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Wickerts, S., Arvidsson, R., Nordelöf, A. et al (2023). Prospective Life Cycle Assessment of Lithium-Sulfur Batteries for Stationary Energy Storage. ACS Sustainable Chemistry & Engineering, 11(26): 9553-9563. <http://dx.doi.org/10.1021/acssuschemeng.3c00141>

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Prospective Life Cycle Assessment of Lithium-Sulfur Batteries for Stationary Energy Storage

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Cite This: *ACS Sustainable Chem. Eng.* 2023, 11, 9553–9563



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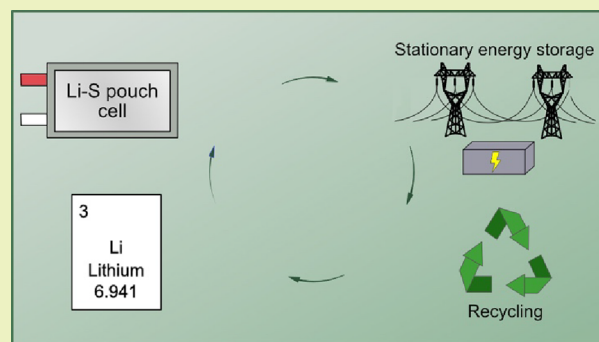
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ABSTRACT: The lithium-sulfur (Li-S) battery represents a promising next-generation battery technology because it can reach high energy densities without containing any rare metals besides lithium. These aspects could give Li-S batteries a vantage point from an environmental and resource perspective as compared to lithium-ion batteries (LIBs). Whereas LIBs are currently produced at a large scale, Li-S batteries are not. Therefore, prospective life cycle assessment (LCA) was used to assess the environmental and resource scarcity impacts of Li-S batteries produced at a large scale for both a cradle-to-gate and a cradle-to-grave scope. Six scenarios were constructed to account for potential developments, with the overall aim of identifying parameters that reduce (future) environmental and resource impacts. The specific energy density and the type of electrolyte salt are the two most important parameters for reducing cradle-to-gate impacts, whereas for the cradle-to-grave scope, the electricity source, the cycle life, and, again, the specific energy density, are the most important. Additionally, we find that hydrometallurgical recycling of Li-S batteries could be beneficial for lowering mineral resource impacts but not necessarily for lowering other environmental impacts.

KEYWORDS: lithium-sulfur batteries, large-scale energy storage, life cycle assessment, recycling, climate change



INTRODUCTION

To reach global climate targets and meet the energy requirements of a growing population, society needs to reduce its dependency on fossil fuels. Renewable energy sources, such as wind power and solar power, can contribute to achieving these targets.¹ However, because solar power and wind power are of variable nature,² they need to be accompanied by energy storage technologies.³ Batteries are used for large-scale energy storage systems due to, for example, their scalability and rapid response time.^{3,4} Developing batteries with low environmental impact is therefore important to reach necessary targets. Additionally, most battery types require raw materials for which the demand is expected to increase. Thus, future battery design and utilization must be coupled with sustainable resource management, particularly for geochemically rare metals.⁵

The lithium-ion battery (LIB) is currently the dominating rechargeable battery technology and is one option for large-scale energy storage. Although LIBs have several favorable properties, such as relatively high specific energy density, long cycle life, and high safety,⁶ they contain varying numbers of rare metals; lithium is present by definition, whereas elements such as cobalt and nickel can be found in some LIB chemistries.⁷ There are also LIB chemistries that contain lithium as the only rare metal, for example, the lithium iron phosphate chemistry. Life cycle assessment (LCA) studies

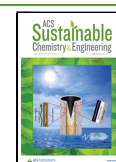
have shown that LIBs can impact the environment considerably throughout their life cycle even when manufactured at a large scale, for example, during battery cell production (in particular, given fossil-based electricity mixes) and nickel sulfate production (for nickel-containing LIBs).^{8,9} For these reasons, there are many ongoing efforts to research and develop new battery chemistries and concepts.¹⁰

One next-generation battery technology considered promising is the lithium-sulfur (Li-S) battery, fundamentally based on a lithium metal foil anode and a sulfur-containing cathode.¹¹ Besides having a high specific energy density,¹² Li-S batteries commonly do not contain any other rare elements than lithium. Because Li-S batteries are not produced at an industrial scale yet,¹³ there are still opportunities to steer Li-S battery development toward minimizing environmental and resource impacts. A handful of LCAs on Li-S batteries have been conducted in recent years with different scopes (Table S1). Deng et al.¹⁴ studied a Li-S battery with a lithium metal anode and carbon black-thiosulfate cathode intended for

