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Cradle-to-gate life cycle assessment of cylindrical sulfide-based solid-state batteries

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Abstract

Purpose Solid-state batteries (SSBs) are a current research hotspot, as they are safer and have a higher energy density than state-of-the-art lithium-ion batteries (LIBs). To date, their production only occurs on a laboratory scale, which provides a good opportunity to analyze the associated environmental impacts prior to industrialization. This paper investigates the environmental impacts of the production of cylindrical SSB, to identify environmental hotspots and optimization potentials.

Methods Here, an attributional cradle-to-gate life cycle assessment (LCA) is performed, focusing on SSBs that use a NMC811/lithium germanium phosphorous sulfide (LiGPS) composite cathode, a sulfide-based solid separator electrolyte, and a lithium metal anode. The life cycle impact assessment (LCIA) is performed in Umberto 11 using the Environmental Footprint 3.1 method with primary and literature data and the Evoinvent 3.9 database for background data.

Results and discussion The results show climate change impacts of 205.43 kg CO_2 eq./kwh (for the base case), with hotspots primarily attributable to the electrolyte and cathode production, and more specifically to the LiPS and LiGPS synthesis as well as to the cathode active material. Additionally, the scenario analysis shows that an upscaling of the LiPS and LiGPS synthesis reduces environmental impacts across all assessed impact categories. In addition, it was shown that the use of an in situ anode further improves the overall environmental performance, while the use of alternative cathode active materials, such as NMC622 and LFP did not lead to any improvements, at least with reference to the storage capacity.

Conclusion The article highlights the environmental hotspots of sulfide-based SSB production, namely electrolyte and catholyte synthesis. The results show that upscaling the synthesis reduces the environmental impact and that cells with higher energy density show a favorable environmental performance. However, SSBs are still in the development stage and no final recommendation can be made at this time.

Keywords Solid-state batteries (SSB) · Solid electrolyte · Sulfide-based electrolyte · Life cycle assessment (LCA) · Environmental impacts · Battery manufacturing

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1 Introduction

The increasing urgency to mitigate climate change has driven policymakers and industry leaders to set targets to reduce global greenhouse gas (GHG) emissions (United Nations Framework Convention on Climate Change 2015; European commission 2019, 2021). The energy and mobility sectors are accountable for more than 55% of global GHG emissions, making them an important lever (Crippa et al. 2022).

Energy storage systems play a crucial role in both the electrification of the mobility sector and in providing necessary energy storage for renewable energy integration (Gül et al. 2020; Ma et al. 2021). Currently, LIBs are the leading technology for rechargeable electrical energy storage and the LIB market is expected to grow significantly in the next years, to 0.5–1.5 TWh in 2025, 2–4 TWh around 2030, and most likely to more than 10 TWh after 2030 (Schmaltz et al. 2022). Yet, the current generation of LIBs faces challenges related to raw material availability, safety concerns, and a limited energy storage capacity (Janek and Zeier 2016; Wu and Kong 2018; Karabelli et al. 2020; Pasta et al. 2020; Kiemel et al. 2021; Kong et al. 2021).

SSB, which are distinguished in inorganic and polymer based ones, are the next step toward lithium-based batteries with improved safety, higher energy and performance (Janek and Zeier 2016; Yao et al. 2016; Ma et al. 2021). They have the same electrochemical structure and operating principle as LIBs, but utilize a solid electrolyte. These electrolytes do not contain flammable organic materials, as is the case with liquid electrolytes, which is beneficial in terms of safety (Yao et al. 2016). In addition, the use of a solid electrolyte can significantly improve other key areas such as the energy density by enabling the use of metal anodes such as lithium metal (Karabelli et al. 2021). Lithium metal has a high specific capacity of 3,860 mAh/g, which is about ten times higher than that of graphite (372 mAh/g), that is used in LIBs (Liu et al. 2018). So far, lithium metal is not used in combination with liquid organic electrolytes, due to the formation of lithium dendrites, which can puncture the separator and thereby lead to an internal short circuit (Karabelli et al. 2021; Ma et al. 2021). In SSBs the solid electrolyte serves as separator and as a physical barrier to the lithium dendrites and thereby enable the use of lithium metal as anode material. Due to these advantages, SSBs have become a new research hotspot in battery development (Pasta et al. 2020; Ma et al. 2021). Nevertheless, their development is still ongoing and several challenges remain to be solved, such as issues regarding the ionic conductivity, electrochemical stability, lithium compatibility, and the upscaling of production (Schmaltz et al. 2022).

The early development stage of SSBs offers a valuable opportunity to analyze the environmental impacts associated with SSBs before they are produced on an industrial scale. Life cycle assessment (LCA) can be an appropriate method to identify the main hotspots and recommendations to minimize them. In recent years, there has been a growing interest in the environmental impacts of LIBs, with several LCAs having been conducted (Peters et al. 2017; Peters and Weil 2018; Porzio and Scown 2021). For SSBs, however, only a limited number of LCAs are available so far.

