electrical hardware.[28]

Bernardes et al.[29] report four disposal routes for batteries; landfill, stabilisation, incineration, and recycling. If household batteries are disposed of in municipal household waste, they may be sent to landfill. Stabilisation aims to treat batteries to prevent the leaching of metals into the environment prior to landfilling but this is a costly process. The incineration of batteries may take place as an alternative to landfilling through this process leads to the emissions of halogen, mercury, lead, and dioxins to the environment.[29] Hydrometallurgical and pyrometallurgical recycling processes allow metals to be recovered from batteries. The pyrometallurgical process involves incineration, smelting, drossing, sintering, melting, and high-temperature reactions in the gas phase. The process results in waste gases and flue dusts which contain halogens leading to potential human health issues. The human toxicity potential of the hydrometallurgy recovery process is lower than that of the pyrometallurgical recovery process, but the required solutions for the hydrometallurgical process are toxic and corrosive. Furthermore, recycling LiBs by pyrometallurgy and hydrometallurgy leads to the production if an untreatable residue which must be landfilled.[30]

2.2. LCAs of Batteries

An abundance of published LiB LCA studies is available, while, due to the infancy of the technology, published LCAs of SSBs are limited in number. A review of relevant studies is provided here. Ellingsen et al.[31] assessed the LCA of LiBs and found vast disparity in the results. For the production phase alone, the lowest result published was 38 kg CO_2 kWh^{-1} compared to the highest result of 356 kg CO_2 kWh^{-1}. The main source of this variance is from the reported energy requirements in the cell manufacture and battery assembly. The group report that, due to the guarded nature of this industry, primary data is hard to come by and consequently published work uses a mixture of estimates, industry reports, previous studies, and primary data for the life cycle inventory (LCI).[31] A variation in the type and amount of material required for production was also highlighted by Ellingsen et al.[31] While most authors agree a similar requirement of material for graphite anodes the amount of cathode material reported by Majeau-Bettez et al.[23] leads to a much high final result. It was also shown that the type of binder used in cathode production can have an adverse impact on the final result.

With respect to the LCA of SBBs, the use of an LCO.LLZ cathode with an LLZ solid-state electrolyte has been investigated by Troy et al.[5] to determine the environmental impacts of a pouch bag housed SSB produced in a laboratory. Overall, the on-site electrical energy requirements were found to account for almost 99% of the global warming potential (GWP) (kg CO_2-eq) impact in this study, and with respect to the manufacturing process, the electrolyte followed by cathode production provided the highest contribution to the GWP impact category. This data was then modified to determine the impacts of production in an idealised laboratory and an industrial process. For the idealised laboratory result, efficiency enhancements for each processing step were assumed and for the industrial manufacturing process, assumptions are made on energy use, a reduced material input and release of solvents. With respect to the industrial manufacturing steps, assumptions are made based on the energy efficiency, reduced release of solvents and material input.[5]

Lastoskie and Dai[32] compare the environmental impacts of two SSB manufacturing processes; lamination and thin-film vacuum vapour deposition. Specifically, for the laminating process, various cathode materials such as lithium cobalt oxide and lithium manganese oxide were studied in conjunction with graphite anodes. With respect to the thin-film vacuum deposition process, these materials and additional alternatives such as lithium vanadium oxide and silver vanadium oxide were also investigated. Lithium vanadium oxide SSBs had the lowest environmental impact across eight mid-point environmental impact categories.[32] Keshavarzmohammadian et al.[33] studied the environmental impacts of sulphur-based solid-state lithium batteries for use in EVs, which solve the safety and technological challenges relating to liquid electrolyte alternatives.[34] Their results attribute the highest environmental impact of the system on the different assembly processes required. Overall, the operation of the clean dry-room, which is required for pyrite battery manufacturing, contributes to 73% of the GWP.[33]

The inclusion of the battery use phase differs from study to study. For example, Zackrisson et al.[26] studied the impact of battery use in an EV modelled as the loss of electricity as a function of battery lifetime plus the additional fuel required to carry the battery during service.[26] Other strategies include that implemented by Majeau-Bettez et al.[23] who estimated the electrical energy waste caused by the internal resistance of a battery,[23] while Hendrickson et al.[35] deemed the environmental impacts of the use phase of EVs to be the same, irrespective of the EOL management scenario and therefore omitted this phase from their study. Similarly, Raugei and Winfield[36] omitted the use phase of the battery from their system boundary as it was not deemed relevant to the goal and scope of the study.[36] These differences in the applied scope of any LCA may lead to high variations in the final results and therefore care must be taken in their comparisons.[35]

Hendrickson et al.[35] employ the functional unit, per kg of battery to present the embodied energy results of $LiMn_2O_4$, $LiFePO_4$ (LFP), and $LiNi_{0.4}Mn_{0.4}Co_{0.2}O_2$ batteries.[35] Similarly, Dunn et al.[17] also made use of this functional unit in their study which addresses a wide range of battery material structures.[17] Majeau-Bettez et al.[23] provide three alternative functional units in their investigation into the LCA of lithium-ion and nickel–metal hydride batteries; battery storage, mass and nominal capacity. The results of each battery's LCA changed dramatically based on the functional unit. For example, the GWP of a nickel–metal hydride battery per kg provides the lowest result of the three battery types compared but when the functional unit is changed to energy storage or capacity the result is the highest of the three analysed, due to the lower energy density of this type of battery. By implementing three functional units, this research provides improved usability and easier comparison with previous research.[23]

LCAs of other battery technologies are available such as nickel–metal hydride batteries, nickel-cobalt-manganese oxide batteries, lithium manganese oxide, lead–acid batteries and sodium-ion batteries.[17,23,35,37,38] From an environmental perspective, the toxicity of lead is well known and the mining of nickel has led to local historical environmental implications such as acidification, heavy metal contamination, and a reduction in biodiversity.[39] As a conflict mineral, the use of cobalt in battery technologies can have a severe impact on local social issues. In the Democratic Republic of Congo, these issues include cobalt found in blood and urine and damage to DNA.[40]

