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Energy use and climate change improvements of Li/S batteries based on life cycle assessment

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HIGHLIGHTS

- LCA showing how to improve energy use and climate change of Li/S cell production.
- Energy use and climate change impact can be reduced by 54 and 93%, respectively.
- Important to reduce cell production electricity and source renewable electricity.
- Best-case climate change is similar for Li/S and lithium ion batteries.

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ABSTRACT

We present a life cycle assessment (LCA) study of a lithium/sulfur (Li/S) cell regarding its energy use (in electricity equivalents, kWh_{el}) and climate change (in kg carbon dioxide equivalents, CO₂ eq) with the aim of identifying improvement potentials. Possible improvements are illustrated by departing from a base case of Li/S battery design, electricity from coal power, and heat from natural gas. In the base case, energy use is calculated at 580 kWh_{el} kWh⁻¹ and climate change impact at 230 kg CO₂ eq kWh⁻¹ of storage capacity. The main contribution to energy use comes from the LiTFSI electrolyte salt production and the main contribution to climate change is electricity use during the cell production stage. By (i) reducing cell production electricity requirement, (ii) sourcing electricity and heat from renewable sources, (iii) improving the specific energy of the Li/S cell, and (iv) switching to carbon black for the cathode, energy use and climate change impact can be reduced by 54 and 93%, respectively. For climate change, our best-case result of 17 kg CO₂ eq kWh⁻¹ is of similar magnitude as the best-case literature results for lithium-ion batteries (LIBs). The lithium metal requirement of Li/S batteries and LIBs are also of similar magnitude.

1. Introduction

Hybrid and electric vehicles (xEVs) are an emerging technology with the potential to reduce the use of fossil fuels. Life cycle assessment (LCA) [1–3] has been used in a number of studies to compare electric vehicles to hybrid and fossil-fueled vehicles and to investigate whether such a replacement would in fact lead to reduced environmental impacts, with a positive result given electricity produced from renewable sources [4]. For xEVs, production of the vehicle and extraction of the required raw materials for the lithium-ion batteries (LIBs) used are then the dominating life cycle phases, in contrast to fossil-fueled vehicles where the use phase typically is most impacting. In particular, the

production of the LIBs is a major contributor [4,5], and the potential increase in use of scarce metals in LIBs, such as lithium, cobalt and nickel, is problematic. Also natural graphite, the preferred LIB anode, might become scarce and is classified as critical to the European Union due to high supply risk [6]. Therefore, the development of batteries and battery concepts less dependent on scarce materials is warranted. The lithium-sulfur (Li/S) battery is one such promising technology requiring no scarce elements except for the lithium metal itself. In addition, it has a promise of higher specific energy densities (400–500 Wh kg⁻¹ at the cell level) than current LIBs (ca. 250 Wh kg⁻¹) [7,8].

While LIBs has been studied extensively by LCA [5,9–13], there is, to the best of our knowledge, only a single cradle-to-grave LCA study on

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Li/S batteries [14]. They considered a wide range of environmental impacts, including climate change, acidification and toxicity. Of the battery materials considered, the electrolyte was the largest contributor to the life cycle energy use (30%), but overall, different life cycle phases dominated different environmental impacts. Since a fossil-dominated electricity mix was assumed for the use phase, this phase dominated the climate change impacts.

Here, we perform an in-depth cradle-to-gate study on the Li/S battery cell, from the extraction of raw materials to the production of the cell. A cradle-to-gate approach is chosen over a cradle-to-grave since data on e.g. life-length and usage conditions are lacking, and hence the total energy throughput and similar parameters cannot yet be accurately estimated. Our aim is to guide Li/S cell developers and producers on how to improve environmental performance, much as Zackrisson et al. [10] did for LIBs. We consider a Li/S cell with typical current state-of-the-art materials choices [15–17]: (i) a lithium metal anode, (ii) a composite carbon/sulfur (C/S) cathode of mesoporous carbon and elemental sulfur, and (iii) a liquid organic solvent-based electrolyte.