Lastoskie and Dai (2015) published first results of a LCA of SSBs, comparing laminated LIBs with vacuum vapor-deposited thin-film SSBs. The results show that the cumulative energy demand (CED) and GWP per unit of energy storage are 25–65% lower for SSBs across all assessed chemistries (Lastoskie and Dai 2015). Troy et al. (2016) analyzed the manufacturing process of SSBs with a lithium cobalt oxide (LCO)/lithium lanthanum zirconate (LLZ) cathode and a LLZ solid-state electrolyte, based on actual laboratory-scale production, finding on-site electricity and high-temperature process steps significantly impacting results. Moreover, the results shows considerable potential for improvement by scaling up the production process, whereby in the GWP category improvements of up to 95% can be achieved compared to the laboratory process. Vandepaer et al. (2017) analyzed stationary lithium metal polymer (LMP) batteries, showing that most of the environmental impacts are attributable to the manufacturing phase and that batteries with a state-of-the-art cell chemistry have significantly greater impacts on global warming and ozone depletion. Conversely, LMP batteries show a greater impact on the eutrophication of water bodies. Keshavarzmohammadian et al. (2018) assessed a sulfide-based solid-state lithium pyrite battery that is suitable for electric vehicles (EV). The estimated GWP is 199 kg CO_2-eq./kWh, with the cleanroom energy accounting for the largest proportion of the total GWP at 73%, followed by cathode paste at 6%. Rossi et al. (2020) performed an LCA of state-of-the-art and innovative batteries for solar home systems, which also included SSB. The analysis shows that the most promising energy storage technologies from a sustainability point of view are nickel–cobalt-manganese (NMC) SSB and nickel–cobalt-aluminum (NCA) LIBs. Smith et al. (2021) performed a comparative LCA of a LIB and a SSB using a lithium lanthanum tantalum zirconia oxide (LLZTO) electrolyte, finding higher environmental impacts for LIBs in most environmental impact categories, due to the lower assumed lifetime of SSBs of 100 cycles. Zhang et al. (2022), assessed a SSB with lithium aluminum titanium phosphate (LATP) electrolyte and compared the results to those of LIBs. Again, SSBs were found to have a higher environmental impact, with the hotspot being the solid electrolyte and its energy-intensive production. The most recent LCA of SSB was carried out by Popien et al. (2023) who investigated the sustainability of current and future traction batteries. The results indicate lower environmental impacts for SSB and lithium sulfur batteries (LSB) as well as SSB-LSB batteries compared to LIB, with the latter having the smallest impact (79.63 kg CO_2 eq/kWh). For the all-solid-state batteries (ASSB) results range from 111 kg CO_2 eq/kWh for the NMC811 and lithium iron phosphate (LFP) chemistry to 123 kg CO_2 eq/kWh for the batteries with NMC622 cathode chemistry (Popien et al. 2023). The decisive factor for the impact category climate change is the electricity demand, particularly in cell assembly, pack assembly and the production of the cathode active material. A scenario analysis has also shown that a switch to 100% renewable energy could result in savings of 26–48% in the climate change impact category.

A detailed overview of the aforementioned LCAs can be found in Mandade et al. (2023), who analyzed the studies regarding inventory data, scope, and LCA results.

Interestingly, among the LCAs on SSBs, only two studies analyze the environmental impacts of sulfide-based SSBs (Keshavarzmohammadian et al. 2018; Popien et al. 2023). Sulfide-based solid electrolytes, however, are considered very promising due to their high ionic conductivity at room temperature (Karabelli et al. 2021). Furthermore, Keshavarzmohammadian et al. (2018) investigate a pyrite battery whose cell chemistry differs from state-of-the-art cathode chemistries. Consequently, there is still a lot of room to investigate sulfide-based SSBs and its supply chain.

Hence, the aim of this paper is to analyze the environmental impacts of sulfide-based SSBs in the cylindrical cell format, across various cell chemistries to identify hotspots and optimization opportunities. This study adds to existing knowledge, as there are limited LCAs available for SSBs, with only two focusing on sulfide-based electrolytes, and only one focusing on cylindrical cells. The latter option, however, is focused on oxide-based cells rather than sulfide-based cells.

2 Methods

2.1 LCA method

LCA is a structured, comprehensive, and internationally standardized method for investigating and quantifying the environmental impacts of a product or service considering the whole life cycle (ISO 14040 (2006); ISO 14044 (2006)). The LCA framework includes four phases: (i) definition of goal and scope, (ii) life cycle inventory, (iii) life cycle impact assessment (LCIA), and (iv) interpretation of results (ISO 14040 (2006); ISO 14044 (2006)). In this paper, an attributional LCA of the manufacturing of a cylindrical sulfide-based SSB is conducted, focusing on a cradle-to-gate analysis. The LCIA was carried out using Umberto 11, employing the Ecoinvent 3.9 database for background data, and the Environmental Footprint 3.1 (EF 3.1) impact assessment method, as recommended in the rules for calculating the Carbon Footprint of Electric Vehicle Batteries (CFB-EV) (Andreasi Bassi et al. 2023). Foreground data were obtained from literature, own calculations, and personal communications. The scenario analysis examines the process that has the greatest impact on the results and explores variations in cell chemistries.

2.2 Goal and scope definition

The objective of this study is to analyze the environmental impacts of the manufacturing of cylindrical (21,700—21 mm diameter and 70 mm length) sulfide-based SSB with a NMC811/LiGPS composite cathode, a lithium metal anode and a lithium phosphorous sulfide (LiPS) electrolyte in order to identify environmental hotspots and optimization measures. The choice of cell chemistry is due to the fact that it is particularly promising for future applications given the high ionic conductivity of sulfide-based solid electrolytes and the high energy density of NMC811 and lithium.

In addition to this, four additional scenarios are investigated. For scenario 1, a solid separator electrolyte and catholyte production with a batch size of 10 kg in comparison to the state of the art of < 50 g was assumed in order to represent the production on a larger scale. The data and assumptions were determined in consultation with the electrolyte manufacturer based on larger production machinery and equipment with a higher efficiency. As a result, the energy required for the production of the separator electrolyte is reduced from 0.32 to 0.003 kWh/g and that of the catholyte from 0.11 to 0.0225 kWh/g (personal communication with an electrolyte manufacturer, 2022 (IPA 2023)). The assumptions regarding the material yield remain unchanged in this assessment. For all other scenarios, the solid electrolyte production in larger batch sizes was assumed as well, as the scale up is inevitable considering a market penetration of sulfide-based SSB. In scenario 2, the cathode active material NMC811 was substituted by NMC622 due to its lower nickel content. In scenario 3, the cathode material LFP was chosen as an alternative in order to completely eliminate nickel and cobalt from the cathode. Finally, scenario 4 considered an alternative anode, namely an in situ anode. In the in situ anode approach, lithium-ions stored in the cathode are deposited onto the negative current collector during the first charging cycles and it is thus possible to avoid the complex handling of metallic lithium during production and significantly increase the energy density of the cell (Karabelli et al. 2021; Schmaltz et al. 2022). For the modeling of this scenario, a steel current collector was assumed.