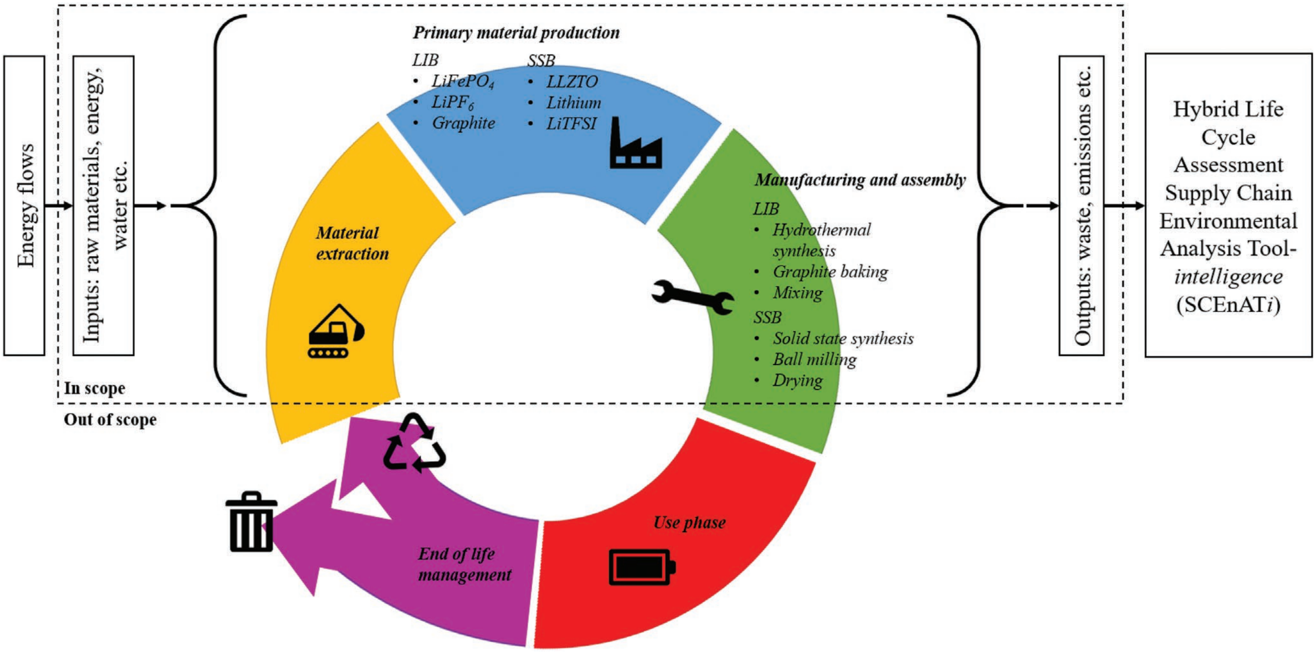

Figure 1. System boundary applied to the comparative hybrid LCA of a LiB cell and a SSB cell, from cradle-to-gate incorporating raw materials and energy inputs, primary material production and the manufacturing process.

From an economic perspective, lithium, nickel, cobalt, manganese, zinc, and rare earth elements (REEs) are classed as economically strategic materials and therefore, LiB may be preferentially recycled over sodium-ion or lead–acid technologies.[41]

This array of research into the environmental impacts of different battery technologies provides a plethora of data, therefore the results provided in this manuscript aim to support previous findings whilst expanding the knowledge of the environmental impacts of SSBs in comparison to LiBs.

3. Methodology

This LCA was performed according to the requirements of BS EN ISO 14040:2006, Environmental management- LCA- Principles and framework the results.[9] The LCA procedure is widely published and requires the completion of four processes; definition of the goal and scope; inventory analysis; impact assessment and interpretation.[9] Each of these procedures is outlined below.

3.1. Goal and Scope Definition

The aim of this investigation is to determine the environmental impacts of a single LiB cell and a single SSB cell, using the HLCA methodology, according to the system boundary shown in **Figure 1**. The functional unit, which provides a reference point during the data collection process, has a significant influence on the LCA results and should be chosen as an accurate representation of the properties of the system under investigation.[19,42] As batteries have different efficiencies during the use phase, a functional unit based on the mass of the battery my not always be relevant[43] and therefore, in this study, two functional units are applied, the environmental impact of 1) delivery of 50 MJ of electrical energy to the device and 2) kg of battery. The use of two functional units in this study allows the results to be compared with other studies more easily.[17,23,35] Despite this, direct comparison must be used with caution depending on the life cycle impact assessment (LCIA) methodology employed by each study.

The energy density of the LiB was given as 99.59 Wh kg^{-1}[44] with a prediction of 2500 cycles[45] and that of the SSB was given as 314 Wh kg^{-1}[18] at 100 cycles,[24] therefore at 80% depth-of-discharge (DoD) the lifetime specific energy capacities were calculated as 718 MJ kg^{-1} for the LiB and 90.4 MJ kg^{-1} for the SSB.

A cradle-to-gate approach was implemented for this study, as shown in Figure 1. The use phase was omitted as the aim of the research is to determine the environmental impact of the SSB in comparison to the LiB at the battery cell stage.[36] As the EOL management of SSB is as yet unknown, this phase of the two battery life cycles was also omitted from the system boundary. Previous work by the authors has evaluated the EOL management of capacitors using the Ecoinvent dataset for the treatment of LiBs as a proxy measure. These results show that EOL management by incineration leads to the highest GWP impact when compared to hydrometallurgical or pyrometallurgical processes.[15]

3.2. Inventory Analysis

LCIA requires the inputs, outputs, and environmental impacts of the product under scrutiny to be gathered.[9] Due to the lack of primary industry data, the LCI of the LiB was provided by[44]

Table 1. List of the environmental impact categories, their corresponding units and LCIA methodologies used in this research.

Environmental impact category	Abbreviation	Unit	LCIA Method
Global warming potential	GWP100	kg CO_2-eq	ReCiPe Midpoint (H) v1.13
Freshwater ecotoxicity potential	FETPinf	kg 1,4-DCB-eq	
Human toxicity potential	HTPinf	kg 1,4-DCB-eq	
Marine ecotoxicity potential	METPinf	kg 1,4-DB-eq	
Terrestrial ecotoxicity potential	TETPinf	kg 1,4-DCB-eq	
Freshwater eutrophication	FEP	kg P-eq	
Marine eutrophication	MEP	kg N-eq	
Terrestrial acidification potential	TAP100	kg SO_2-eq	
Cumulative energy demand	CED	MJ-eq	Cumulative energy demand

while that of the SSB was derived from both literature[24,25,46,47] and on-going Li-ion projects at the Functional Materials and Devices Laboratory, Materials Science and Engineering, University of Sheffield. A full outline of each battery LCI is provided in Tables S1–S21, Supporting Information. It was assumed that the thermal and electrical energy requirements of each battery type were equal (using published data)[44] except in the production of the SSB electrolyte, for which the thermal and electrical energy requirements were calculated separately (as shown in Table S22, Supporting Information) to account for the high energy requirements of ceramic processing.[24,25,46,47] Finally, published data was also used to account for the transportation and infrastructure requirements.[44]

3.3. Impact Assessment

HLCA is an integration of two key LCA modelling methodologies: Process LCA and EIO.[16] The Process LCA is a bottom-up methodology mainly used to evaluate the environmental impacts of products with clearly defined supply chains. The distinct inputs into the supply chain are identified (see Section 3.2) which allows the emission intensities relating to each input to be sourced and therefore the overall environmental impact can be discretely calculated. The Process LCA was employed to calculate the environmental impact using Equation (1) by attributing the chosen environmental impacts categories (see **Table 1**) to each data point highlighted in the LCI.[19]

$$\text{Environmental Impact} = \sum_{i=1}^{n} A_{\mathrm{p}(i)} \times E_{\mathrm{p}(i)} \tag{1}$$

The inputs (i) into the supply chain are represented by A_p, according to the constraints of the system boundary provided in Figure 1. These inputs include the bill of quantities of data on the extraction of raw material, energy usage, production, transportation, and infrastructure. The total number of inputs (i) is given by n and the emission intensity of the environmental impact categories (Table 1) are given by E_p for each input (i) into the supply chain. The final units of this calculation relate to the chosen environmental impact category (E_p), a selection of which are provided in Table 1.[19]