2. Method and materials

All material and energy flows related to the cradle-to-gate production of a Li/S battery cell are quantified in terms of a functional unit (FU) reflecting the function of the product, in this case 1 kWh of specific energy storage. Flows crossing the boundary between the technosphere and the environment are calculated into environmental impact categories [18]:

$$I = \sum_{i,j} q_{i,j} C_i \quad (1)$$

where I is the environmental impact, e.g. energy use or climate change [impact FU^{-1}], q is the quantity of emitted substance or resource used [amount FU^{-1}], C is a characterization factor (CF) that reflects the impact of the emitted substance or resource used [impact amount $^{-1}$], i is the emitted substance or resource type (e.g. carbon dioxide or hard coal), and j is a process in the product's life cycle.

We perform an attributional LCA of the Li/S cell and of different improvements, effectively identical to a consequential LCA where only first-order (linear) physical flow consequences are considered [19] – or “a consequential LCA based on the attributional [LCA] framework” [20].

2.1. Technology studied and system boundaries

The Li/S cell material composition and balance (Table 1) was obtained from a cell developer, and hence no exact amounts and sources can be provided for confidentiality reasons. A generic organic liquid electrolyte of 1 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) in dioxalane/dimethoxyethane (DIOX/DME) with 0.2 M lithium nitrate (LiNO_3) as additive [15] was used. For the composite C/S cathode, a mesoporous carbon, CMK-3, and elemental sulfur [16] was employed. Other carbon materials are considered in a scenario analysis (Section

Table 1
Composition of the Li/S cell.

Component	Material	Mass [g cell $^{-1}$]
Anode	Lithium foil	~4
Cathode + Current collector	C/S composite	~4
	Aluminium foil	~3
		~9
Electrolyte	DIOX	~9
	DME	~11
	LiTFSI	~8
	LiNO_3	~0.5
Separator	Tri-layer PP-PE-PP membrane	~6
Total		~45

2.4). An aluminium foil is used as current collector for the cathode, a lithium metal foil is both active material and current collector for the anode, and the separator is a typical micro-porous polyolefin membrane made of polypropylene (PP) and polyethylene (PE) in a tri-layer configuration (PP-PE-PP). The two main differences as compared to the cell of Deng et al. [14] is that they employed a copper current collector for the anode and a C/S composite made from graphene oxide and sodium thiosulfate.

In this cradle-to-gate study, the gate is the exit of the battery cell production facility (Fig. 1). A cut-off limit of 1% of the main product mass was employed for each unit process. Water flows were excluded because of the low impact for water for the considered impact categories, e.g. $\sim 0.0005 \text{ kWh}_{\text{el}} \text{ kg}^{-1}$ and $\sim 0.001 \text{ kg CO}_2 \text{ eq kg}^{-1}$ for deionised water produced in Europe [21].

2.2. Allocation

In the production system, by-products are produced, such as chlorine gas (Cl_2) during the production of lithium from lithium chloride (LiCl) and various by-products from petroleum refining (see the Supplementary data). Since economic value is the driver of most industrial processes [22,23], economic allocation based on price was applied to partition the environmental impact between products and by-products [24]:

$$P_i = \frac{n_i x_i}{\sum_i n_i x_i} \quad (2)$$

where P_i is the partitioning factor of product i , n_i is the mass of i produced and x_i is its price. Price data (2005) were obtained from the Ecoinvent database [21]. In addition to economic allocation, in a sensitivity analysis we apply allocation by mass and no allocation, i.e. allocating all impact to the main product. Mass allocation has been advocated because mass relationships are more fundamental than the prices of products [24]. The no-allocation approach is unconventional in LCA and can be seen as a worst case for the Li/S cell with regard to allocation.