The material choices for the base case and the scenarios 1–4 are depicted in Table 1.

The function of the analyzed cell is to store energy; thus, the functional unit is set to 1 kWh of storage capacity after production, which allows to take into account the energy density. At this point it should be mentioned that performance-related factors such as the charge and discharge efficiency or lifetime are not captured in this study, as due to the early stage of SSB development, no well-founded assumptions can be made regarding performance and lifetime. The reference flow is set by the number of cells required for 1 kWh. The storage capacity for each assessed cell is specified in Sect. 2.3.2. The system boundaries comprise all upstream processes including the production of the cell (cradle-to-gate), while the use phase and the end-of-life phase are excluded.

This is because the application of SSB is not yet defined so the use phase is not considered, while the end-of-life (EoL) phase is excluded, because recycling processes for

Table 1 Overview of the analyzed scenarios with changes to the base case highlighted in grey

	Base case	Scenario 1	Scenario 2	Scenario 3	Scenario 4
Cathode	NMC811 (on Al)	NMC811 (on Al)	NMC622 (on Al)	LFP (on Al)	NMC811 (on Al)
Catholyte	LiGPS (production in small batches)	LiGPS (production in large batches)	LiGPS (production in large batches)	LiGPS (production in large batches)	LiGPS (production in large batches)
Electrolyte	LiPS (small batches)	LiPS (large batches)	LiPS (large batches)	LiPS (large batches)	LiPS (large batches)
Anode	Lithium metal (on Cu)	Lithium metal (on Cu)	Lithium metal (on Cu)	Lithium metal (on Cu)	In-situ (steel foil)

SSB are still at an early stage of research (Doose et al. 2021). Taking such an approach may, however, underestimate the actual environmental impacts of a product and make comparability with studies using other system boundaries more difficult, potentially leading to incomplete assessments of environmental performance. Therefore, the system boundaries of the study should be taken into account in the interpretation.

The study is based on own calculations and secondary data from literature as foreground data, and the Ecoinvent 3.9 database as background data (Ecoinvent 2022). The foreground data was obtained from a research project at the Fraunhofer Institute for Manufacturing Engineering and Automation focusing on the development of cylindrical sulfide-based SSBs (Fraunhofer IPA 2023). To assess the SSB production at an industrial scale, several assumptions have been made, which are described along the article and in the supplementary information (SM1, SM2).

2.3 Life cycle inventory analysis

The following section describes the system under consideration, i.e., the manufacturing process of the SSB under investigation, as well as the assumptions and calculations used to compile the life cycle inventory (LCI).

2.3.1 System description

The SSB battery manufacturing process involves three main steps: 1. Electrode and electrolyte layer production, 2. Cell assembly and 3. Cell finishing (Heimes et al. 2018). This is illustrated in Fig. 1. In general, various different production methods can be used, which mainly depend on the material choices. The process that is described in the following refers to the production of sulfide-based SSB and to the material choices made in this LCA.

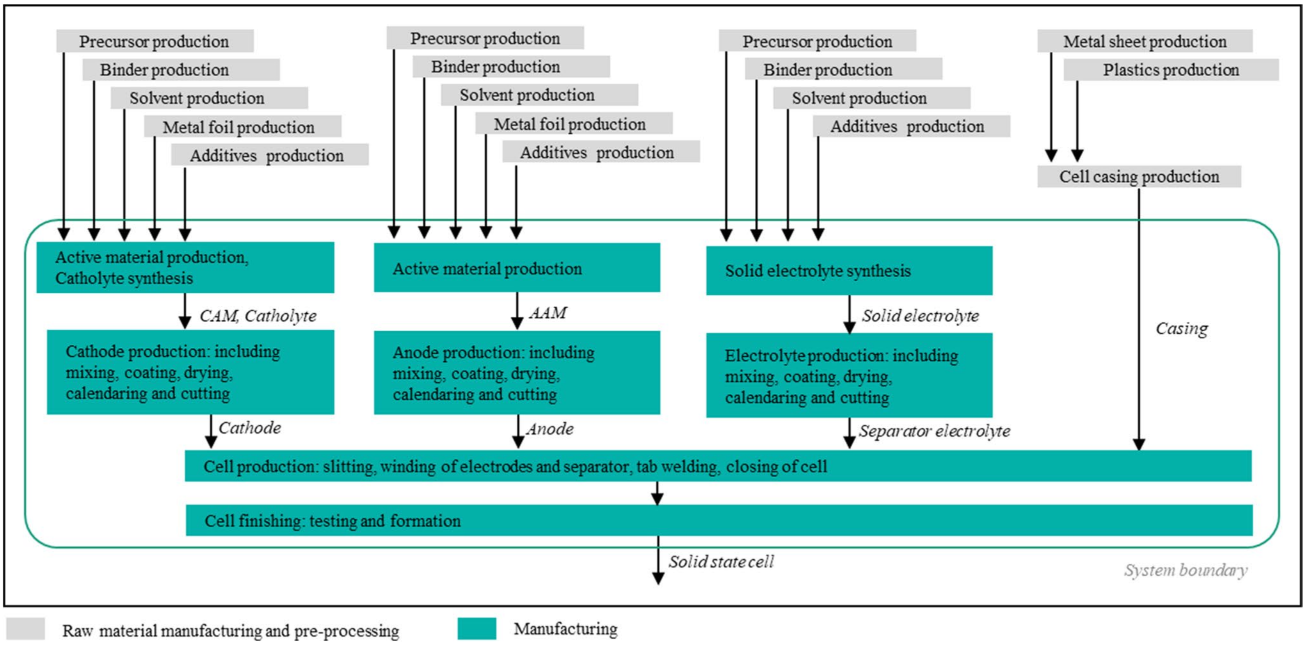

Fig. 1 System boundaries of the LCA of a solid-state battery. Each square represents a process

Electrode and electrolyte layer production The production of electrodes and electrolytes includes the composite cathode production, the anode production, and the electrolyte layer production.