It has been shown that the variance in the accuracy caused by implementing a system boundary can be higher than 50% and therefore the Process LCA methodology is not capable of determining the environmental impacts of global and complex supply chains. As such, the EIO methodology (a top-down methodology) utilises Multi-Regional Input–Output data, combined with sector emissions intensities to calculate a broader result by capturing the indirect inputs into the supply chain. The limitations relating to this methodology include sector aggregation, quantity conversions and proportionality and homogeneity assumptions. Equation (2) gives the calculation for the direct and indirect environmental impact of the supply chain using the EIO methodology in its simplest form.[19]

$$\begin{array}{l}\text{Direct and indirect sectoral} = E_{\mathrm{io}} \cdot (I - A)^{-1} \cdot y \\ \text{environmental impact}\end{array} \tag{2}$$

where A_io gives the technical coefficient IO matrix, (I) represents the identity matrix, E_io signifies the emission intensity of the environmental impact categories and y represents the final demand. The total (direct and indirect) emissions intensities of each industry associated with the supply chain are given as $E_\mathrm{io} \cdot (I - A)^{-1}$. Again, the final units of this calculation relate to the chosen environmental impact category (E_io).[19]

By integrating these two methodologies, HLCA provides a robust and complete system boundary that incorporates the whole supply chain without double counting.[19] Thus, the total environmental impact (direct and indirect) is calculated by the HLCA methodology is given in Equation (3).[48]

$$\text{Emissions impact} = \begin{bmatrix} E_p & 0 \\ 0 & E_{io} \end{bmatrix} \begin{bmatrix} A_p & -C_d \\ -C_u & I - A_{io} \end{bmatrix}^{-1} \begin{bmatrix} y \\ 0 \end{bmatrix} \tag{3}$$

where E_p relates to the process inventory environmental extension matrix, E_io corresponds to the multi-regional input–output environmental extension matrix for each of the chosen environmental impact categories, A_p denotes the square matrix representation for the process LCA inventory and A_io provides the input–output technology coefficient matrix. I represents the identity matrix, C_u gives the upstream cut-off matrix, C_d the downstream cut-off matrix, while $\begin{bmatrix} y \\ 0 \end{bmatrix}$ denotes the functional unit column matrix. The final units of this calculation relate to the chosen environmental impact category (E_io).[19,48]

In the HLCA model, the element C_u in the algebra matrix which is the upstream cut-off matrix can only be calculated using the unit cost (i.e., £/kg) of the individual process inventory (e.g., $LiFePO_4$), which is already captured within the process matrix, A_p. The unit cost is then multiplied by the input (i.e., the physical quantity) of the process (e.g., $LiFePO_4$ as in the case of this study), yielding the total cost (£) of the process (i.e.,[£/kg] × [kg]) and represents the cost of $LiFePO_4$ that is required to produce the battery based on the functional unit under consideration. The costs obtained are integrated using only the missing inputs from the process component of the HLCA, to avoid double counting. The use of cost estimates is essential, given that impacts via the EIO model are calculated on the basis of transforming financial flows to environmental flows.[49,50] For detailed steps for calculating the C_u matrix within an HLCA framework, readers are referred to Wiedmann et al.,[51] Suh and Huppes,[52] and Ibn-Mohammed et al.[19,53]

The Supply Chain Environmental Analysis Tool-*intelligence* (SCEnAT*i*) whose underlying algorithm and calculation steps are informed by Equation (3), was used to perform the environmental lifecycle modelling, according to the functional unit of kg of battery.[48,54] The tool enables a chosen supply chain to be mapped and requires the following inputs: the sector from which the input is sourced (chosen from a dropdown list) and the associated classification (chosen from a dropdown list), the quantity of the input in line with the chosen functional unit (given by the LCI), the emissions intensity of the input (provided by databases such as Ecoinvent)[55] and the unit cost of the item.[48,54] Finally, the modeller must choose those inputs missing from the Process LCA to be incorporated into the HLCA. The missing inputs applied to the system boundary are given in Table S23, Supporting Information. These inputs ensure the balance and completeness of Equation (3) for adequate implementation. Only the results for the GWP are provided as this result was the dominant impact when normalised and compared to the other impact categories studied. The results are then broken down into the process emissions (as per the Process LCA) and the "upstream indirect emissions"; the indirect impacts are aggregated across 18 economic sectors. For this study, due to a lack of robust data, manufacturing facilities were omitted from the hybrid analysis and their cost was assumed to be included in that of the material.

To compare the environmental impacts, this study uses the ReCiPe Midpoint (H) v1.13 and cumulative energy demand (CED) LCIA methodologies, assembled using the Ecoinvent database,[55] to provide the environmental impact category data appropriate to the scope of the study, including the impact of associated emissions. Where any publically available data was lacking, published literature was sought and guidelines were followed relating to the chemical characteristics or functionally similar materials.[56] Table 1 outlines the chosen impact categories, their units and methodologies.

4. Results and Discussion

4.1. Process LCA

This section provides analysis and detailed discussion of the environmental impacts of the Process LCA applied from cradle-to-gate to for each battery type. Table 1 relates the details of the environmental impact categories and **Table 2** outlines the results of the process LCA within the constraints of the two chosen functional units; 1) the environmental impact per 50 MJ of electrical energy delivered and 2) the environmental impact per kg of battery.

4.1.1. Material Impact

GWP calculates the change in the earth's temperature following the emissions of greenhouse gases over a given 100-year time horizon and is measured as tonnes or kg CO_2-equivalent (eq).[57] Based on the functional unit delivery of 50 MJ of electrical energy, the GWP of the LiB is 1.60 kg CO_2-eq, compared to 43.76 kg CO_2-eq for the SSB. In terms of the functional unit kg battery cell, the results reveal that the GWP of the LiB is 22.97 kg CO_2-eq and that of the SSB is 79.11 kg CO_2-eq. When this result is benchmarked against other published literature (given as 22 kg CO_2-eq kg^{-1}),[23] the result per kg of battery is very similar. The main discrepancy according to the functional unit for the delivery of 50 MJ of electrical energy relates to the assumed cycle life of the LiB. Majeau-Bettez et al.[23] use a cycle life of 6000 cycles to support their lithium iron phosphate

Table 2. The environmental impact of the LiB and SSB technologies for each of the environmental impact categories listed in Table 1.

Environmental impact category	Unit	LiB		SSB	
		50 MJ	kg	50 MJ	kg
Global warming potential	kg CO_2-eq	1.60	22.97	43.76	79.11
Freshwater ecotoxicity potential	kg 1,4-DCB-eq	0.39	5.67	0.98	1.77
Human toxicity potential	kg 1,4-DCB-eq	2.98	42.76	17.09	30.89
Marine ecotoxicity potential	kg 1,4-DB-eq	0.35	5.08	0.86	1.56
Terrestrial ecotoxicity potential	kg 1,4-DCB-eq	4.88E-04	7.00E-03	5.37E-03	9.70E-03
Freshwater eutrophication	kg P-eq	1.37E-03	0.02	0.02	0.04
Marine eutrophication	kg N-eq	5.30E-03	7.61E-03	1.00E-02	1.81E-02
Terrestrial acidification potential	kg SO_2-eq	0.01	0.17	0.18	0.32
Cumulative energy demand	MJ-eq	28.46	408.69	698.45	1262.79

battery, while this research uses a cycle life of 2500 cycles[45] as this provides a more up to date reference. In line with the availability of other published results, the individual impact of the cathode material can be benchmarked. In this case, the results of this study give the GWP impact of the LFP LiB cathode material as 12.65 kg CO_2-eq kg^{-1}, compared to only 3 kg CO_2-eq kg^{-1} given by Dunn et al.[17] These differences may be attributed to differences in the scope of the study, battery specification or other supply chain inputs.[35] In the absence of benchmarking data for the SSB, and therefore comparing it to the LiB, it is clear that at this early stage of development of the SSB the GWP impact far outweighs that of the commercialised LiB.