Received: January 9, 2023

Revised: May 29, 2023

Published: June 16, 2023



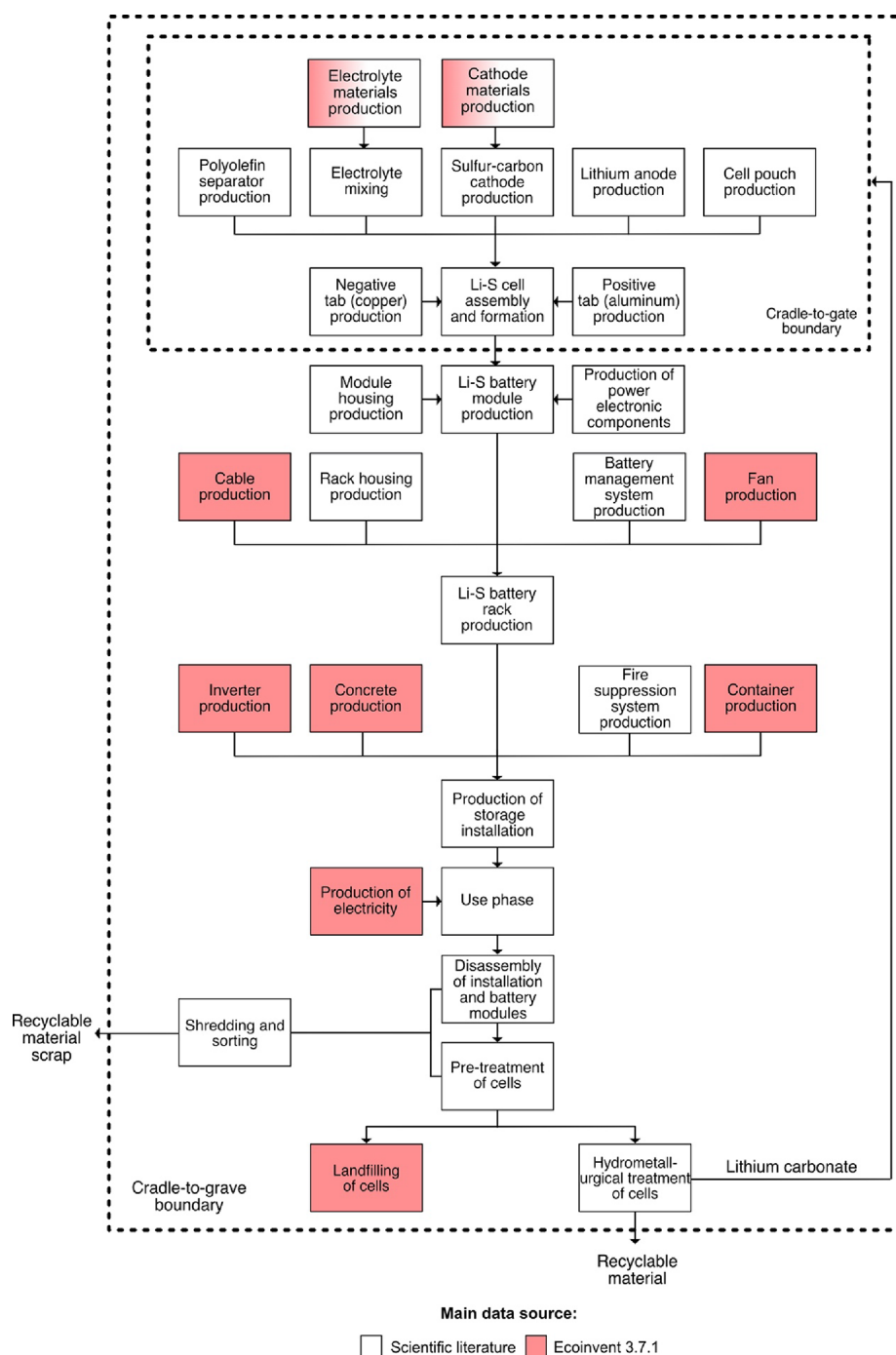


Figure 1. Flowchart for the Li-S battery production system with the two system boundaries illustrated by dashed lines. Landfilling and hydrometallurgical recycling represent two different EoL treatments for the cell materials, where one or the other was included depending on the scenario; see Table 1. The colors show the main type of underlying data source, that is, the scientific literature (white) and the Ecoinvent database v 3.7.1 (red). For some processes, the modeling was based on both these types of data sources, which are shown in gradient red/white color.

automotive applications. Cerdas et al.¹⁵ carried out a comparative LCA between a Li-S battery and a LIB, both used as traction batteries. Arvidsson et al.¹⁶ looked into a number of potential environmental improvements of a state-of-the-art Li-S cell with a lithium metal anode and a carbon-sulfur cathode in a cradle-to-gate study. Wolff et al.¹⁷ assessed the cradle-to-grave impacts of a Li-S cell developed within the HELIS project. Lopez et al.¹⁸ conducted an LCA of Li-S cells with five different advanced cathode materials on battery pack level. Benveniste et al.¹⁹ carried out a cradle-to-grave LCA,

where they also compared a Li-S battery and a LIB used as traction batteries. Finally, Barke et al.²⁰ considered an all-solid-state Li-S battery with a lithium metal anode, a solid sulfur cathode, and different solid-state electrolytes. The battery is intended to be used in an electric aircraft, and both environmental and social impacts were assessed.

With this study, our first aim is to identify development paths that could result in improved environmental and resource performance of future Li-S batteries produced at a large scale. To this end, a number of new unit process data sets

for emerging battery materials were provided, specifically lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), lithium triflate (LiOTf), polyethylene glycol (PEG), sulfolane (SL), and a mesoporous carbon material. Furthermore, all previous LCA studies of Li-S batteries that included the use phase considered electric vehicle applications (Table S1) because the high specific energy density of Li-S batteries could enable longer driving ranges.¹¹ However, Li-S batteries might also be valuable as storage solutions for the variable renewable energy in the electricity grid.²¹ The second aim is therefore to assess the environmental and resource scarcity impacts of Li-S batteries used for large-scale stationary energy storage. Finally, we want to investigate the potential environmental and resource benefits from recovering rare metals, in this case, lithium. The third aim is thus to assess the environmental and resource impacts from dedicated recycling of lithium from Li-S battery cells compared to a landfilling scenario. Scientists (both fundamental and applied) involved in battery research, as well as companies developing Li-S batteries, funding bodies of battery research, and LCA practitioners studying batteries, are the primary intended audience of this study.

MATERIALS AND METHODS

Modeled System. Two system boundaries were considered (Figure 1). The first was a cradle-to-gate boundary, where the cradle was the extraction of raw materials and the gate was that of the Li-S cell manufacturing facility. The functional unit (FU) in this case was set to 1 kWh of the theoretical storage capacity from a pouch cell. The second system boundary was a cradle-to-grave boundary, where the cradle was again raw material extraction but the grave was the disposal of waste and the handing of secondary materials from the recycling of Li-S batteries and other components. The use phase entails large-scale energy storage of wind-based electricity using the Li-S batteries; thus, an FU of 1 MWh of AC electricity delivered to the grid over 20 years was selected, as also applied in other LCAs of batteries for stationary storage.^{22–24} The manufacturing of Li-S cells was based on the work by Chordia et al.,⁸ see a complete description in Section S3.6. A configuration shown in Ainsworth²⁵ was used to model the energy storage installation. Battery cells are placed in a housing structure together with power electronic components, forming a battery module.²⁶ Battery racks are formed by placing modules in a shelf system with a battery management system (BMS) and a cooling system. The racks are placed inside an intermodal shipping container, standing on a concrete foundation, with a fire suppression system and inverters,²⁷ together constituting the installation (Figure 2). In reality,