For the composite cathode production, wet chemical processing is assumed (Schnell et al. 2018; Heimes et al. 2018; Schmaltz et al. 2022). In this process route, the active material (NMC811), the solid electrolyte (LiGPS), the conductivity additive (carbon black), the binder (styrene-butadiene rubber (SBR) and the solvent (xylene) are mixed and subsequently coated onto an aluminum current collector. For the production of the catholyte LiGPS, the precursors Li_2S, GeS_2, and P_2S_5 are ground in a planetary mill for 3 h, then tempered at 700 °C for 7 h and milled again in a planetary mill (personal communication with an electrolyte manufacturer, 2022 (Fraunhofer 2023)). After the coating, the solvent is evaporated and the cathode is compacted to ensure good contact between the particles and with the current collector.

For the anode, lithium foil production through extrusion of lithium metal and subsequent high-pressure calendaring is assumed (Schnell et al. 2018; Heimes et al. 2018; Duffner et al. 2021; Schmaltz et al. 2022). The lithium metal foil is then laminated onto the copper current collector.

For the solid separator electrolyte layer, wet chemical processing is assumed (Schnell et al. 2018; Schmaltz et al. 2022). After mixing, the separator electrolyte slurry is coated directly onto the cathode via tape casting and subsequently compacted through calendaring (Schnell et al. 2018; Schmaltz et al. 2022). For the LiPS synthesis, the precursors Li_2S and P_2S_5 are mixed with the solvent tetrahydrofuran (THF) for 16 h, then dried for three days (5 h at 80 °C and then at 140 °C), and finally milled in a planetary mill. Currently, the electrolyte synthesis process is still under development and only small batch sizes (< 50 g) are produced (personal communication with an electrolyte manufacturer, 2022 (Fraunhofer 2023)).

Cell assembly As outlined in literature, the individual layers are subsequently cut to the desired geometry and wound into a coil. This coil is then inserted into the cell casing and contacted to the current collector tabs. Thereafter, the cell is sealed and washed (Heimes et al. 2018; Duffner et al. 2021; Schmaltz et al. 2022).

Cell finishing After the cell assembly, the formation and aging of the cells takes place. When lithium metal is used as the anode material, the SSB is already in a charged state after assembly, resulting in a reduced formation time (Heimes et al. 2018). For the aging, the cells are stored in aging shelves and their performance is tracked. Compared to LIBs, a shorter aging time is expected as stable properties are achieved more quickly due to the solid electrolyte (Heimes et al. 2018).

Miscellaneous In addition to cell production, the dry room energy requirement is taken into account as well. This is due to the requirement that sulfides place on the production environment. The cathode and electrolyte production of sulfide-based SSB must take place in a dry environment (< 100 ppm H_2O) or even in an inert gas atmosphere, because sulfide-based solid electrolytes are susceptible to ambient moisture, resulting in the formation of harmful H_2S (Schnell et al. 2018; Duffner et al. 2021; Hatzell and Zheng 2021; Schmaltz et al. 2022).

2.3.2 Life cycle inventory data

As SSBs are still under development, several assumptions have had to be made to derive the mass inventory and the manufacturing energy demand. Existing tools that facilitate battery design according to user-specified application requirements, such as BatPAC (Nelson et al. 2019) or SolidPAC (Dixit et al. 2022), were only employed as source for default values (material density, porosity, nominal capacity, and open circuit voltage), as they either do not include solid electrolytes or do not support a cylindrical cell design.

Mass inventory For the LCI of the SSB, first the materials and the thicknesses of the individual cell layers were determined. The material selection is based on Ates et al. (2019). Layer thicknesses were defined based on target values from the literature and in consultation with the project team (Duffner et al. 2021; Schmaltz et al. 2022). The cathode thickness was set to 65 µm, the anode thickness to 20 µm, and the separator electrolyte thickness to 25 µm. In addition, a thickness of 10 µm was assumed for the negative current collector and 15 µm for the positive current collector. It should be noted that the anode is typically sized based on the capacity ratio between the anode and cathode; this approach was not used for the sizing because the thickness of the lithium layer would otherwise be less than the thickness of commercially available lithium foils. Subsequently, the volume of each cell layer was calculated based on the jellyroll dimensions that fit into a cylindrical 21,700 cell casing, taking the dimensions reported by Quinn et al. (2018) as a reference. The number of windings, and thus the critical scale for the sheet length, was calculated based on the approach described by Waldmann et al. (2020) which is based on the Archimedean spiral. For the coating, double-sided coating was assumed, resulting in a total cell layer thickness of 255 µm (Fig. 2).

This total layer thickness, an assumed void of 1 mm and a coil diameter of 20.6 mm result in a total sheet length of 1.30 m. Moreover, a width of 64.6 mm was assumed, which was determined based on the height of a 21,700 cylindrical cell. For simplicity, the same width was assumed for all cell layers, although this is normally not the case in order to

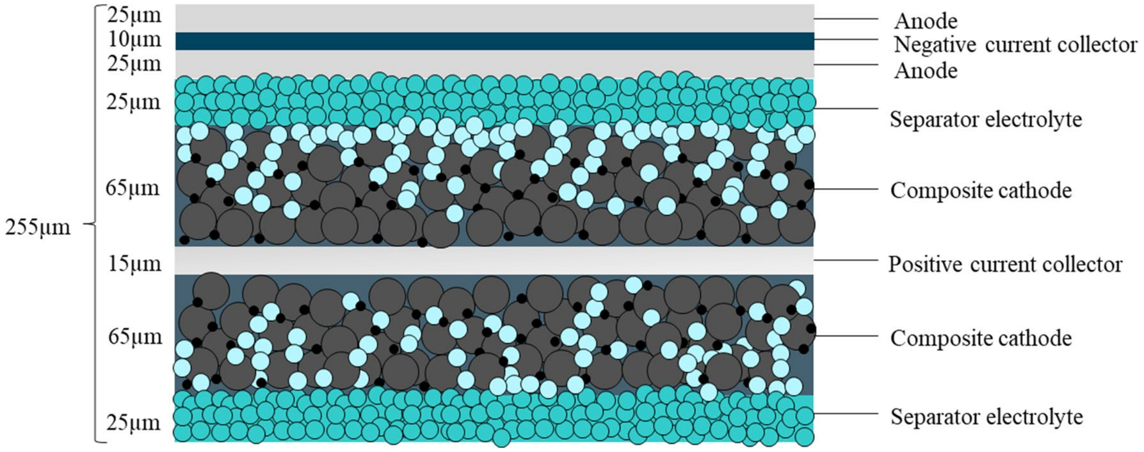

Fig. 2 Schematic electrode stack design for the analyzed SSB, assuming double sided coating of anode and cathode, own illustration