The FETPinft, HTPinf, METPinft, and TETPinft relate to the toxicity impact of these materials and measure the emissions of 1,4-Dicholorobenzene equivalent to freshwater, urban air, seawater, and industrial soil respectively.[58] The results of this study show that the HTPinf for the LiB is 42.76 kg 1,4-DCB-eq per kg compared to 66 kg 1,4-DCB-eq per kg, provided by previously published results.[23] Comparatively, the results of the FETPinft, METPinft, and TETPinft environmental impact categories are much higher in this study than in previously published research.[23] These toxicity impacts can be attributed to the use of copper in the anode current collector. With respect to the human toxicity of copper, low doses of copper in humans can lead to deficiencies, but high doses can lead to adverse effects, for example, liver toxicity.[59] Furthermore, research has reported increased levels of arsenic in the urine of those living close to copper smelters.[60] Copper has been found to be toxic to aquatic animals due to the effect of the metal on the water such as its hardness, pH, and suspended solids.[61] Overall, the SSB shows a lower environmental impact per kg of battery for the HTPinf, FETPinft and METPinft environmental impact categories compared to the LiB and only a very small increase is observed in the TETPinf environmental impact category. Therefore, from a toxicity point of view, the SSB is superior to the LiB.

The CED or embodied energy calculates the primary energy accumulated during the life cycle of a product,[62] it is measured in MJ-eq and is calculated as the sum of untransformed energy sources, that is, fossil fuels.[19] The embodied energy of LFP LiBs has been reported to range from 64.4 to 541 MJ kg^{-1};[35] this study provides a result of 409 MJ kg^{-1}. Furthermore, the CED of the SSB was found to be 1263 MJ kg^{-1}. The highest contributor to this result is the electricity requirement for the manufacturing of the SSB electrolyte, discussed in more detail in section 4.1.2. Furthermore, the use of lanthanum hydroxide in the structure leads to the second-highest contribution to the environmental impact across all categories studied. Lanthanum is an REE, the mining and downstream processing requirements of which are energy-intensive, leading to a high CED. Furthermore, as the quality of the ore decreases, energy consumption and the related CO_2 emissions increase.[63] REEs are listed as a critical material using numerous methodologies.[64–66] The separation and refining processes of rare earth elements are energy and water-intensive, require extensive chemical use and result in the emission of heavy metals, SO_2, and dust among other pollutants.[63] Regardless of these environmental impacts, the use of REEs is continuing to increase.[67,68]

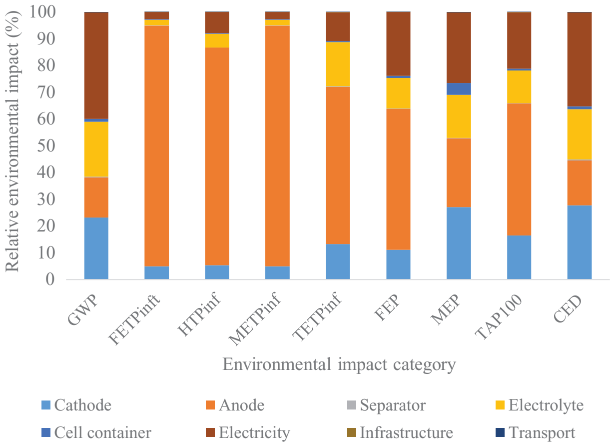

Figure 2. The percentage contribution of each component required in the production of a LiB cell for each environmental impact categories listed in Table 1.

Eutrophication concerns the accumulation of chemical nutrients in ecosystems which ultimately leads to the disproportionate growth of plant life thereby reducing the quality of the water and the populations of animals. Marine eutrophication is expressed as kg N-eq and freshwater eutrophication is expressed as kg P-eq as nitrates, nitrogen oxide, and phosphorous all have an adverse effect on eutrophication.[57] By means of comparison, this study shows that both the freshwater and marine eutrophication impacts are higher for the SSB than the LiB, independent of the functional unit applied. In turn, when compared to previously published results with respect to per kg of battery, the results provided for the LiB in this study are lower.[23] These results are mirrored in the results for terrestrial acidification, which is the measure of the effect of acid rain on different ecosystems.[57]

The contribution of each component in the production of LiBs and SSBs to the overall environmental impact of the twelve impact categories listed in Table 1 are shown in **Figures 2** and **3**, respectively. The indicators are normalised to

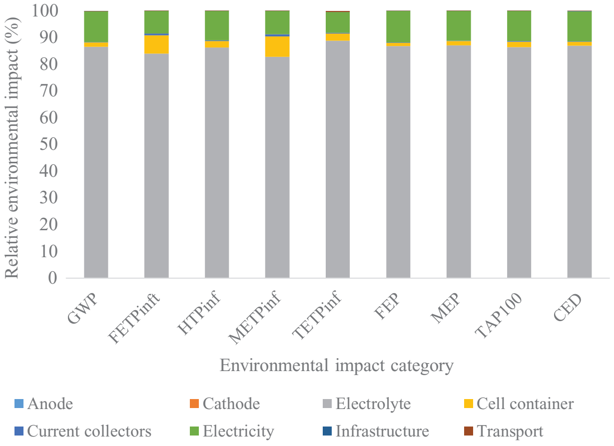

Figure 3. The percentage contribution of each component required in the production of an SSB cell for each environmental impact category listed in Table 1.

provide an absolute indicator of 100% for each value, shown in Figure 2 for the LiB and Figure 3 for the SSB.[19]

By breaking down the total of each environmental impact for both battery technologies, it is possible to determine where the carbon hot-spots lie within the structures. Figure 2 shows that the majority of the environmental impact of the LiB is split across the cathode, anode, electrolyte, and electricity use in the manufacturing stage. The impact of the LiB anode is dominant for the FETPinft, HTPinf, METPinft, TETPinft, FEP, and TAP100 environmental impact categories. Conversely, Figure 3 shows that the electricity requirements for manufacturing lead to the highest percentage contribution for the SSB. This is discussed in more detail in section 4.1.2.

Overall, when the functional unit for the delivery of 50 MJ of electrical energy is applied, the environmental impacts are lower for the LiB. When the functional unit of kg of battery is applied, the SSB has a lower environmental impact in the case of those environmental impact categories concerned with (eco)toxicity. The effect of the cycle life and DoD, of the SSB is considered in the sensitivity analysis to understand how the functionality of the architecture must be improved to out-perform the LiB across all environmental impact categories.

4.1.2. Impact of Energy Consumption Due to Fabrication

It is the use of the lithium iron phosphate and *N*-methyl-2-pyrrolidone solvent in the LiB cathode that leads to the high environmental impact across the GWP and CED environmental impact categories. These environmental impact categories are mainly affected by the electricity requirements of the electrolyte production. Zackrisson et al.[26] show that when the NMP solvent is changed to water the GWP impact is reduced by approximately 23%.[26]

This study uses the global electricity, medium voltage[kWh] dataset from Ecoinvent to provide an average global impact for the electricity use in battery production.[55] Previous research has shown how the location of the electrical energy source can affect the overall environmental impact of a component. For example, electricity sourced from China was found to have an impact of over ten times larger than that sourced from France.[15] A reduction in the GWP of electricity generation can be attributed to decarbonisation of national grids. With respect to the UK, the key factors to achieving a reduction the GWP are attributed to a reduction in demand, an increase in gas utilisation and the implementation of renewable energy sources such as wind.[69] Comparatively, in France, the road to decarbonisation is paved by nuclear power generation.[70] Therefore, it can be concluded that the impact of the electricity use in the electrolyte (and, in fact, the rest of the battery) could either decrease or increase according to the chosen manufacturing location.