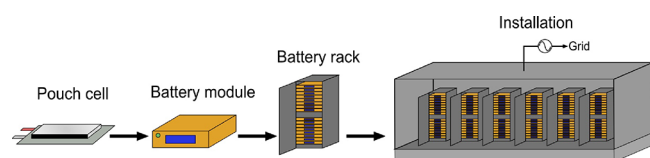


Figure 2. Schematic illustration of pouch cells being placed in battery modules, which in turn are assembled in battery racks. Multiple racks are then placed in a container, which constitute a stationary energy storage installation. Inverters and the fire suppression system are not shown in the figure. For the exact number of modules and racks for one installation, see Section S3.11.

battery cells that have lost a predefined percentage of their initial capacity are sent to end-of-life (EoL) treatment and replaced. Here, as a simplification, all battery modules were assumed to be replaced after a predefined number of cell cycles equal to their cycle life (Table 1). After 20 years, the installation with all its components was assumed to be dismantled and sent to EoL treatment, yielding both waste and recycled materials. Table S30 provides the dimensioning, operational

parameters for the installation, and number of battery module changes per FU.

Modeling with two different system boundaries was done to enable comparisons on two levels: (i) against other cell chemistries, such as LIBs, on the cell level and (ii) against other batteries used for stationary energy storage at the application level. Only one multifunctional process was modeled explicitly within this work, trimethylsilyl chloride production, with the coproducts dimethyldichlorosilane, methyltrichlorosilane, dichloromethylsilane, and dimethylchlorosilane. Because of the challenges of estimating future prices,²⁸ mass-based allocation was selected as the partitioning approach for this process, following Wickerts et al.²⁹ In addition, several data sets obtained from the Ecoinvent database were already preallocated, which is often the case in LCA databases.³⁰ These data sets were applied without altering the partitioning.

Prospective Modeling of Production and Use Phase. Li-S batteries are currently produced at the lab scale or as prototypes, which correspond to a manufacturing readiness level (MRL) of 4–5.³¹ When the production volume increases over time, technical characteristics might improve, and also, surrounding systems can change, such as the electricity supply. Prospective LCA can be used for assessing early-state technologies at a future point in time, representing the mature state.³² In this study, six scenarios of future large-scale mature production, use, and EoL handling of Li-S batteries were considered (Table 1). All scenarios assume large-scale production (MRL = 10), which requires upscaling of data from smaller-scale operations. Although the exact timing of such production is uncertain, it is unlikely to occur before 2030. For emerging battery materials, the framework by Piccinno et al.³³ was used for upscaling the production. For the battery cell production, a large-scale “gigafactory” model of LIB production⁸ was adapted to Li-S cell production. More detailed information on the upscaling can be found in Section S3.6. All other upstream materials were assumed to be produced as they are today, meaning that their production was modeled as in Ecoinvent v3.7.1. This implies that, for example, sulfur, being an important element in Li-S cells, was assumed to be produced as a byproduct from petroleum refinery operations to a large extent.

In the “Base scenario”, electricity supply to all production processes was modeled with a medium emission intensity mix, which here represents a future worst case in terms of, for example, carbon intensity. This mix was modeled as the EU mix available in Ecoinvent v3.7.1. Furthermore, electricity required to maintain a functioning installation when not in use,²⁴ for example, through thermal management, was also modeled with the medium emission intensity mix. However, the electricity stored in the use phase was assumed to be wind power because the function of the stationary energy storage installation in this study is to store and deliver such electricity. In effect, this means that the losses occurring during the use of the energy storage installation were wind power. The cell materials are based on a state-of-the-art Li-S cell composition¹⁶ but with updated modeling of several processes; see Section S3.6. That cell consists of a lithium metal foil anode that also functions as a current collector, and a composite cathode based on elemental sulfur, the mesoporous carbon material CMK-3, as well as a polyvinylidene difluoride (PVDF) binder. The electrolyte consists of the salts LiTFSI and lithium nitrate dissolved in the organic solvents dimethoxyethane (DME) and 1,3-dioxolane (DOL). The cell also contains a separator made of polypropylene (PP) and polyethylene (PE), as well as an aluminum-based current collector for the cathode. A specific energy density of 150 Wh/kg at the cell level and a cycle life of 1500 cycles were selected as performance starting points.²⁵ Regarding round-trip efficiency, data specific to Li-S batteries were not available. Instead, we apply 70% as reported by Schimpe et al.³⁴ for stationary energy storage solutions with LIBs.

In the “Material selection scenario”, the cell materials are different from the “Base scenario” and are instead based on a patent by Kolosnitsyn and Karaseva³⁵ with different electrolyte and cathode materials: LiOTf dissolved in SL functions as electrolyte, and the composite cathode consists of elemental sulfur, carbon black (CB), and a PEG binder.

Table 1. Characteristics of the Prospective Scenarios^a

scenario parameter	base scenario	material selection scenario	energy system scenario	technical performance scenario	recycling scenario	combined scenario
electricity supply for production, installation, operation (other than storage losses), and EoL	medium emission intensity mix	medium emission intensity mix	low emission intensity mix	medium emission intensity mix	medium emission intensity mix	low emission intensity mix
cell materials and composition ^b	A1	B1	A1	A2	A1	B2
specific energy density (Wh/kg)	150	150	150	500	150	500
cycle life	1500	1500	1500	3000	1500	3000
round-trip efficiency (%)	70	70	70	90	70	90
EoL treatment of Li-S cells	landfilling	landfilling	landfilling	landfilling	hydrometallurgical treatment	hydrometallurgical treatment

^aThe scenario parameters below the two solid lines are only relevant at the cradle-to-grave level. ^bDescriptions of A1, A2, B1, and B2 are found in Table 2.

In the “Energy system scenario”, all electricity supplied to production processes and the operation of the installation was assumed to be wind power, which represents an example of a low emission intensity mix.¹

In the “Technical performance scenario”, a specific energy density of 500 Wh/kg at the cell level was assumed, which might be achievable in the future.^{11,25} The cell composition by Robinson et al.¹² was considered because that enables a theoretical specific energy density just over 500 Wh/kg. In addition, the cycle life was doubled, and a round-trip efficiency of 90% was assumed.⁴

Finally, a “Combined scenario” with altered parameter values from all four latter scenarios was considered. This scenario should represent a best case for Li-S battery production and application in stationary energy storage. All scenario characteristics are presented in Table 1, and the battery cell materials and compositions are provided in Table 2. The “Recycling scenario” is described in the next section.