avoid internal contact between the anode and cathode and thus a short circuit. Next, the mass of the respective materials of a SSB cell was calculated based on the product of the volume of the individual cell layer, the weight share of each material, the material density, and the porosity as specified in the SolidPAC tool (15%) (Ates et al. 2019; Dixit et al. 2022).

Equation 1 Material mass

$$\textit{material mass}\ (g)_x = \textit{density}\left(\frac{g}{cm^3}\right)_x \cdot \textit{layer volume}\left(cm^3\right)_y \cdot \textit{weight share}\ (\%)_x \cdot \left(1 - \textit{porosity}(\%)_y\right) \tag{1}$$

Here *x* stands for the corresponding material and *y* for the corresponding cell layer.

To calculate the solvent mass, a liquid content of 60% was assumed for the cathode slurry and 70% for the electrolyte slurry (Dixit et al. 2022). The LCI data of the cell casing, namely the steel container and the insulation ring, were taken from Chordia et al. (2021), who focused on cylindrical 21,700 LIB cells. The mass of the current collector tabs was calculated based on the material density as specified in the SolidPAC tool (Dixit et al. 2022) and the volume of the tabs, assuming a width of 5 mm, a length of 75 mm and a thickness of 0.1 mm. Furthermore, scrap rates as described by Hanisch et al. (2015) were taken into account for the calculation of the LCI. Scrap in this context is production waste such as slurry losses, cuttings, and defective cells.

Based on the material inventory, the gravimetric energy density of the cell was calculated, which is decisive regarding the functional unit of 1 kWh of storage capacity. The following equations were used for this purpose:

Equation 2 Gravimetric energy density

$$\textit{Gravimetric energy density}\left(\frac{Wh}{kg}\right) = \frac{\textit{Cell energy}\ (Wh)}{\textit{Total cell weight}\ (kg)} \tag{2}$$

Equation 3 Volumetric energy density

$$\textit{Volumetric energy density}\ \left(\frac{Wh}{l}\right) = \frac{\textit{Cell energy}\ (Wh)}{\textit{Total cell volume}\ (l)} \tag{3}$$

Equation 4 Cell energy

$$\textit{Cell energy}\ (Wh) = \textit{Cell capacity}\ (Ah) \cdot \textit{Open circuit voltage at}\ 50\%\ \mathrm{SOC}\ (V) \tag{4}$$

Equation 5 Cell capacity

$$\textit{Cell capacity}\ (mAh) = \textit{Active material weight}\ (g) \cdot \textit{Nominal capacity cathode}\ \left(\frac{mAh}{g}\right) \tag{5}$$

The values of the nominal capacity of the cathode active material and the open circuit voltage at 50% state of charge (SOC) were obtained from the SolidPAC tool (Dixit et al. 2022).

The calculated cell energy for all assessed cell chemistries is shown in Table 2.

Manufacturing energy demand In order to calculate the manufacturing energy demand, large-scale cell production was assumed, since laboratory-scale production results in

Table 2 Calculated cell parameters for the assessed cell chemistries, NMC811, NMC622, LFP und NMC811 (in situ)

Cell chemistry	NMC811	NMC622	LFP	NMC811 (in situ)
Active material capacity [mAh/g]	200.00	180.00	150.00	200.00
Cell capacity [Ah]	5.98	5.38	3.33	7.44
Open circuit voltage at 50% SOC [V]	3.75	3.75	3.28	3.75
Cell energy [Wh]	22.42	20.18	10.91	27.89
Cell weight [g]	70.82	70.82	64.98	81.16
Cell volume [l]	0.01	0.01	0.01	0.01
Gravimetric energy density [Wh/kg]	316.62	284.96	167.94	343.66
Volumetric energy density [Wh/l]	2903.58	2613.22	1413.20	3611.66
Number of cells required to provide 1 kWh	44.60	49.55	91.63	35.85

significantly higher environmental impacts (Jinasena et al. 2021). For this purpose, the specific energy consumption provided by Degen et al. (2023) was multiplied by the previously calculated cell energy. The detailed calculation can be found in the supplementary information (SM1).

LCI data Based on the calculation of the material mass and the manufacturing energy demand the following LCI was derived for the base case (Table 3).

3 Results and discussion

3.1 Life cycle impact assessment results

The results of the LCIA for the EF 3.1 midpoint indicator for the functional unit of 1 kWh of storage capacity are presented in Table 4.

Furthermore, the contribution of the individual process steps relative to the overall result per impact category is shown in Fig. 3 in order to highlight the environmental hotspots of SSB manufacturing.

The contribution analysis shows that a large part of the environmental impact is attributable to the precursor materials for the production of cathodes, anodes, and electrolytes.

The catholyte material LiGPS contributes significantly to the climate change impact category, with a share of 17.1%. It also contributes 18.4% to the impact category of ozone layer depletion and 17% to fossils. These high values are mainly due to the high energy demand during the synthesis of the catholyte material, especially due to the laboratory scale production and the assumption of a German electricity mix. In addition, the heat required for the synthesis of germanium sulfide further adds to this high contribution.