2.3. Impact categories

Two key impact categories are considered: energy use and climate change. All energy used is re-calculated into electricity equivalents [kWh_{el}] to enable comparisons between different energy flows with electricity as a ‘common currency’ [25,26]: (i) electricity is added as is, (ii) heat is converted at an efficiency of 37% to represent an electricity-generating turbine [27], and (iii) chemical energy in the form of energy carriers (e.g. diesel) and materials (e.g. polyethylene) were traced back to their respective primary sources, both renewable and non-renewable, and converted at efficiencies of 43% for natural gas, 32% for coal and 33% for crude oil, biomass and uranium [28] (Fig. S1).

Climate change [$\text{kg CO}_2 \text{ eq}$], sometimes referred to as global warming potential [29], is modelled according to the ReCiPe 2016 impact assessment method [30] with a 100-year time-frame.

In addition to these two key impact categories, due to the potential future scarcity of lithium [31], lithium use is quantified as input mass to the production system. Due to the ongoing discussion on the relevance of different methods for assessing mineral resource depletion [32,33], this impact category is not considered.

2.4. Scenario analysis and sensitivity

In addition to a base scenario (Table 1), we consider five improvement scenarios. First, battery cell production electricity requirement is reduced. Deng et al. [14] modelled an operational pilot-scale production facility and arrived at 47 kWh kg^{-1} of Li/S cell, but also a potential future industrial-scale production with a ca. 77% reduction to 11 kWh kg^{-1} , which we apply. For the C/S composite cathode,

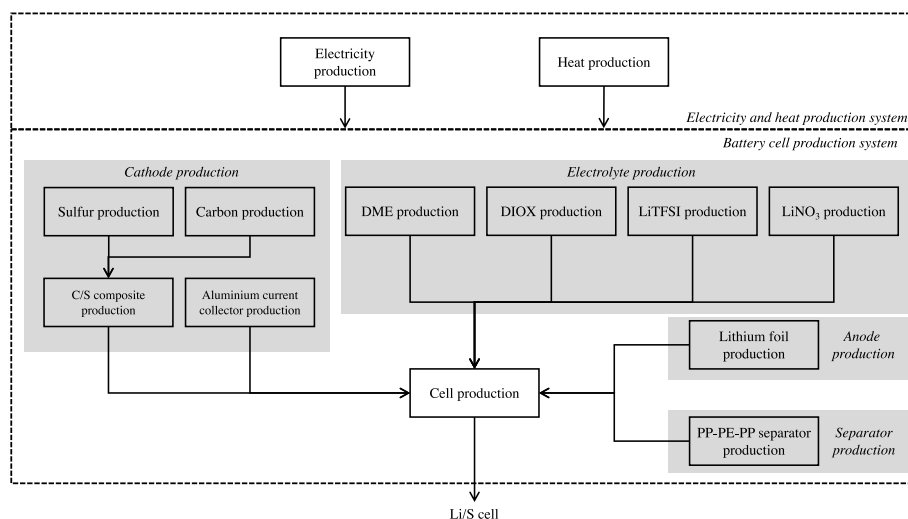


Fig. 1. Flow chart of the Li/S cell production system.

different carbon materials are continuously being assessed for Li/S cells: carbon black, carbon nanofibers, carbon nanotubes, graphite, graphene, and graphene oxide [34]. The second scenario therefore considers a shift from CMK-3 to graphene, disregarding any (current) performance drawbacks. Third, we consider a shift from graphene to the high-volume, low-cost, low-impact material carbon black, again disregarding any performance drawbacks. Fourth, the electricity and heat production systems are altered from coal power and natural gas heat to renewables – solar power and biogas heat, which generally have a lower environmental impact [35,36]. Fifth, the specific energy of the Li/S cell is targeted, with an increase in Wh kg^{-1} logically leading to lower environmental impact per kWh. The base scenario is set to 300 Wh kg^{-1} at the cell level and the improvement to a challenging 500 Wh kg^{-1} [7].