Table 2. Cell Materials and Compositions^a

cell component	cell materials A	cell materials B	cell composition 1 ^b (wt %) ¹⁶	cell composition 2 ^a (wt %) ¹²
anode	lithium foil	lithium foil	8.5	16
sulfur source (cathode)	elemental sulfur	elemental sulfur	4.8	18
conductive additive (cathode)	CMK-3	CB	3.8	2.7
binder (cathode)	PVDF	PEG	2.1	2.7
electrolyte	LiTFSI and LiNO ₃ in DOL and DME	LiOTf in SL	61	44
current collector	aluminum foil	aluminum foil	6.4	3.1
separator	PP-PE-PP	PP-PE-PP	13	9.9

^aCB = carbon black, DME = dimethoxyethane, DOL = 1,3-dioxolane, SL = sulfolane, LiOTf = lithium triflate, LiTFSI = lithium bis(trifluoromethanesulfonyl)imide, PE = polyethylene, PEG = polyethylene Glycol, PP = polypropylene, and PVDF = polyvinylidene difluoride. ^bThe cell composition does not add up to 100% because the cell container and tabs are not included in the table.

Prospective Modeling of End-of-Life. For the EoL stage, the overall assumption of a mature state was ascribed the meaning that Li-S batteries are widely used in society, and the lithium market is therefore heavily influenced by the Li-S market. All six prospective scenarios include collection and EoL treatment of the installation, including racks, modules, Li-S battery cells, and other components. To simplify the modeling of the EoL stage, the collection rate was set to 100% for the complete installation. This modeling can be argued to reflect a future with high demand on battery recycling, in line with

ambitions in, for example, the European Union’s Battery Directive (2006/66/EC) and its proposed follow-up regulation (EU) No. 2019/2020. The whole installation and then subsequently also the battery modules were assumed to be manually disassembled. The battery modules then yield battery cells, module housings, and power electronic components. The battery cells were assumed to be deactivated and manually disassembled. All noncell components and materials of the installation as well as certain parts of the cells (cell pouch, tabs, and the aluminum current collector) were assumed to go through a shredding and sorting process in preparation for recycling (however, this recycling was outside the scope of this product life cycle). Furthermore, for the cell remaining, we modeled two different EoL treatment processes that represent possible best and worst case scenarios. The best case was modeled as hydrometallurgical treatment (in the Recycling and Combined scenarios), whereas landfilling was modeled as the worst case in all other scenarios (Table 1). A detailed flowchart of the EoL modeling as well as a complete description of the disassembly and EoL treatment processes for the cells can be found in Section S3.13.

The hydrometallurgical process constitutes a dedicated recovery of lithium carbonate from Li-S cells. This recycling was therefore modeled as a closed loop.³⁶ In a future when Li-S cells occupy a considerable share of the market, such recycling can be a way to avoid a bottleneck for their own diffusion.³⁷ In effect, it means that for the scenarios with hydrometallurgical treatment, the input of primary lithium carbonate only covers for the lithium losses occurring in the production and EoL processes of the life cycle. Oppositely, for the scenarios with landfilling, Li-S cells do not contribute to the recycling of lithium, and primary lithium was considered for the input but this time covering the full lithium demand of the cells as it is all lost in the landfill. However, any other material leaving the product life cycle was assumed to be recycled through an open loop back to material production and was modeled by the cutoff approach.³⁶ This means that the current recycled contents of all products and materials as provided by Ecoinvent v3.7.1 were assumed to remain over time, except for that of lithium.

The electricity supply to all separation and waste handling processes during EoL was modeled with the medium emission intensity mix in the Base, Material selection, Technical performance, and Recycling scenarios, whereas the low emission intensity mix was modeled in the Energy system and Combined scenarios.

Data Acquisition. As shown in Figure 1, some unit process data were based on our own data collection and modeling, whereas data for other unit processes were found in the literature or in the Ecoinvent database (v3.7.1, cutoff). For the aluminum current collector, a wrought aluminum alloy was assumed to be processed into a foil using the sheet rolling process in Ecoinvent.³⁸ The modeling of lithium foil production was based on Deng et al.¹⁴ Because lithium needs an inert or dry environment,³⁹ it was assumed that the foil production occurs inside a dry room. Energy requirements for the dry room were based on the model by Chordia et al.,⁸ and detailed data were retrieved from personal communication with the first author of that study. The production of DOL and