Likewise, the cathode active material NMC811 makes a significant contribution to the overall environmental impacts, accounting for more than 15% in all categories except freshwater eutrophication. The contribution to the water scarcity, minerals and metals, and ozone layer depletion indicators is particularly high at 57.7%, 41.9%, and 37% respectively. This is mainly due to NMC811 hydroxide production, which is responsible for 97.1% of water scarcity, 95.5% of minerals and metals, and 80.0% of ozone layer depletion.

The solid electrolyte synthesis, namely that of LiPS, has a significant impact on the overall results as well, particularly in the categories of freshwater eutrophication (55.9%), ionizing radiation (49.8%), and climate change (43.2%). This is due to the high electricity demand for synthesis and the assumed German electricity mix. The electricity consumption contributes to the eutrophication of fresh and marine waters, as fossil fuels release nitrogen and phosphorus compounds. The operation of nuclear power plants also leads to the release of radioactive substances and thus contributes to ionizing radiation. The share of lignite in the employed electricity mix contributes significantly to the category climate change. In comparison, the production of precursors such as Li_2S and P_2S_5 contributes little to the environmental impact.

Anode production contributes mostly to the categories of non-carcinogenic effects (51.6%), minerals and metals (41.9%), and carcinogenic effects (23.1%), which is due to the copper current collector foil onto which the lithium metal is laminated.

The cell casing impacts particularly the acidification of freshwater and terrestrial ecosystems with a share of 52.5%, which is due to the nickel coating of the casing.

The environmental impacts of assembly and cell finishing are negligible, with a share of less than 1% in all impact categories. The same applies to miscellaneous processes, which do not exceed a share of 2.6%. This is interesting, as this process takes into account the energy requirements for the dry and clean rooms, which are higher for SSB than for other battery types such as LIB due to higher requirements for the production environment.

3.2 Scenario analysis

In this section, a scenario analysis is performed to examine the environmental impact variations regarding the electrolyte production, the cathode active material, and the anode, as those processes represent the environmental hotspots and

Table 3 LCI for the base case, showing the process steps cell finishing, cell assembly and anode, cathode and electrolyte sheet production

	Inputs		Outputs	
Formation, aging, quality check, miscellaneous				
SSB	1.05	units		
Electric energy	1.05E-01	kWh		
Gas energy	1.22E+00	MJ		
SSB			1	units
Hazardous waste			3.54	g
Assembly				
Anode sheet	10.09	g		
Cathode sheet	40.64	g		
Electrolyte	8.87	g		
Negative current collector tabs	3.34E-03	g		
Positive current collector tabs	1.01E-03	g		
Case				
Container	8.87	g		
Lid	1.90	g		
Fastening tape	0.66	g		
Insulation ring	1.22	g		
Electric energy	2.80E-02	kWh		
SSB			1	unit
Hazardous waste			1.44	g
Anode sheet production				
Lithium foil	2.48	g		
Negative current collector	8.38	g		
Electric energy	7.93E-03	kWh		
Anode sheet (anode + negative current collector)			10.09	g
Hazardous waste			0.10	g
Copper scrap			0.67	g
Cathode sheet production				
NMC811	33.43	g		
LiGPS	6.16	g		
Carbon black	0.39	g		
NBR (nitrile butadiene rubber)	0.30	g		
Xylene	60.43	g		
Positive current collector	3.81	g		
Electric energy	7.47E-03	kWh		
Gas energy	1.41E-01	MJ		
Cathode sheet			40.64	g
Hazardous waste			3.15	g
Aluminum scrap			0.30	g
Solvent scrap			60.43	g
Electrolyte sheet production				
LIPS	9.30	g		
NBR (nitrile butadiene rubber)	0.14	g		
Xylene	21.36	g		
Electric energy	3.84E-03	kWh		
Gas energy	1.36E-01	MJ		
Electrolyte sheet			8.87	g
Hazardous waste			1.83	g
Solvent scrap			20.09	g

Table 4 Overview of cradle-to-gate environmental impacts of solid-state battery cell production for the EF 3.1 midpoint indicator

Indicator	Value	Unit
Climate change	205.43	kg CO_2-eq
Carcinogenic effects	1.52E-07	CTUh
Non-carcinogenic effects	5.92E-06	CTUh
Respiratory effects, inorganics	9.02E-06	Disease incidences
Ozone layer depletion	5.73E-06	kg CFC-11-eq
Photochemical ozone creation	6.62E-01	kg NMVOC- eq
Ionizing radiation	35.26	kBq U_{235}- eq
Freshwater ecotoxicity	1682.34	CTUe
Freshwater eutrophication	0.18	kg P- eq
Marine eutrophication	0.23	kg N- eq
Terrestrial eutrophication	1.88	mol N- eq
Freshwater and terrestrial acidification	2.35	mol H+—eq
Fossils	3121.25	MJ
Minerals and metals	8.38E-03	kg Sb- eq
Land use	618.05	points
Water scarcity	278.35	m^3 world- eq

because the electrolyte production in the base case is only performed at a very small scale (batch size < 50 g).

An overview of the climate change impacts exemplarily for each scenario compared to the base case is shown in Fig. 4. The LCIA results for all other impact categories are provided in the supplementary information (SM3). Climate change impacts were reduced by 38% in scenario 1, by 28% in scenario 2, by 8% in scenario 3, and by 46% in scenario 4. This reduction is reasonable considering that the electricity required for the solid electrolyte synthesis is relatively high due to the small batch size. Thus, a significant reduction in energy demand can be achieved by upscaling the production process for solid electrolyte powder, which leads to a reduction in environmental impacts.

