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Comparative life cycle assessment of high performance lithium-sulfur battery cathodes

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Lithium-sulfur (Li–S) batteries present a great potential to displace current energy storage chemistries thanks to their energy density that goes far beyond conventional batteries. To promote the development of greener Li–S batteries, closing the existing gap between the quantification of the potential environmental impacts associated with Li–S cathodes and their performance is required. Herein we show a comparative analysis of the life cycle environmental impacts of five Li–S battery cathodes with high sulfur loadings (1.5–15 mg cm–2) through life cycle assessment (LCA) methodology and cradle-to-gate boundary. Depending on the selected battery, the environmental impact can be reduced by a factor up to 5. LCA results from Li–S batteries are compared with the conventional lithium ion battery from Ecoinvent 3.6 database, showing a decreased environmental impact per kWh of storage capacity. A predominant role of the electrolyte on the environmental burdens associated with the use of Li–S batteries was also found. Sensitivity analysis shows that the specific impacts can be reduced by up to 70% by limiting the amount of used electrolyte. Overall, this manuscript emphasizes the potential of Li–S technology to develop environmentally benign batteries aimed at replacing existing energy storage systems.

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1. Introduction

Rechargeable batteries are key enablers to transit from the current fossil-fuel based transportation model to a sustainable and efficient renewable energy-based zero emissions transportation system (Knobloch et al., 2020). Rechargeable batteries have already triggered a notorious revolution in portable electronics field (Liang et al., 2019). Since they were firstly commercialized in 1991 by Sony, secondary lithium-ion batteries (LIBs) have dominated the rechargeable battery market (Choi et al., 2012; Goodenough and Park, 2013). However, LIBs are approaching their theoretical limit with an energy density that hardly goes beyond 250 Wh kg–1. This energy density is insufficient to boost the growing demand for electrified transportation (for example in plug-in battery electric vehicles, EVs) and represents a limitation towards clean stationary energy storage systems (Abraham, 2015). EVs can result in further positive impacts as they can be charged from home by simply plugging in (in comparison with the fueling of conventional vehicles), avoiding the risks associated with petroleum supply chain disruption or the exposure to viruses such as Covid-19 (Horsley, 2020). A priori, the use of EVs fed with renewable electricity may reduce greenhouse gas emissions in comparison with conventional vehicles. For instance, a direct 14% of emission reduction is achieved (in a cradle-to-grave analysis) from a conventional EURO-5 1.200 kg petrol car (265.71 kgCO2 eq per 100 km) having an EV with the current electric generation mix in France. When an EV is fed with solar photovoltaic electricity, a 64% reduction from conventional car emissions has been detected (Table S1; Moreno Ruiz et al., 2019).

The cathode is recognized as the most critical part of the battery determining its performance (Eftekhari and Kim, 2017; Liu et al., 2016). As a consequence, great academia and industry efforts are being focused towards the design and fabrication of battery cathodes which effectively store lithium ions. The ability to store lithium using naturally abundant elemental sulfur cathodes is larger in comparison with
traditional LIB cathodes, which mostly rely on the use of lithium cobalt oxide (LiCoO2) (Zhao et al., 2020), lithium manganese oxide (LiMn2O4) (Cusenza et al., 2019), lithium iron phosphate (LiFePO4) (Hänsel et al., 2019), or lithium-nickel-manganese-cobalt-oxide (NMC) (He et al., 2020), together with carbon black and a polymeric binder as cathode materials. As a result, lithium-sulfur (Li–S) batteries present a theoretical specific capacity of 1.672 m Ah g–1 (Song et al., 2013), making them attractive to displace LIBs. Paired with a metallic Li as the anode, Li–S battery enables a remarkable theoretical specific energy density in a full cell of 2.600 Wh kg–1, markedly higher than the <250 Wh kg–1 provided by conventional LIBs (Xu et al., 2014). Moreover, sulfur is a readily available element in comparison with the elements needed in common LIB cathodes. For instance, sulfur accounts for 0.042% of the Earth’s crust, while cobalt is only present by 0.003%. Sulfur is considered a more environmentally benign than the heavy metals used in LIBs, and Li–S batteries present fewer safety concerns in comparison with LIBs (Manthiram et al., 2019).