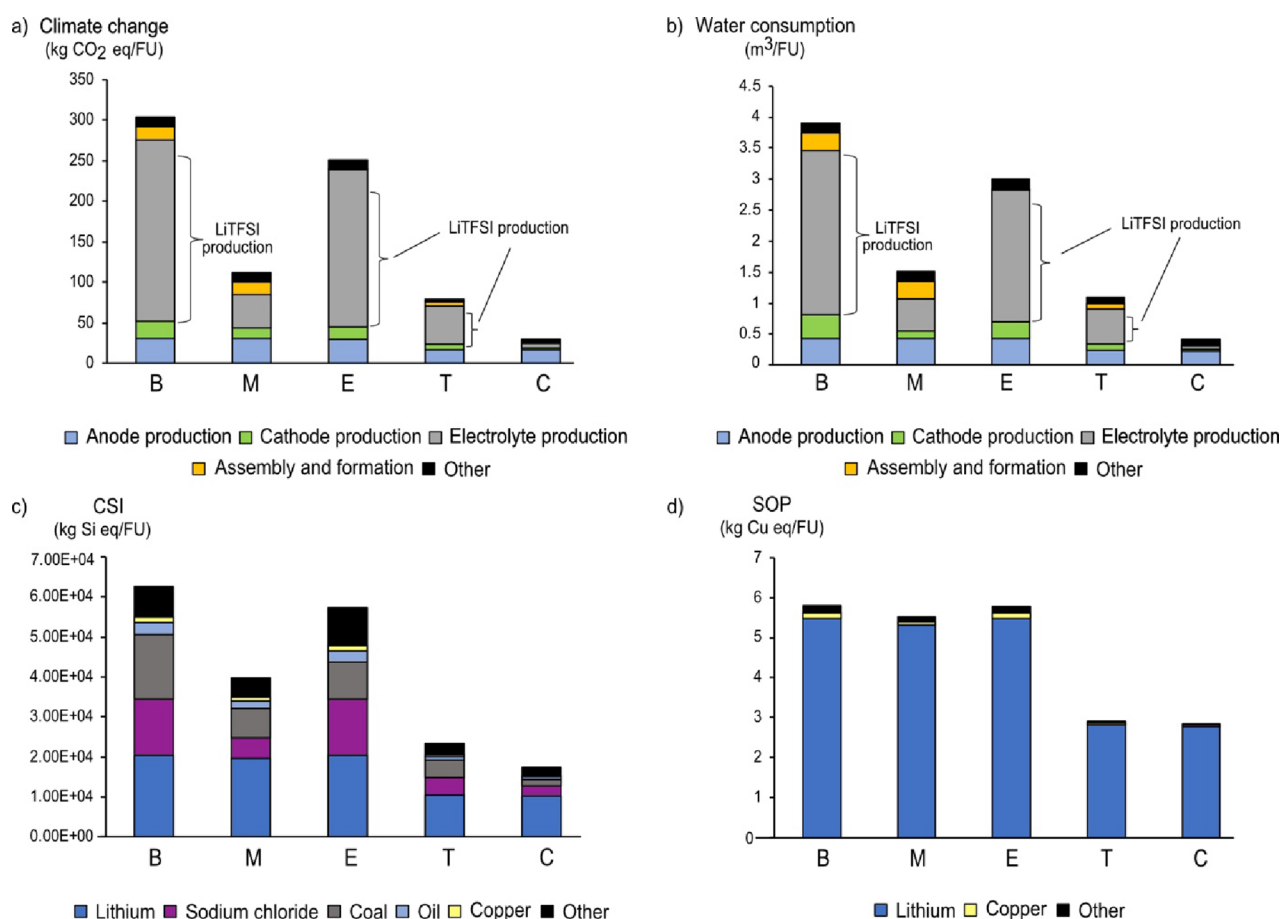


Figure 3. Cradle-to-gate results for (a) climate change, (b) water consumption, (c) the crustal scarcity indicator (CSI), and (d) the surplus ore potential (SOP) of the Li-S battery cell. The high share of impacts from LiTFSI production in the electrolyte production is shown with brackets for three scenarios in panels a and b. The FU is 1 kWh of theoretical storage capacity. B = Base scenario, M = Material selection scenario, E = Energy system scenario, T = Technical performance scenario, and C = Combined scenario.

lithium nitrate was based on Deng et al.¹⁴ Data for the production of carbon black and elemental sulfur were obtained from Ecoinvent. CMK-3 and SBA-15 production based on Jun et al.⁴⁰ was modeled. To produce SBA-15, a polymer called Pluronic P123 is required, for which a production based on Moulijn et al.⁴¹ and Noshay and McGrath⁴² was modeled. The binder in the Base, Energy, and Technical scenarios, PVDF, was approximated as polyvinylfluoride, for which production data were obtained from Ecoinvent. Production of PEG based on Dausgs et al.⁴³ was modeled. A trilayer separator consisting of polypropylene (PP) and polyethylene (PE) was modeled based on Li et al.⁴⁴ and Deng et al.¹⁴ The pouch consisting of aluminum and PE, as well as the production of the tabs, was modeled using Ellingsen et al.⁴⁵ as the data source.

Production of battery modules was based on the “3 kWh Rack Mounted Battery” in Ainsworth²⁵ regarding outer dimensions and weight of the battery cells. However, because the weight distribution of other components is not specified in Ainsworth,²⁵ data for battery modules in Peters and Weil²⁶ were used but modified to match the cell weight modeled in this study. The rack production was based on Peters and Weil.²⁶ Because no data for BMS production specifically developed for stationary applications could be found, the production process in Ellingsen et al.⁴⁵ was used for the BMS composition, whereas Nordelöf et al.⁴⁶ and Nordelöf⁴⁷ were used for some BMS subcomponents’ composition and assembly. The total BMS quantity required for the installation was obtained from Ellingsen et al.⁴⁵ The need for a fire suppressant and cooling system per kWh of storage capacity was based on Pellow et al.²⁷ Data for the production of inverters, the container, the concrete foundation, and the cooling system were obtained from Ecoinvent. Data from Tillman et al.⁴⁸ were

obtained for the shredding and sorting process. The deactivation, separation, and grinding of Li-S cells (all belonging to the process “pretreatment of Li-S cells” in Figure 1) were modeled based on Schwich et al.⁴⁹ Data for landfilling were obtained from Ecoinvent, whereas a hydrometallurgical treatment process based on Schwich et al.⁴⁹ was modeled.

For production processes for materials (e.g., LiTFSI, LiOTf, PEG, and SL) and the recycling process, for which complete production data representing large scale were missing, unit processes based on the approach suggested by Piccinno et al.³³ were constructed. The suggested approach is used for scaling lab processes into large-scale production; however, because complete lab protocols were not found for all materials, additional approaches were used in combination with this scaling procedure. For cases where no data on material inputs and outputs were available, stoichiometry was used together with a yield factor. Although it is difficult to assume a universal reaction yield if no process-specific yield is available, 95% was assumed in such cases.⁵⁰ Parameters such as reaction time and reaction temperature are required to calculate, for example, the heating demand.³³ When these were not available, specific assumptions were made for each unit process as described in the Supporting Information (SI). If outputs other than the main product are generated, they were categorized as either co- or byproducts. Compared to the main product, coproducts have similar economic value, whereas byproducts have significantly lower economic value.³³ In the former case, mass-based partitioning of burdens was applied, whereas in the latter case, because of uncertainties with regard to future utilization, waste treatment was assumed. Solvent recycling by distillation, which is common in the industry,⁵¹ was considered for all relevant processes, following