Overall, the main contributors to the environmental impact of the SSB are the electrolyte and the electricity usage in production. 86.5% of the GWP impact of the SSB can be attributed to the electrolyte and 11.56% to electricity use, as shown in Figure 3. On further inspection, electrolyte impact is dominated by the energy requirements associated with the manufacturing process, for example, electricity used for sintering. The ceramic industry is known to be an energy-intensive sector, in this case, temperatures of 900 °C are required for the heating of the electrolyte constituents and 1140 °C is required to complete the sintering process.[25] Therefore, to reduce the environmental impact of the electrolyte (and consequently that of the SSB technology) research is required into reducing the processing parameters required for the densification of the ceramic. Cold sintering, which aims to produce dense ceramic materials below 200 °C, is a line of investigation that is currently underway to determine if the energy intensity of LLZTO production can be reduced. The process employs a transitory, usually liquid, phase to allow mass transfer to produce dense ceramics using uniaxial pressure. The transitory phase usually evaporates during the cold sintering process, driving densification by solution–precipitation.[71]

Troy et al.[5] also show a high contribution to the environmental impact is attributed to the electricity requirements relating to the manufacturing phase of an SSB. Particularly, they show the percentage contribution of the electricity requirements for the production of the LLZTO electrolyte to be 80.8%, which aligns with the findings of this study at 86.5%.[5] Sulphur-based SSBs are suitable for mass production, offer another possible alternative to LIBs and do not rely on the sintering process during manufacturing. Despite this, the electricity required to operate the clean-dry room leads to a significant environmental impact across the impact categories studied.[33] Therefore, in all cases, due to the high impact of electricity use for SSB manufacturing, it is prudent to consider renewable sources of electricity generation to reduce the overall environmental impact.[15]

While for the majority of the chosen environmental impact categories the results relating to a kg of SSB are higher than a kg of LiB, the FETPinf, HTPinf, and METinf environmental indicators are higher for the LiB. These categories relate to the toxicity of the battery types and, as discussed earlier in this section, this high impact is caused by the use of copper in the anode current collector. Therefore, from the point of view of the decision-maker, when comparing the environmental impacts of these battery types using this functional unit, a decision must be made to determine which environmental impact categories are the most important.

4.2. IO LCA (Upstream Environmental Impact)

SCEnAT*i* allows for the input-output upstream GWP of each battery type to be reported separately, as such, **Figure 4** provides the results of the IO components of the HLCA based on the functional unit of kg of battery. Overall, the GWP upstream emissions of the LiB were found to be 10.86 kg CO_2-eq kg^{-1} and that of the SSB technology, 6.44 kg CO_2-eq kg^{-1}. As outlined above, SCEnAT*i* requires the unit cost of each supply chain input to be supplied into the model. Due to a lack of primary data, arbitrary costs for 1 kg of each battery type were calculated using commercial suppliers on the internet. The total cost of the supply chain inputs for 1 kg of LiB was calculated to be approximately £369 and that of the SSB, to be approximately £367. Despite the relatively low cost of the water required for manufacturing the LiB, this leads to the highest overall cost due to the large amount utilized. In comparison, it is the cost of

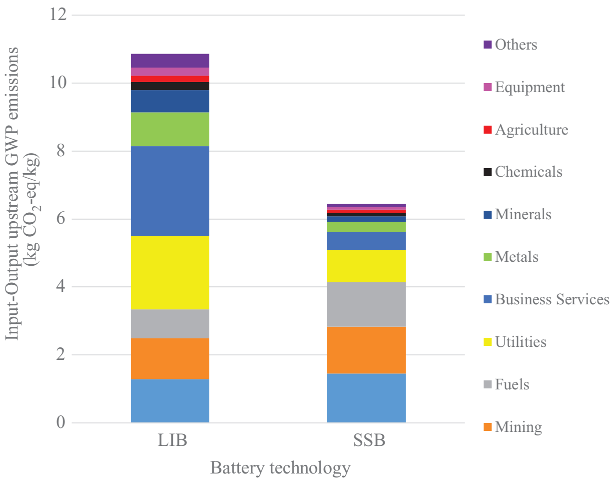

Figure 4. The input–output upstream GWP emissions of the LiB and SSB battery technologies given by the HLCA methodology for each economic sector. NB: "Others" relates to those sectors contributing less than 1% to the overall result.

the electricity required to manufacture the solid-state electrolyte that overshadows the costs of the other inputs in the SSB.

For the LiB technology, the "Business Services" economic sector has the highest percentage contribution at 23% of the total impact and for the SSB technology, the "Transport and Communication" economic sector has the highest percentage contribution to the total impact, also at 23%. Those economic sectors contributing less than 1% to the total result were collated and referenced as "Others". These impacts can be broken down further to show that the LiB cathode and assembly have the highest impact on the upstream emissions, while it is the electrolyte of the SSB that has the highest percentage contribution. This result is affected by the emissions intensities of each industry associated with the supply chain, the cost of the input and the "additional impacts" supplied by the modeller. Therefore, this result may differ if different supply chain costs were applied and different "additional inputs" were modelled. Overall, the upstream environmental impacts of the LiB across the economic sectors considered are higher than that of the SSB technology due to their higher cost of constituent materials and supply chain requirements.

4.3. Sensitivity Analysis

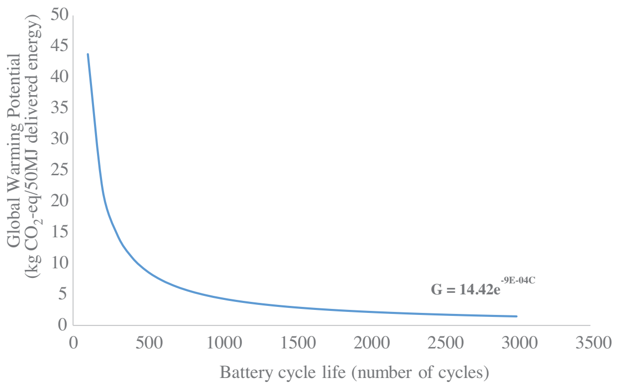

Figure 5. The GWP of the SSB technology as the cycle life is increased from 100 cycles to 3000 cycles in 100 cycle increments at a specific energy of 314 Wh kg^{-1} at 80% DoD.

Due to the low technology readiness level of the SSBs, sensitivity analysis was performed to ensure the result is representative.[20] To determine the GWP impact of the SSB in the event of an increase in cycle life, the LCA was performed from 100 to 3000 cycles at 100 cycle intervals, assuming a specific energy of 314 Wh kg^{-1} at 80% DoD.[18] The cycle life pertains to the number of times a battery can be charged and discharged prior to failure and is often influenced by DoD which is a function of the total battery capacity level that has been utilized. Specific energy relates to the amount of energy stored per unit weight. The results of the sensitivity analysis are shown in **Figure 5**, where the GWP of the SSB technology is assessed according to an increase in cycle life of the battery; the cycle life was increased from 100 cycles to 3000 cycles in incremental steps of 100 cycle. The results show that the GWP impact decreases exponentially as the cycle life is increased. As shown above, a LiB with a predicted cycle life of 2500 cycles has a GWP impact of 1.60 kg CO_2 50 MJ^{-1}, therefore to reduce the environmental impact of the SSB below that of the LiB, the technology would have to reach 2800 cycles, this results in a GWP impact of 1.56 kg CO_2-eq 50 MJ^{-1} delivered energy. The equation of the resulting curve ($G = 14.42e^{-9E\text{-}04C}$) indicates that an increase from 100 cycles will significantly reduce the GWP of the battery, but further improvement will suffer from diminishing returns.