In addition to these scenarios, summarized in Table 2, the sensitivity of some parameters was investigated: the yield for specialty chemicals production (87–97%), the electricity use for specialty chemicals production (0.19–1.4 kWh kg^{-1}), the steam use for specialty chemicals production (1.2–7.7 kg kg^{-1}) [37], the methane purification energy use (0.058–0.69 kWh kg^{-1}) [38], the energy requirement for steam (2.7–3.4 MJ kg^{-1}) [39], graphene ultrasonication yield (95–99%) [40], and ultrasound effect (150–300 WL^{-1}) [40]. However, since no notably different results compared to our base scenario were obtained, average values for these parameters were used in all scenarios. Prices have fluctuated notably since 2005, almost by an order of magnitude for lithium [41] and petroleum products such as heavy fuel oil [42]. Therefore, the price for each of the 16 main products and 24 by-products were increased by a factor of 10 in a sensitivity analysis. Due to the high correlation between petroleum refinery product prices [43], all prices of petroleum products (diesel, heavy fuel oil, kerosene, light fuel oil, liquefied petroleum gas, naphtha and petrol) are increased collectively rather than separately.

2.5. Data acquisition

Li/S cell production data were obtained from Deng et al. [14]. For production of lithium foil, we use data on extrusion and lithium metal production from the Ecoinvent database [21]. For production of the C/S composite cathode, we assume a 66% sulfur loading, and a melt diffusion process [44] and heating. Due to lack of specific data on industrial melt diffusion, production data for generic specialty chemicals from Geisler et al. [37] are employed regarding yield, steam and electricity use. Elemental sulfur production data were obtained from the Ecoinvent database [45] (v2.2, no data in v3.3). No use of solvents is reported for the melt diffusion. Production of CMK-3 is based on Jun et al. [46], but scaled linearly from g to kg scale for the input materials: mesoporous silica SBA-15, sucrose, sulphuric acid (H_2SO_4) and hydrofluoric acid (HF). Water was reported as solvent for CMK-3 production. Yield, electricity and steam requirements are again from Geisler et al. [37]. The amount of HF is calculated stoichiometrically based on Fogler et al. [47], who describe its reaction with silica, mimicking the silica-based SBA-15. While data on production of sucrose, H_2SO_4 and HF are all available in the Ecoinvent database [21], production of SBA-15 is not. It is therefore based on Jun et al. [46] (scaled from g to kg) and Geisler et al. [37] regarding yield and energy requirements. Again, water was reported to be the solvent used for SBA-15 production. Three input materials are required for SBA-15 production: tetraethyl orthosilicate, hydrochloric acid (HCl) and Pluronic P123. While the two first materials are available in the Ecoinvent database [21], Pluronic P123, a co-polymer of ethylene and propylene oxide in a 40/70 M monomer ratio [48], is not. The ratio is applied to estimate the amounts needed, while the amount of seed propylene glycol required [48] is disregarded. Again, yield, electricity and steam requirements are based on Geisler et al. [37]. No use of solvent was reported for Pluronic P123 production. Detailed data regarding material input for the C/S composite, CMK-3, SBA-15 and Pluronic P123 can be found in Table 3. Although inert gas is often used during production of specialty chemicals [37], none is reported for the specialty chemicals considered here [44,46,48].

Table 2
Improvement scenarios for the Li/S cell with improvements in *italic*.

	Base case	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5
Cell production electricity	47 kWh kg^{-1}	<i>11 kWh kg^{-1}</i>	<i>11 kWh kg^{-1}</i>	<i>11 kWh kg^{-1}</i>	<i>11 kWh kg^{-1}</i>	<i>11 kWh kg^{-1}</i>
Carbon material	CMK-3	CMK-3	<i>Graphene</i>	<i>Carbon black</i>	<i>Carbon black</i>	<i>Carbon black</i>
Electricity production	Coal power	Coal power	Coal power	Coal power	<i>Solar power</i>	<i>Solar power</i>
Heat production	Natural gas	Natural gas	Natural gas	Natural gas	<i>Biogas</i>	<i>Biogas</i>
Specific energy	300 Wh kg^{-1}	300 Wh kg^{-1}	300 Wh kg^{-1}	300 Wh kg^{-1}	300 Wh kg^{-1}	<i>500 Wh kg^{-1}</i>

Table 3
Material inputs for the modelling of production of the four specialty chemicals.