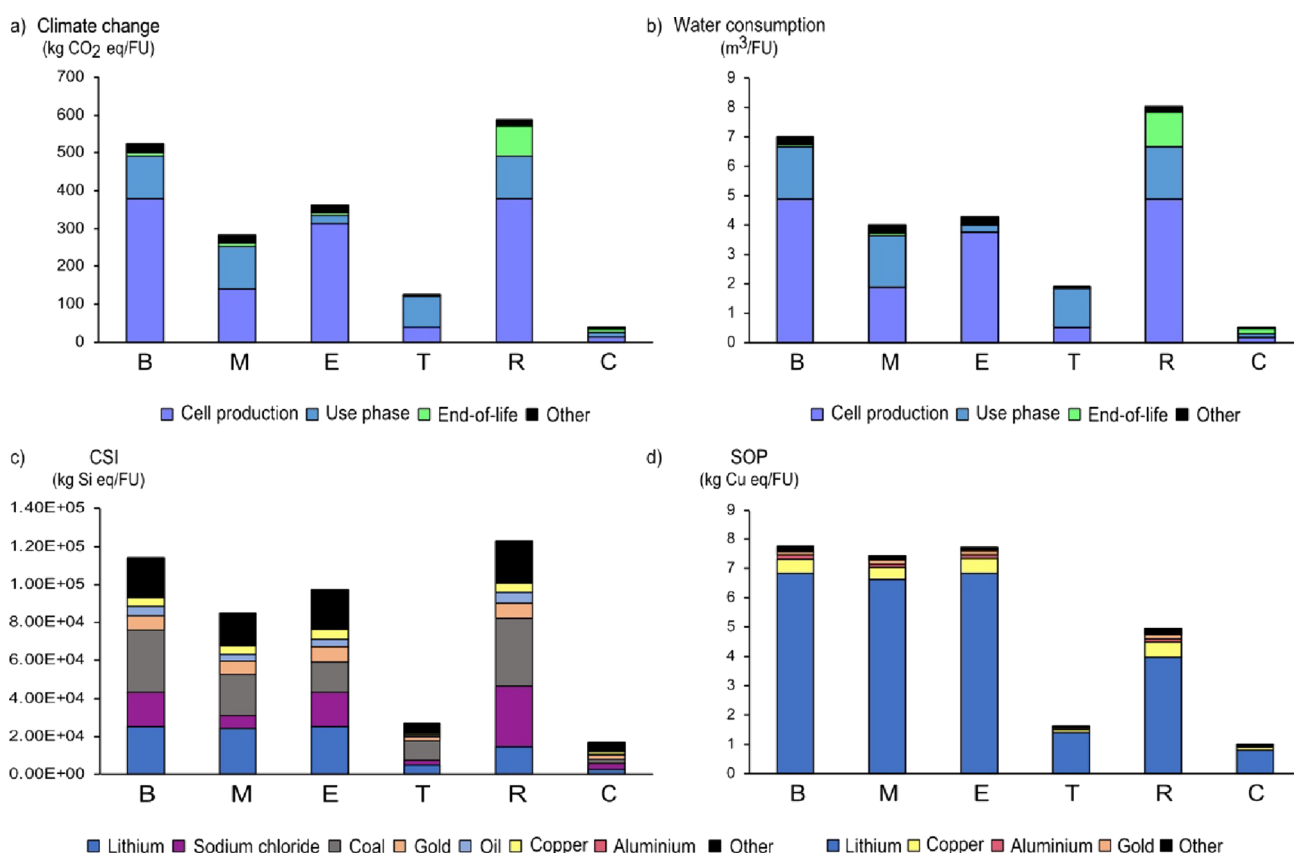


Figure 4. Cradle-to-grave results for (a) climate change, (b) water consumption, (c) the crustal scarcity indicator (CSI), and (d) the surplus ore potential (SOP) of the Li-S battery used for stationary energy storage. The FU is 1 MWh of AC electricity delivered to the grid over 20 years. B = Base scenario, M = Material selection scenario, E = Energy system scenario, T = Technical performance scenario, R = Recycling scenario, and C = Combined scenario.

Piccinno et al.³³ Waste amounts were calculated from mass balances. Because future fugitive emissions are challenging to estimate, only direct process emissions were included. A complete description of this modeling procedure can be found in Section S2 in the SI.

Life Cycle Impact Assessment. Nine impact categories were assessed in this study: climate change, mineral resource use, water use, acidification, eutrophication, ozone formation, fine particulate matter formation, stratospheric ozone depletion, and fossil resource scarcity. Climate change and mineral resource use are particularly relevant impact categories for batteries.^{52,53} With respect to climate change, the purpose of this battery application is to enable low-carbon energy supplies, such as wind power. Climate change is here modeled as the change in radiative forcing using characterization factors from IPCC AR5 with a 100 year time horizon.⁵⁴ For mineral resource use, this is an aspect where Li-S batteries are considered promising compared to other battery technologies such as LIBs.^{14,15} Because of the future-oriented perspective of the study, longer-term impacts of mineral resource use were of interest. The crustal scarcity indicator (CSI) is aligned with this perspective and was therefore applied in this study.⁵⁵ To obtain a thorough understanding of resource impacts, the CSI was complemented with another resource metric: the surplus ore potential (SOP), which is part of the ReCiPe 2016 package. Even though there are time-related parameters used in the SOP, it was selected as the second resource metric because of its comprehensiveness in terms of elementary flows included.⁵⁶ Water consumption has also been pointed out as a potentially important impact category for battery LCAs⁷ and was therefore also included, with water use characterization factors from the ReCiPe package.⁵⁷ Results for acidification, eutrophication, ozone formation, fine particulate matter formation, stratospheric ozone depletion, and fossil resource scarcity are provided in Section S4 in the SI, all using characterization factors from ReCiPe 2016. Most of these are highly correlated with the

climate change results. Impact assessment methods were applied as implemented in the OpenLCA package (version 2.1.2).

RESULTS AND DISCUSSION

Climate Change and Water Consumption at the Cell Level. The climate change and water consumption results show similar patterns (Figures 3a,b). For the Base scenario, it can be noted that, for cell materials, LiTFSI (including its production) is the main contributor; when switching to another electrolyte (containing a different electrolyte salt) as is done in the Material selection scenario, the impacts are reduced by more than 60%. LiTFSI has such a high impact due to a complex multistep synthesis route (Figure S1), where some of the upstream inputs have high impacts. For example, trimethylsilyl chloride production alone stands for 30% of the LiTFSI impact. LiOTf, the alternative electrolyte salt in the Material selection scenario, has a simpler synthesis route (Figure S2) and considerably lower impact than LiTFSI. Therefore, an LiOTf-based electrolyte could be a better choice for future Li-S cells if performance criteria, such as high ion conductivity and chemical stability,⁵⁸ can be met.