Furthermore, to determine the impact of the DoD on the GWP of the SSB technology, the DoD was altered from 30–100% across 100, 500, 100, 1500, and 2000 cycles. These results are shown in **Figure 6** which indicates that as the cycle life of the SSB technology increases, the GWP is less affected by the DoD of the battery. Therefore, this research shows that by increasing the cycle life, the environmental impact of the SSB battery is lower than that of the LiB. Schnell et al.[18] point out that changing an LLZ based electrolyte to one based on $Li_{1.3}Al_{0.3}Ti_{1.78}(PO_4)_3$ could increase the energy density of the battery to 405 Wh kg^{-1}. This provides scope for further work into how

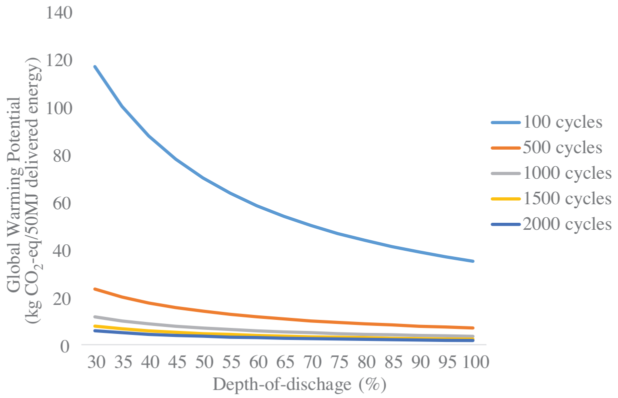

Figure 6. The GWP of the SSB technology as the DoD is increased from 30% to 100% across 100, 500, 1000, 1500, and 2000 cycles at a specific energy of 314 Wh kg^{-1}.

the SSB structure can be optimised for both performance and reduced environmental impact.

4.4. Material and Process Selection

As criticality becomes increasingly important in a circular economy,[72] it is necessary to consider the materials chosen for battery technologies. As mentioned above, the move from nickel–metal hydride batteries to LiBs, has reduced the impact of rare earth oxide use (classified as critical raw materials by the European Union[65]) but increased the use of other materials, particularly lithium.[22] According to the EU's methodology, lithium is not yet classed as critical but as approximately 2 million EV LIBs will reach their EOL in the US and China alone in 2020, approximately 20 000 tonnes of LIB cathodes will become waste. These materials must be recycled to avoid the generation of hazardous waste, reduce environmental pollution, ensure the availability of lithium sources into the future, and reduce raw material costs.[73,74] Comparatively, the SSB structure using the LLZTO returns to a reliance on critical REEs, specifically lanthanum. Although the LLZTO is not the only available SSB structure, alternatives such sulphide based systems (Li_4SnS_4-Li_3SbS_4)[75] and lithium niobate cathodes with $Li_{10}GeP_2S_{12}$ solid electrolytes[76] also rely on critical materials such as antimony and germanium. Therefore, it is paramount for the environmental impacts of different SSB structures to be compared prior to their commercialisation to ensure that the technology with the lowest environmental impact is preferentially manufactured.

With respect to manufacturing techniques, when the lamination and thin-film vacuum vapour deposition techniques are compared in the production of $LiCoO_2$, $LiMn_2O_4$, and $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ SSBs, those produced by the vapour deposition method had a lower CED and GWP impact; though this pattern was not observed across the remaining environmental impact categories studied.[32] 96–98% densities of LLZO have been achieved through the hot-pressing technique. Unfortunately, this manufacturing route does not lend itself to continuous mass production and is economically unattractive.[25] SSB structures containing an LLZTO/polyethylene oxide electrolyte have been manufactured using a solvent-free method in a Haake Rheomixer chamber. This resulted in a high speed, efficient and large-scale manufacturing process.[77] Furthermore, it has been recognised that further investigation is required to reduce the cost and difficulties relating to the compatibility of the LLZTO electrolyte with the lithium metal anode.[78]

As outlined above, the ceramic industry is inherently energy-intensive due to the high temperatures required for sintering, therefore extensive research is ongoing into cold sintering. This process not only reduces the manufacturing temperature but also the run time from hours to minutes.[71] Therefore, the cold sintering process provides a useful mechanism to reduce the environmental impact of the LLZTO electrolyte in the SSB.

4.5. Limitations and Future Work

The main limitations of this research relate to the lack of primary data for each of the battery technologies and therefore the life cycle inventories were established from published literature. This limitation can only be completely overcome through the use of primary data from battery manufacturers which is difficult to obtain due to its confidential nature.[31] Furthermore, the low technology readiness level of the SSB technology limits the output of this study. To reduce the uncertainty that accompanies this limitation, sensitivity analysis was performed to determine the environmental impact of this battery type with an increased cycle life.[20] The HLCA methodology presents a level of subjectivity in that it is the responsibility of the modeller to decide which of the missing supply chain inputs are added into the SCEnAT*i* decision support tool. Future work in this the area of battery HLCA must consider the requirements of the additional inputs, thereby reducing the level of subjectivity.

5. Conclusion

This study is the first to present a comparative assessment of the environmental impacts of a LiB and an SSB containing an electrolyte of LLZTO using the hybrid LCA methodology. The study applies two functional units; the delivery of 50 MJ of electrical energy and kg of battery, to allow for a robust result to be provided. Furthermore, sensitivity analysis is performed to determine how the cycle life of the SSB technology affects the overall environmental impact.

With a functional unit of 50 MJ delivered energy, the environmental impact of the LiB is lower across all of the environmental impact categories studied. While, according to a kg of battery technology, the environmental impact of the SSB is higher across all environmental impact categories except the FETPinf, HTPinf, and METPinf, for which the LiB has a higher environmental impact. The upstream impacts of the LiB were slightly higher than that of the SSB technology and, through sensitivity analysis, it was shown that the GWP impact of the SSB technology is lower than that of the LiB at 2800 cycles.

With these results in mind, stakeholders must determine which environmental impact categories are the most important and progress must be made to increase the number of cycles of SSBs to provide a functional and safe battery with reduced environmental impact. Furthermore, the environmental impact of the SSB technology can be lowered through a reduction of the energy requirements needed for sintering. To date, the most promising process to achieve this is through the development of the cold sintering process. Alternative SSB material structures also exist, such as sulphide based systems based on Li_4SnS_4-Li_3-SbS_4[75] and lithium niobate cathodes with $Li_{10}GeP_2S_{12}$ solid electrolytes[76] and therefore future work should also concentrate on determining which SSB structure leads to the lowest environmental impact.