Furthermore, scenarios 1 to 4 show that the greatest reduction can be achieved by using an in situ anode in combination with large-scale solid electrolyte production scenarios 2 and 3 show no improvement over scenario 1, although improvements over the base case could be achieved by improved electrolyte production. This is particularly due to the lower energy density of the NMC622 and LFP materials compared to NMC811 and the functional unit of 1 kWh per storage capacity. In the case of LFP, the advantages lie

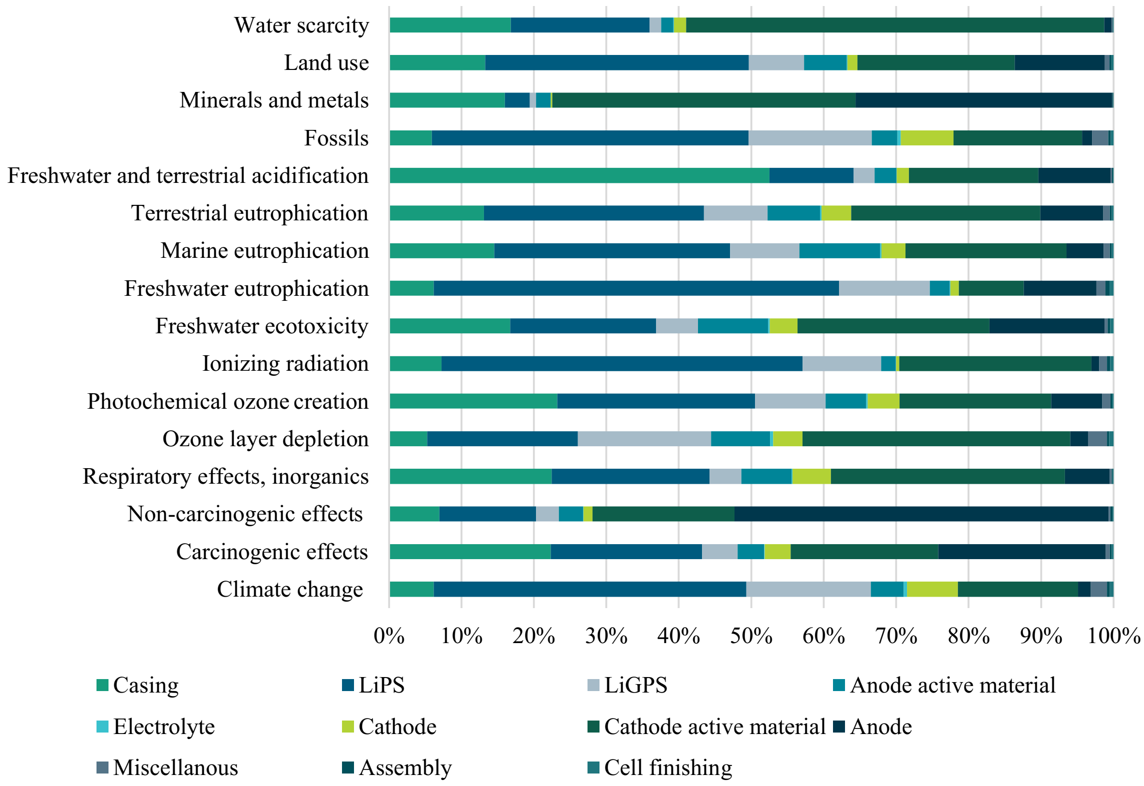

Fig. 3 Contribution analysis of selected impact indicators for the solid-state battery cell

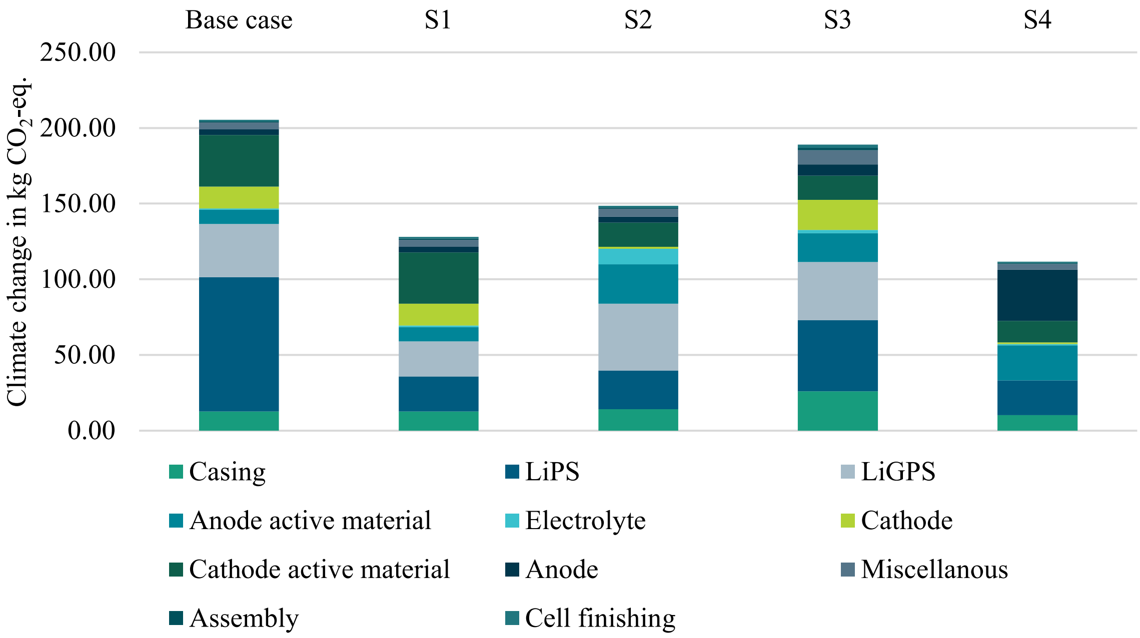

Fig. 4 Climate change impacts for the SSB for the base case, scenario 1 (up-scaled electrolyte and catholyte production), scenario 2 (NMC622 as cathode), scenario 3 (LFP as cathode) and scenario 4 (in situ anode) in kg CO$_2$-eq./kWh of storage capacity

primarily in a longer lifetime and are therefore not considered by the functional unit. LFP based SSB would be beneficial for a functional unit based on mass or the lifetime specific storage capacity. Consequently, the improvements in scenarios 2 and 3 compared to the base case are due to the upscaling of the electrolyte synthesis, but not to the change in cathode material.