	C/S composite	CMK-3	SBA-15	Pluronic P123
Input materials (kg kg ⁻¹)	Sulfur: 0.61 Carbon: 0.48	SBA-15: 1.08 Sucrose: 2.2 H ₂ SO ₄ : 0.25 HF: 1.02	Pluronic P123: 0.54 TEOS: 1.2 HCl: 1.2	Ethylene oxide: 0.33 Propylene oxide: 0.76
Solvent	None reported	Water ^a	Water ^a	None reported

^a Disregarded since no water flows are considered in this study.

Data for carbon black are available in the Ecoinvent database [21], while data for graphene are obtained from Arvidsson et al. [40] for its production by ultrasonication that requires electricity, graphite and diethyl ether solvent. 90% solvent recovery, which is not uncommon in the chemical industry [37], is assumed. Data for natural graphite and diethyl ether production were obtained from the Ecoinvent database [21]. The aluminium current conductor was modelled as aluminium wrought alloy processed to a foil, available in the Ecoinvent database [21]. Production data for DME were obtained from the Ecoinvent database [21] and for DIOX from Deng et al. [14].

For the complex production system of LiTFSI there is a lack of data sources. Therefore, we use data from Deng et al. [14], who constructed a model based on 11 input materials, and generic data from Geisler et al. [37], thus in a similarly generic way as done here for the materials in Table 3. Data for LiNO₃ were obtained from Dai [49] and data for the PP-PE-PP separator were obtained from an LCA study of a LIB [50].

Data for electricity and heat production were obtained from the Ecoinvent database [21]. In the base scenario, we assume electricity produced from hard coal. For the solar power, a multi-silicon 3 kW flat-roof installation is considered (average yield: 1099 kWh kW_{peak}⁻¹). Natural gas and biogas heat are both provided by boilers. The energy requirement for steam from Nieuwlaar et al. [39] is applied throughout the life cycle. A more detailed record of data sources is provided in the Supplementary data.

3. Results and discussion

Here, results for the two impact categories energy use and climate change are presented, followed by the results for lithium use. The effects of the different scenarios are reported, departing from a base case and continuing with different improvement scenarios according to Table 2. Comparisons to previous studies are done when possible. We also provide results showing the influence of different allocation approaches and price fluctuations.

3.1. Energy use

The base scenario results in an energy use of approximately 580 kWh_{el} kWh⁻¹ (Fig. 2a). The two most contributing processes are LiTFSI production (37%) and cell production (27%). In Scenario 1, reducing electricity use during cell production lowers the energy use by 21%. Shifting also from CMK-3 to graphene in Scenario 2 does not change the results notably. Although not investigated here, it can be noted that some other carbon nanomaterials applied in C/S cathodes are energy-intensive: 13 000–120 000 MJ kg⁻¹ for fullerenes [51] and 10 000–1000 000 MJ kg⁻¹ for carbon nanotubes [52], and thus neither are these expected to lead to a tangible improvement. In Scenario 3, a 3% reduction is achieved by switching from graphene to carbon black. Scenario 4 has no influence on energy use results. In Scenario 5, the 67% increase in the Li/S cell specific energy causes an additional linear (hence by 40%) down-scaling in all energy use per FU, as we disregard any possibly needed cell design alterations. Overall, this in total leads to a 54% reduction in energy use.

Since Deng et al. [14] performed a cradle-to-grave study, report

results per vehicle km, and reported primary energy instead of electricity equivalents, we cannot do any direct comparisons. Yet, they found the electrolyte to have the largest contribution (30%) to the energy use of the battery materials, while we obtain ca. 49% of the base scenario energy use. The studies are thus in agreement with respect to the significance of the electrolytes.