Battery technologies and manufacturing processes are continually improving and therefore it is key to ensure that future HLCA research considers systems at low technology readiness levels to ensure that environmental impacts can be designed out of new structures, rather than allowing for impacts to be generated.[19] This is particularly crucial for batteries and other energy storage systems in a society where energy demand is increasing.[79,80]

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

energy storage, hybrid life-cycle assessment, lithium-ion batteries, solid-state batteries, sustainability

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[1] H. Chen, T. N. Cong, W. Yang, C. Tan, Y. Li, Y. Ding, *Prog. Nat. Sci.* **2009**, *19*, 3.
[2] S. H. Lee, J. Y. Hwang, J. Ming, Z. Cao, H. A. Nguyen, H. G. Jung, J. Kim, Y.-K. Sun, *Adv. Energy Mater.* **2020**, *10*, 20.
[3] L. Oliveira, M. Messagie, S. Rangaraju, J. Sanfelix, M. Hernandez Rivas, J. Van Mierlo, *J. Cleaner Prod.* **2015**, *108*, Part A.
[4] J. G. Kim, B. Son, S. Mukherjee, N. Schuppert, A. Bates, O. Kwon, M. J. Choi, H. Y. Chung, S. Park, *J. Power Sources* **2015**, *282*, 1.
[5] S. Troy, A. Schreiber, T. Reppert, H.-G. Gehrke, M. Finsterbusch, S. Uhlenbruck, P. Stenzel, *Appl. Energy* **2016**, *169*, 1.
[6] Y. E. Durmus, H. Zhang, F. Baakes, G. Desmaizieres, H. Hayun, L. Yang, M. Kolek, V. Küpers, J. Janek, D. Mandler, S. Passerini, Y. Ein-Eli, *Adv. Energy Mater.* **2020**, *10*, 2000089.
[7] F. Han, Y. Zhu, X. He, Y. Mo, C. Wang, *Adv. Energy Mater.* **2016**, *6*, 8.
[8] P. Oh, H. Lee, S. Park, H. Cha, J. Kim, J. Cho, *Adv. Energy Mater.* **2020**, *10*, 24.
[9] International Organization for Standardization, ISO 14040:2006, *Environmental Management-Life cycle assessment- Principles and framework*, **2006**.
[10] E. L. Ager-Wick, M.-B. Guillaume, S. Bhawna, S. A. Kumar, V. L. Ole, S. A. Hammer, *J. Ind. Ecol.* **2014**, *18*, 1.
[11] L. Ahmadi, S. B. Young, M. Fowler, R. A. Fraser, M. A. Achachlouei, *Int. J. Life Cycle Assess.* **2017**, *22*, 111.
[12] J. Peters, D. Buchholz, S. Passerini, M. Weil, *Energy Environ. Sci.* **2016**, *9*, 1744.
[13] A. J. Davidson, S. P. Binks, J. Gediga, *Int. J. Life Cycle Assess.* **2016**, *21*, 1624.
[14] M. Hiremath, K. Derendorf, T. Vogt, *Environ. Sci. Technol.* **2015**, *49*, 8.
[15] L. Smith, T. Ibn-Mohammed, S. C. L. Koh, I. M. Reaney, *Appl. Energy* **2018**, *220*, 496.
[16] T. Ibn-Mohammed, I. M. Reaney, S. C. L. Koh, A. Acquaye, D. C. Sinclair, C. A. Randall, F. H. Abubakar, L. Smith, G. Schileo, L. Ozawa-Meida, *J. Eur. Ceram. Soc.* **2018**, *38*, 4922.
[17] J. B. Dunn, L. Gaines, J. C. Kelly, C. James, K. G. Gallagher, *Energy Environ. Sci.* **2015**, *8*, 916.
[18] J. Schnell, H. Knörzer, A. J. Imbsweiler, G. Reinhart, *Energy Technol.* **2020**, *8*, 1901237.
[19] T. Ibn-Mohammed, S. C. L. Koh, I. M. Reaney, A. Acquaye, D. Wang, S. Taylor, A. Genovese, *Energy Environ. Sci.* **2016**, *9*, 3495.
[20] N. Thonemann, A. Schulte, D. Maga, *Sustainability* **2020**, *12*, 1192.
[21] J. B. Goodenough, K.-S. Park, *J. Am. Chem. Soc.* **2013**, *135*, 1167.
[22] K. Richa, C. W. Babbitt, G. Gaustad, X. Wang, *Resour., Conserv. Recycl.* **2014**, *83*, 63.
[23] G. Majeau-Bettez, T. R. Hawkins, A. H. Strømman, *Environ. Sci. Technol.* **2011**, *45*, 10.
[24] F. Du, N. Zhao, Y. Li, C. Chen, Z. Liu, X. Guo, *J. Power Sources* **2015**, *300*, 24.
[25] Y. Li, Z. Wang, C. Li, Y. Cao, X. Guo, *J. Power Sources* **2014**, *248*, 1.
[26] M. Zackrisson, L. Avellán, J. Orlenius, *J. Cleaner Prod.* **2010**, *18*, 1519.
[27] S. Saxena, C. L. e. Floch, J. MacDonald, S. Moura, *J. Power Sources* **2015**, *282*, 265.
[28] L. Ahmadi, M. Fowler, S. B. Young, R. A. Fraser, B. Gaffney, S. B. Walker, *Sustainable Energy Technologies and Assessments* **2014**, *8*, 9.
[29] A. M. Bernardes, D. C. R. Espinosa, J. A. S. Tenório, *J. Power Sources* **2004**, *130*, 291.
[30] K. Briffaerts, C. Spirinckx, A. Van der Linden, K. Vrancken, *Waste Manage.* **2009**, *29*, 2321.
[31] L. A.-W. Ellingsen, C. R. Hung, A. H. Strømman, *Transportation Research Part D: Transport and Environment* **2017**, *55*, 1.
[32] C. M. Lastoskie, Q. Dai, *J. Cleaner Prod.* **2015**, *91*, 158.
[33] A. Keshavarzmohammadian, S. M. Cook, J. B. Milford, *J. Cleaner Prod.* **2018**, *202*, 770.
[34] X. Yao, N. Huang, F. Han, Q. Zhang, H. Wan, J. P. Mwizerwa, C. Wang, X. Xu, *Adv. Energy Mater.* **2017**, *7*, 17.
[35] T. P. Hendrickson, O. Kavvada, N. Shah, R. Sathre, C. D. Scown, *Environ. Res. Lett.* **2015**, *10*, 014011.
[36] M. Raugei, P. Winfield, *J. Cleaner Prod.* **2019**, *213*, 926.
[37] C. Spanos, D. E. Turney, V. Fthenakis, *Renewable Sustainable Energy Rev.* **2015**, *43*, 478.
[38] D. A. Notter, M. Gauch, R. Widmer, P. Wäger, A. Stamp, R. Zah, H.-J. Althaus, *Environ. Sci. Technol.* **2010**, *44*, 7744.
[39] G. M. Mudd, *Ore Geol. Rev.* **2010**, *38*, 9.
[40] C. Banza Lubaba Nkulu, L. Casas, V. Haufroid, T. De Putter, N. D. Saenen, T. Kayembe-Kitenge, P. M. Obadia, D. K. W. Mukoma, J.-M. L. Ilunga, T. S. Nawrot, O. L. Numbi, E. Smolders, B. Nemery, *Nat. Sustain.* **2018**, *1*, 495.
[41] P. Meshram, B. D. Pandey, Abhilash, *Resour. Policy* **2019**, *60*, 1.
[42] W. Klöpffer, B. Grahl, *Life Cycle Assessment (LCA): A Guide to Best Practice*, Weinheim, Germany, **2014**.
[43] J. Matheys, W. Van Autenboer, J.-M. Timmermans, J. Van Mierlo, P. Van den Bossche, G. Maggetto, *The International Journal of Life Cycle Assessment* **2007**, *12*, 191.
[44] Argonne National Laboratory, BatPac: Battery Manufacturing Cost Estimation, https://www.anl.gov/tcp/batpac-battery-manufacturing-cost-estimation, (accessed: April 2020).
[45] G. J. May, A. Davidson, B. Monahov, *J. Energy Storage* **2018**, *15*, 145.
[46] D.-R. Shi, J. Fu, Z. Shadike, M.-H. Cao, W.-W. Wang, Z.-W. Fu, *ACS Omega* **2018**, *3*, 7.
[47] J.-Y. Choi, D. J. Lee, Y. M. Lee, Y.-G. Lee, K. M. Kim, J.-K. Park, K. Y. Cho, *Adv. Funct. Mater.* **2013**, *23*, 17.
[48] S. C. L. Koh, A. Genovese, A. A. Acquaye, P. Barratt, N. Rana, J. Kuylenstierna, D. Gibbs, *Int. J. Prod. Res.* **2013**, *51*, 2092.
[49] T. Ibn-Mohammed, R. Greenough, S. Taylor, L. Ozawa-Meida, A. Acquaye, *Build. Environ.* **2014**, 72.
[50] A. Acquaye, T. Ibn-Mohammed, A. Genovese, G. A. Afrifa, F. A. Yamoah, E. Oppon, *Eur. J. Oper. Res.* **2017**, 269.