3.3 Validation with other LCA studies

In order to verify the plausibility of the results, they are compared with literature. For this purpose, LCAs of SSBs were selected that have the same system boundaries as those selected in this study and for which the environmental impacts per kWh were specified or a conversion to this unit was possible. A detailed overview of the studies in comparison can be found in the Supplementary information (SM4).

A comparison with Troy et al. (2016) shows that the base case results of this study are significantly lower for the industrial scale in all impact categories examined, except for the marine and freshwater eutrophication categories (by a factor of about 150 in the climate change category). The reason for this is that Troy et al. (2016) conducted their assessment at a laboratory scale. In their scenario for industrial production, the values are smaller, but still about a factor of five higher than the results in this study. This shows that the scale considered is critical and that improved environmental performance can be achieved through economies of scale. The effects in the eutrophication categories in this study are due to the LiPS synthesis and the cathode active material and here in particular the nickel sulfate used, which also explains the difference as other materials are found in Troy et al. (2016) with LCO and LLZO. Furthermore, the environmental hotspot in the climate change impact category in Troy et al. (2016) is on the electrolyte at 83%. This is due to the heat required for calcination and sintering. Since a sulfide-based electrolyte is used here, the hotspots are distributed over the cathode and electrolyte as no sintering step is required.

The results of Keshavarzmohammadian et al. (2018) are particularly relevant because they also assess sulfide-based SSB. Their environmental impacts are of similar magnitude in all categories. However, the hotspots are different. In their LCA, the energy demand of the drying room is by far the largest contributor with 72% of the climate change impacts, which is not consistent with the results of this LCA, where the hotspots are in electrolyte production. Keshavarzmohammadian et al. (2018) only consider the production of the electrolyte powder by mixing the precursors Li_2S and P_2S_5, while this study also considers the heating steps, which cause a higher energy demand. In addition, the energy demand for the dry room calculated by Keshavarzmohammadian et al. (2018) is much higher (195 Wh/Wh) than the value considered in this LCA (18.25 Wh/Wh), which explains the discrepancy in the results. However, the large discrepancies highlight the need for primary data collection and more in-depth analyses.

A comparison with Zhang et al. (2022) shows that their results are significantly higher in all categories. This is due to the production scale, which is at the laboratory level. Furthermore, the hotspots are mainly in the electrolyte production, which is due to the sintering steps required for oxide-based electrolytes.

The results of Popien et al. (2023) are lower for all cell chemistries except the in situ cell in scenario 4, although they assume a lower specific energy for NMC811 and NMC622 and a lower manufacturing energy requirement. The reason for the lower impact could be the consideration at the pack level, as pack components such as module housing, pack housing, or battery management system are included in the total mass and the environmental impacts.

3.4 Uncertainties

The main uncertainties arise from the use of secondary data and the associated assumptions and calculations of the SSB mass composition and energy requirements. The former is based on targeted future layer thicknesses and electrode compositions obtained from literature. Yet, due to obstacles related to ionic conductivity, interfacial contact, cyclability, or processability, these will most likely change in the future and the current assumptions are rather optimistic, especially with respect to the calculated energy density. In addition, the binder content may have to be increased due to the cylindrical design, which negatively affects the energy density. The comparison with literature has shown that the assumptions, particularly regarding the electrolyte and the cathode, can significantly affect the overall results, with, for instance, the environmental impacts of pure cathodes being much lower than those of composite cathodes. In addition, the environmental impacts of miscellaneous processes, such as the dry room operation, yielded significantly lower values than in the other LCA that evaluated sulfide-based SSBs. Moreover, the electricity mix for Germany reported in the Ecoinvent datasets was adopted. Since SSB production is not expected to start until after 2025, an assessment based on the future electricity mix would be advisable. Furthermore, results vary greatly with production scale. In this work, large-scale production is assumed, whereas the data for solid electrolyte synthesis refers to small batch sizes (in the base case), which results in inconsistencies regarding the scale of the overall assessment. With respect to the solid electrolyte, it should also be mentioned that no data sets for the precursors Li_2S, P_2S_5, and GeS_2 as well as for germanium are not available in the Ecoinvent database, meaning that those data had to be modeled based on literature values.

4 Conclusion and outlook

The aim of this article was to analyze the environmental impacts of the production of sulfide-based SSBs by means of LCA. More specifically, the focus is on the production of cylindrical cells that use a NMC811/LiGPS composite cathode, LiPS as the separator electrolyte, and a lithium metal anode. Furthermore, an upscaled LiPS and LiGPS synthesis were investigated, as well as using NMC611 and LFP as cathode materials and an in situ anode. The results show that the production of SSBs generates climate change impacts of 205.43 kg CO_2-eq. per kWh of storage capacity. The environmental hotspots are mainly observed in electrolyte and composite cathode production and are attributable to the energy requirements of the LiPS and LiGPS synthesis, as well as to the cathode active material production. The high contribution of the solid electrolyte can be ascribed to the production scale, and by upscaling the synthesis process, a reduction in environmental impacts is achieved, with a 38% reduction in the category climate change. In addition, it is shown that the use of an in situ anode can improve the overall environmental performance, while the alternative cathode active material NMC622 and LFP did not result in any improvements on a storage capacity basis.

In conclusion, in the case of SSBs, primary data for most manufacturing processes is not available yet. Thus, future research should focus on validating the results obtained in this LCA with industry data once it is available. Furthermore, it is necessary to analyze the use phase and the end-of-life phase in order to obtain a holistic picture of environmental impacts and to avoid burden shifting. It would also be interesting to analyze the SSB in a bipolar set up, as this further improves the energy density of the cell. Finally, the environmental impact of other sulfide-based solid electrolytes should be investigated, especially as electrolytes such as LiGPS are expensive and therefore not viable for mass production.

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Data availability All data generated or analyzed during this study are included in this published article and its supplementary information files.

Declarations

Competing interests The authors declare no competing interests.

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