3.2. Climate change

The base scenario climate change is approximately 230 kg CO₂ eq kWh⁻¹ (Fig. 2b). Cell production dominates (72%), while LiTFSI and lithium foil production contribute notably as well (11% and 9%, respectively). A 55% reduction is achieved in Scenario 1 by reducing electricity use for the cell production. Since the carbon material (CMK-3) has a minor contribution, switching to other carbon materials in Scenarios 2 and 3, respectively, has only minor, if any, positive influence. In Scenario 4, the move from coal to solar power and from natural gas to biogas reduces, in total, the climate change by 72% compared to Scenario 3. The heat source-related reduction is notably smaller (2%), while the reduction due to the shift in electricity source is the major contributor (70%), especially in the electricity-demanding and dominating cell production and lithium foil production processes. In Scenario 5, the 67% specific energy improvement again leads to a 40% down-scaling for all processes. In total, a remarkable 93% reduction of the cell's climate change is achieved given all the improvements.

In their cradle-to-gate study of LIBs, Kim, et al. [9] obtained 63 kg CO₂ eq kWh⁻¹ for cell manufacturing and 28 kg CO₂ eq kWh⁻¹ for the cell materials, i.e. 91 kg CO₂ eq kWh⁻¹ in total. This is similar to Scenarios 1–3 (Fig. 2b), where electricity use corresponding to industrial-scale cell production was employed, and could thus reflect the fact that LIB cells are already produced at industrial scale. Kim et al. [9] also reviewed other LCA studies on LIB cell production, finding a range of 35–460 kg CO₂ eq kWh⁻¹. The reasons for these large differences between different LCA studies are e.g. differences in reported greenhouse gas emissions from cell production and different assumptions on material input [9] and is mirrored in our study; 17–230 kg CO₂ eq kWh⁻¹ given different scenarios. Our best case of 17 kg CO₂ eq kWh⁻¹ (Scenario 5) is of similar magnitude as the best case reported for LIBs: 35 kg CO₂ eq kWh⁻¹.

3.3. Lithium use

Lithium is extracted for use in lithium foil, LiTFSI and LiNO₃. In the base scenario, the lithium use is 0.55 kg kWh⁻¹, whereof 95% is due to lithium foil production, 4% due to LiTFSI production, and 1% due to LiNO₃ production. Only Scenario 5 affects the lithium use, resulting in 0.33 kg kWh⁻¹, as the aforementioned 40% decrease applies here as well. Current LIBs have lithium requirements of about 0.2 kg kWh⁻¹ on battery level [31], indicating that the lithium requirements of LIBs and Li/S batteries are of similar magnitude.

3.4. Influence of allocation approach and price fluctuations

Mass allocation results in approximately the same energy use and climate change for the base scenario as when using economic allocation (580 kWh_{el} kWh⁻¹ and 210 kg CO₂ eq kWh⁻¹, respectively; compare to Fig. 2). The contributions of different processes do not change much, except for a reduced share from the lithium foil production since producing lithium metal from LiCl results in 5 kg of Cl₂ gas as by-product for each 1 kg of lithium metal. Given the much higher price of lithium metal (€200 kg⁻¹) vs. Cl₂ (€0.2 kg⁻¹), a larger share of impact in economic allocation is logical. Applying no allocation for the base scenario results in a notably higher energy use but a similar climate change: 1000 kWh_{el} kWh⁻¹ and 240 kg CO₂ eq kWh⁻¹, respectively. Energy use is affected more since climate change is dominated by electricity use in the cell production, where no by-products are