Another important parameter in the cradle-to-gate comparison is the specific energy density. Comparing the Base and the Technical performance scenarios, shifting to 500 Wh/kg entails a reduced number of cells needed to store 1 kWh of energy. This is true even though a larger amount of active material is likely required (Tables 1 and 2). In addition, using cleaner electricity sources, such as wind power in the Energy system scenario, has a slightly positive effect on the impacts,

and combining the change of cell materials, utilizing wind power, and increasing the specific energy density, as in the Combined scenario, will yield the lowest impacts.

Comparing the cradle-to-gate impacts to other Li-S studies, a climate impact of 117 kg CO₂ equiv/kWh was provided in Cerdas et al.,¹⁵ whereas Arvidsson et al.¹⁶ reported an interval of 17–230 kg CO₂ equiv/kWh for the same impact category, depending on the scenario. Lopez et al.¹⁸ reported climate change impacts in the range of 50–250 kg CO₂ equiv/kWh for a wide range of advanced cathode materials and differing electrolyte/sulfur ratios. The cell manufacturing is the dominant phase in Cerdas et al.¹⁵ and in the small-scale cell production scenario in Arvidsson et al.,¹⁶ whereas in the large-scale cell production scenario in Arvidsson et al.,¹⁶ LiTFSI production is dominant. The present study, which models large-scale cell production in the form of a gigafactory, confirms a reduction of climate impacts when the production is scaled up and the then-dominant role of LiTFSI production (60–70% of impacts in all applicable scenarios).

As compared to other battery chemistries and based on the data herein, Li-S batteries could achieve similar climate impacts, for example, 50–110 kg CO₂ equiv/kWh for NMC-graphite LIBs produced at a large scale⁸ but only with an LiOTf-based electrolyte and/or an increase in specific energy density. For water consumption, Li-S batteries perform similarly to or better than the LIBs modeled in Chordia et al.⁸

Mineral Resource Use at the Cell Level. For mineral resource use, the contribution analysis is shown on an elementary flow-level because individual contributions of the metals and other materials are of interest rather than process contributions (Figures 3c,d). With respect to CSI results, shifting from LiTFSi to LiOTf and increasing the specific energy density should be considered to obtain lower impacts. Regarding SOP, only an increased specific energy density reduces the impact significantly. For all scenarios, lithium, sodium chloride, and coal are the three main contributors in the CSI results, whereas lithium makes up almost the entire impact in the SOP results. Lithium has a high characterization factor in both impact assessment methods and contributes to the results to a varying extent, depending on cell composition and specific energy density. Although sodium chloride, which is utilized in, for example, electrolyte production, has a significant contribution to the CSI in the Base and Energy system scenarios, it should be noted that the compound is not scarce in general, as there are vast amounts of sodium chloride in the oceans. The high contribution is however due to a combination of the large usage of the compound and the CSI indicator being based on the abundance of materials in Earth's crust.⁵⁵ Coal is related to electricity production in background processes, and the slight differences in the results depend on differences in cell materials, cell composition, and electricity source.

No other Li-S studies report mineral resource use results based on the CSI and/or SOP methods. Li-S batteries seem to have an advantage compared to NMC-graphite batteries, which have a reported CSI at 65,000 kg Si equiv/kWh and an SOP at 10 kg Cu equiv/kWh.⁸ LIBs utilizing a lithium-iron-phosphate (LFP) cathode could be even more interesting to compare with because of the absence of cobalt and nickel, but we could not find any study of LFP batteries that use either the CSI or the SOP method.

Climate Change and Water Consumption for the Full Life Cycle. Climate change and water consumption impacts

show a similar pattern also for the cradle-to-grave perspective (Figure 4a,b). Cell production, which includes both the actual production of cells and raw material extraction, influences the total results the most in the Base, Material selection, Energy system, Recycling, and Combined scenarios, whereas the use phase is the largest contributor for the Technical performance scenario. Use phase impacts originate from system losses related to storing the wind-based electricity as well as electricity required to operate the installation.

Comparing the Material selection to the Base and Energy system scenarios, it had the lowest impact among the three for the cradle-to-gate study, and it also had the lowest impact for the full life cycle. However, the difference in impacts between the Material selection and Energy system scenario is smaller than on the cell level: the advantage of the Material selection scenario from substituting LiTFSI is countered by the reduced use phase impact in the Energy system scenario due to the shift from medium to low emission intensity mix for the electricity source for all foreground processes.

The Recycling scenario yields the highest impact of all scenarios, as the hydrometallurgical treatment contributes notably to the total impact, implying that there is an environmental cost in terms of climate impacts and water consumption to recover the lithium contained in the cell and process it to lithium carbonate with the required purity. This result is surprising; however, the high impact is mainly caused by the large amounts of input materials needed, such as sodium hydroxide, as well as treating the high waste amount that is generated from the process. The Technical performance and the Combined scenarios yield a lower impact for both impact categories due to an increased specific energy density, longer cycle life, and lower system losses in the Technical performance scenario, as well as wind power in the Combined scenario. However, the use phase is the dominant life cycle phase in the Technical performance scenario because the electricity source was modeled with a medium emission intensity mix; see Table S31 for details.

The cycle life is dependent on how the battery is used, that is, dependent on parameters such as the depth of discharge (DOD) and charge and discharge rates.²⁷ For example, a lower DOD results in a longer cycle life, and vice versa. A longer cycle life would in turn result in cells needing to be replaced less frequently, which would likely reduce the impacts for all impact categories. That said, depending on parameters such as the DOD, the amount of retrieved electricity will vary, which will also affect the results. In this study, a DOD of 80% was assumed; see details in Section S3.12.

As the load profile—and thereby the number of cycles per year—is an assumption of a yearly wind pattern, it was of interest to assess what effect an altered load profile would have on the results. It is not likely that it is windy more than 300 days per year; however, less than 300 windy days is possible. Therefore, the influence on climate change in the Base and Combined scenarios was assessed, assuming 100 windy days per year, resulting in 100 cycles per year. Although the batteries do not need to be replaced as often when assuming 100 cycles per year (for the Combined scenario, no battery replacement is needed), the lower amount of electricity delivered compared to when assuming 300 cycles per year results in higher impacts per functional unit for both scenarios.