[51] T. Wiedmann, S. Suh, K. Feng, M. Lenzen, A. Acquaye, K. Scott, J. R. Barrett, *Environ. Sci. Technol.* **2001**, *45*, 13.
[52] S. Suh, G. Huppes, *J. Cleaner Prod.* **2005**, 13.
[53] T. Ibn-Mohammed, S. C. L. Koh, I. M. Reaney, A. Acquaye, G. Schileo, K. B. Mustapha, R. Greenough, *Renewable Sustainable Energy Rev.* **2017**, 80.
[54] A. Lake, A. Acquaye, A. Genovese, N. Kumar, S. C. L. Koh, *Int. J. Prod. Res.* **2015**, *53*, 6495.
[55] Ecoinvent, https://www.ecoinvent.org/, (accessed: May 2018).
[56] G. Geisler, T. B. Hofstetter, K. Hungerbühler, *Int. J. Life Cycle Assess.* **2014**, *9*, 1.
[57] A. P. Acero, C. Rodriguez, A. Ciroth, G. Delta, https://www.openlca.org/wp-content/uploads/2016/08/LCIA-METHODS-v.1.5.5.pdf, (accessed: October 2020).
[58] CML2001. think step GaBi, https://www.gabi-software.com/international/support/gabi/gabi-lcia-documentation/cml-2001/, (accessed: October 2020).
[59] P. G. Georgopoulos, S. W. Wang, I. G. Georgopoulos, M. J. Yonone-Lioy, P. J. Lioy, *J. Exposure Sci. Environ. Epidemiol.* **2006**, *16*, 397.
[60] L. Polissar, K. Lowry-Coble, D. A. Kalman, J. P. Hughes, G. van Belle, D. S. Covert, T. M. Burbacher, D. Bolgiano, N. K. Mottet, *Environ. Res.* **1990**, *53*, 29.
[61] R. J. Erickson, D. A. Benoit, V. R. Mattson, E. N. Leonard, H. P. Nelson Jr., *Environ. Toxicol. Chem.* **1996**, *15*, 2.
[62] M. A. J. Huijbregts, L. J. A. Rombouts, S. Hellweg, R. Frischknecht, A. J. Hendriks, D. van de Meent, A. M. J. Ragas, L. Reijnders, J. Stuijs, *Environ. Sci. Technol.* **2006**, *40*, 3.
[63] N. Haque, A. Hughes, S. Lim, C. Vernon, *Resources* **2014**, *3*, 614.
[64] D. Bauer, D. Diamond, J. Li, D. Sandalow, P. Telleen, B. Wanner, U.S. Department of Energy Critical Materials Strategy, https://energy.gov/sites/prod/files/DOE_CMS2011_FINAL_Full.pdf, (accessed: July 2020).
[65] *Communication from the commission to the European Parliament, the Council, The European Economic and Social Committee and the Committee of the Regions, on the 2017 list of Critical Raw Materials for the EU*, **2017**.
[66] T. E. Graedel, R. Barr, C. Chandler, T. Chase, J. Choi, L. Christoffersen, E. Friedlander, C. Henly, C. Jun, N. T. Nassar, D. Schechner, S. Warren, M. Yang, C. Zhu, *Environ. Sci. Technol.* **2012**, *46*, 2.
[67] M. Tanaka, T. Oki, K. Koyama, H. Narita, T. Oishi, in *Handbook on the Physics and Chemistry of Rare Earths*, Vol. *43* (Eds: G. B. Jean-Claude, K. P. Vitalij), Elsevier, New York, **2013**, Ch. 255.
[68] K. Binnemans, P. T. Jones, B. Blanpain, T. Van Gerven, Y. Yang, A. Walton, M. Buchert, *J. Cleaner Prod.* **2013**, *51*, 1.
[69] I. Staffell, *Energy Policy* **2017**, *102*, 463.
[70] C. Cany, C. Mansilla, G. Mathonnière, P. da Costa, *Energy* **2018**, *150*, 1.
[71] T. Ibn-Mohammed, C. A. Randall, K. B. Mustapha, J. Guo, J. Walker, S. Berbano, S. C. L. Koh, D. Wang, D. C. Sinclair, I. M. Reaney, *J. Eur. Ceram. Soc.* **2019**, *39*, 5213.
[72] European Commission, *Commission Staff Working Document Report, on Critical Raw Materials and the Circular Economy*, **2018**.
[73] T. Yang, Y. Lu, L. Li, D. Ge, H. Yang, W. Leng, H. Zhou, X. Han, N. Schmidt, M. Ellis, Z. Li, *Adv. Sustainable Syst.* **2020**, *4*, 1.
[74] X. Li, F. Dogan, Y. Lu, C. Antunes, Y. Shi, A. Burrell, C. Ban, *Adv. Sustainable Syst.* **2020**, *4*, 8.
[75] Z. Zhang, J. Zhang, Y. Sun, H. Jia, L. Peng, Y. Zhang, J. Xie, *J. Energy Chem.* **2020**, *41*, 1.
[76] X. Li, L. Jin, D. Song, H. Zhang, X. Shi, Z. Wang, L. Zhang, L. Zhu, *J. Energy Chem.* **2020**, *40*, 1.
[77] Z. Huang, R.-a. Tong, J. Zhang, L. Chen, C.-A. Wang, *J. Power Sources* **2020**, *451*, 1.
[78] Y. Tian, F. Ding, H. Zhong, C. Liu, Y.-B. He, J. Liu, X. Liu, Q. Xu, *Energy Storage Mater.* **2018**, *14*, 49.
[79] B. Kang, G. Ceder, *Nature* **2009**, *458*, 190.
[80] A. Ahmed, I. Hassan, T. Ibn-Mohammed, H. Mostafa, I. M. Reaney, S. C. L. Koh, J. Zu, Z. L. Wang, *Energy Environ. Sci.* **2017**, *10*, 3.