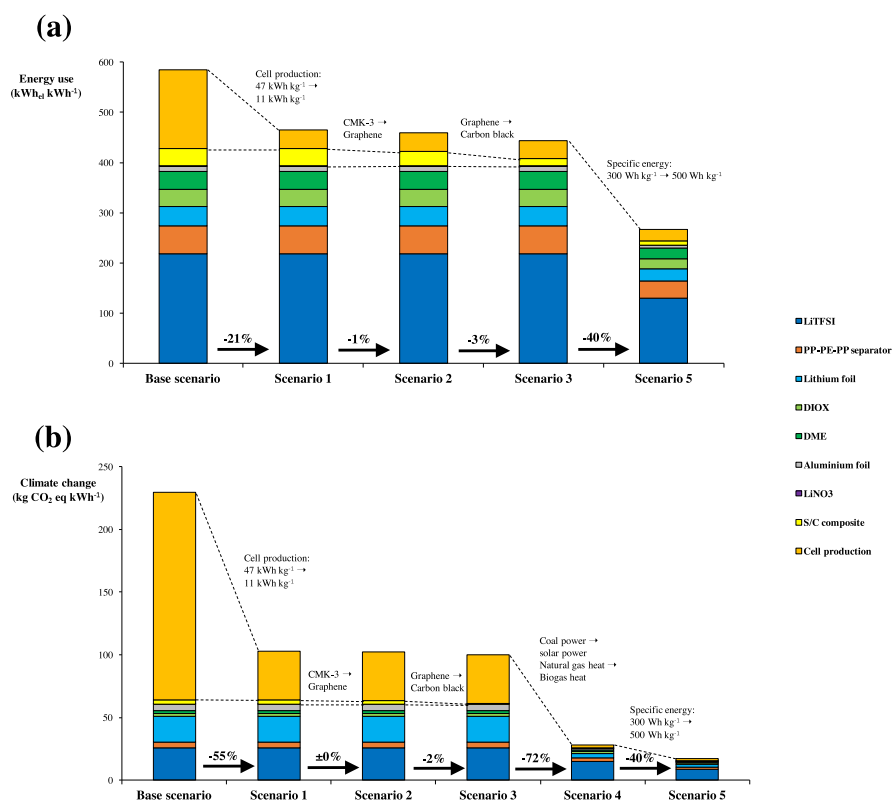


Fig. 2. Energy use (a) and climate change (b) of the Li/S cell production, showing cumulative improvements with bars and marginal step-wise improvements in percent. Scenario 4 has no influence in energy use results and is therefore not shown in (a).

produced.

Increasing prices on at a time by a factor of 10 for the economic allocation resulted only in modest variations in the results: a range of 580–600 kWh_{el} kWh⁻¹ for energy use, whereas climate change was effectively unaltered at 230 kg CO₂ eq kWh⁻¹. There might be additional price correlations not accounted for here, but overall price fluctuations seem to be of minor significance to the calculated environmental impact.

4. Concluding remarks

From our base case and the applied scenarios, we conclude that there are several ways to improve the environmental performance of a Li/S cell in terms of energy use and climate impact: (i) reducing electricity consumption of cell production, (ii) sourcing renewable electricity such as solar or wind power, (iii) improving the specific energy of the Li/S cell and (iv) shifting to low-impact carbon materials. Some of these improvements are possible to conduct without any further research, such as sourcing renewable electricity. Others will require additional research and development, such as reducing electricity use in cell production and achieving higher specific energy. An inherent weakness of our study is that Scenarios 2 and 3 do not couple directly to Scenario 5, while in reality the efforts in altering the C/S cathode, and the type of carbon material, are primarily driven by targets of higher specific (and volumetric) energy.

There are two processes in particular that warrant further investigation regarding data quality due to their large contributions to impacts (Fig. 2): cell production and LiTFSI production. Although the cell production modelled by Deng et al. [14] is detailed, it reflects only one specific pilot-production facility and estimated industrial-scale data. First-hand, or additional estimates of, industrial-scale data would shed further light on the environmental impacts of this process. The estimation of inventory data for LiTFSI by Deng et al. [14] was based on

generic data for specialty chemicals rather than the exact reaction scheme used, and is therefore also in need of further investigations.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jpowsour.2018.02.054>.

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