There are no Li-S cradle-to-grave studies assessing large-scale energy storage, but the results can be compared to those for other battery chemistries. da Silva Lima et al.²⁴ modeled

large-scale energy storage for the integration of renewable energy to the grid using an NMC-graphite LIB and a vanadium redox flow battery (VRFB), where the assessment of the latter was based on data from Weber et al.²³ For climate change impacts, da Silva Lima et al.²⁴ reported values of 95 kg CO₂ equiv/MWh for NMC-graphite LIBs and 100 kg CO₂ equiv/MWh for VRFBs. Only the Technical performance and Combined scenarios have impacts of similar magnitude, implying that technical performance parameters (specific energy density, cycle life, and round-trip efficiency) are important parameters to improve for making Li-S batteries environmentally competitive.

Mineral Resource Use for the Full Life Cycle. For the CSI results presented in Figure 4c, similar profiles as for the cradle-to-gate scope are seen. However, there is a larger difference in impacts between the first three scenarios and the Technical performance and Combined scenarios because, apart from the higher specific energy density in these two, the longer cycle life also results in a lower number of cells required to deliver the FU. Developing Li-S cells with high specific energy that are used in such a way that they can be cycled many times is therefore more important than changing cell components, for example, the electrolyte salt, for reducing the CSI. Notably, the Recycling scenario has a similar impact to that of the Base scenario, implying that landfilling and hydrometallurgical treatment have similar impacts. The reason for this is that the CSI indicator is inclusive regarding characterized flows, so although the contribution from lithium is reduced because of hydrometallurgical treatment, contributions from other materials added in the recycling process increase. Furthermore, it can be noted that gold has a significant contribution in the Base, Material selection, Energy system, and Recycling scenarios. Gold is required for certain components in the installation's BMS, which is why the metal only appears in the cradle-to-grave scope results. The BMS amount needed is the same for all scenarios, but because the Technical performance and Combined scenarios deliver more electricity, the relative impact from gold becomes lower.

The SOP results shown in Figure 4d also have a pattern similar to that of the cradle-to-gate scope. There are no significant differences between the Base, Material selection, and Energy system scenarios, whereas the impact for the Recycling scenario is almost 40% lower due to the recycling of lithium carbonate. The SOP indicator does not characterize as many flows as the CSI indicator, which means that the impacts of some inputs to the hydrometallurgical treatment are not considered by this indicator. Again, the impacts for the Technical performance and Combined scenarios are the lowest because of the improved technical performance. For both the CSI and the SOP, copper and other resources have slightly larger relative contributions compared to those for the cradle-to-gate results. The higher copper impacts originate from power electronic components on the module and rack level. Gold does not contribute notably to the SOP results.

Some of these results can be benchmarked to those of da Silva Lima et al.,²⁴ who reported an SOP of 5.9 kg Cu equiv/MWh for both the NMC-graphite and VRFB battery. These results are lower than for the three worst scenarios in Figure 4d, similar to the Recycling scenario, and considerably higher than for the Technical performance and Combined scenarios. This indicates that Li-S batteries could be a promising alternative from a resource point of view at the cradle-to-grave level, but it depends on the cell characteristics. Again,

LFP batteries constitute the more promising type of LIBs resource-wise, but no study of LFP batteries using the CSI and/or SOP could be found.

LIMITATIONS AND FUTURE STUDIES

In summary, to reduce impacts on the cradle-to-gate level, it is most important to substitute the LiTFSI salt and to increase the specific energy density. For the cradle-to-grave level, however, the benefit that arises from changing electrolyte salt is minor compared to electricity losses during the energy storage and energy requirements to operate the installation. Consequently, electricity losses should be reduced in combination with developing Li-S cells with high specific energy density that are used in a way that ensures a long cycle life. Additionally, comparing landfilling to hydrometallurgical treatment, the latter one is clearly beneficial for recovering lithium.

It should be noted that the Ecoinvent database does not contain prospective data sets. All such data applied therefore effectively constitute status-quo scenarios, which is a limitation of this study. Although the status quo might be reasonable in some cases, some materials that are today produced as byproducts from fossil resources, such as sulfur,⁵⁶ may have to carry their own environmental burden in a future with less fossil fuel extraction. This could result in recycling processes, such as the hydrometallurgical one modeled in this study, becoming increasingly important to lower impacts of material extraction. However, this study shows that the hydrometallurgical treatment can have high environmental impacts. We therefore recommend that efforts are made to develop less impacting recycling and waste treatment processes for Li-S batteries.

Another input material that supply might change in the future is lithium. Chordia et al.⁵⁹ showed notable differences in impacts between current and near-term lithium supplies. For example, lithium from Atacama Desert brines might get more than twice the current climate impact and water use due to declining brine grades. Contrary, lithium from spodumene mineral might get almost half the current climate impact if mined with less fossil energy. Investigating how impacts of Li-S cells change with future lithium supplies is recommended for future research.

Wind power storage was modeled in this study, but the storage of solar power would be interesting to consider in future studies. Because wind power and solar power have similar impacts per kWh for several impact categories,¹ the impacts of their respective storages might also become similar. Furthermore, in addition to the comparison between the Li-S battery and other battery energy storage systems performed in this study, comparisons to different energy storage technologies, such as hydrogen,⁶⁰ would be interesting for future studies. Future studies might also investigate the influence of changing the partitioning approach, for example, to economic allocation by estimating future prices,⁶¹ or by applying the main product bears all burden approach as a worst case scenario.¹⁶

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.3c00141>.

A comparison between LCAs of Li-S batteries (Section S1, Table S1), description of the prospective modeling

approach for cell materials and recycling (Section S2), specific process descriptions and unit process tables (Sections S3.1–S3.14, Tables S2–S39), and cradle-to-gate as well as cradle-to-grave results for all impact categories (Section S4, Tables S40 and S41) (PDF)

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Notes

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ACKNOWLEDGMENTS

We would like to thank the Swedish Energy Agency (grant number 50099-1) and the Vinnova competence center Batteries Sweden (BASE) for financially supporting this study.

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