However, the development of Li–S batteries faces many challenges which in turn lower their energy density and cycling life. Firstly, the electronically insulating character of sulfur limits its full utilization as active material. Therefore, early attempts were focused to mix sulfur with carbonaceous conducting fillers. Unlike conventional insertion cathode materials, sulfur suffers structural and compositional changes upon cycling, yielding soluble lithium polysulfides (Li2Sx, 4 ≤ x ≤ 8) which contaminate the battery and lower the battery stability and performance (Manthiram et al., 2013). To mitigate this shuttle mechanism and increase cycle stability and system efficiency, different cathode configurations, separator formulations and electrolyte chemistries have been explored. The volumetric change during continuous cycling and unstable solid-electrolyte interphase (SEI) are other issues contributing to capacity fade. Since these problems are exacerbated at high sulfur loadings (being the 5–10 mg cm–2 range generally considered as a high sulfur loading), most of the published articles have been limited to modest mass loadings of ~2 mg cm–2 (Eftekhari and Kim, 2017). This poor mass loading constitutes a serious bottleneck towards the translation of obtained fundamental designs into practical applications. As a consequence, no Li–S batteries have been commercialized to date (Zhu et al., 2019). Such drawbacks mostly lie on the insulating nature of sulfur (S8) and lithium (di)sulfides (Li2S and Li2S2) (Liu et al., 2017), the large volumetric change of sulfur-electrodes during charge/discharge (Barai et al., 2016), and the lithium polysulfide (LiPS) shuttling effect between electrodes that leads to low coulombic efficiencies (Ali and Yan, 2020).

Several manuscripts have successfully addressed the obstacles faced by Li–S cathodes through different and often complex chemical approaches (Shaibani et al., 2020; Chong et al., 2018; Li et al., 2020; Huang et al., 2020). Additionally, scarce and toxic raw materials with potential supply chain bottleneck such as lithium, cobalt, vanadium or nickel are generally used in batteries, which increase their environmental impact (Goodenough and Park, 2013; Olivetti et al., 2017). These studies failed to report the energy and environmental burdens associated with their synthetic approach, which is essential towards up-scaling. Consequently, a careful assessment of the environmental impacts associated with the fabrication of Li–S cathodes should be carried out to get a complete picture aimed at reducing the pressure on natural resources and ecosystems. Life cycle assessment (LCA) is the most widely used methodology to “evaluate the environmental impact of a product throughout its life cycle encompassing extraction and processing of the raw materials, manufacturing, distribution, use, recycling, and final disposal” (Ilgin and Gupta, 2010; Muralikrishna and Manickam, 2017). To date, LCA has been extensively used to quantify the environmental impacts of products/processes as varied as bio-based and fossil-based plastics (Walker and Rothman, 2020), CO2 capture and conversion (Jens et al., 2019), batteries (Wang et al., 2020), algae cultivation (Bessette et al., 2020) or magnets (Marx et al., 2018). Accordingly, here we performed the LCA of five recently reported high-performance Li–S battery cathodes with high sulfur loadings to provide cues towards the development of novel energy storage alternatives with reduced environmental impact.

The five cathodes analyzed in this research are listed hereafter. Shaibani et al. faced the quick capacity fade of Li–S batteries resulting from the cathode fragmentation by introducing a small amount of sodium carboxymethylcellulose (NaCMC) binder through a simple fabrication process (cathode termed as NaCMC-sulfur) (Shaibani et al., 2020). NaCMC acted as a glue, forming stiff links between neighboring particles but without covering them (no interference with electrochemical reactions and ion mobility was achieved), obtaining a cathode with a sulfur loading of 15 mg cm–2 able to deliver >1200 m Ah g–1. This approach lies on the use of the environmentally friendly and non-toxic NaCMC binder, in comparison with traditional petroleum-derived polyvinylidene difluoride (PVDF) which entails serious environmental burdens (Chong et al., 2018). A cathode consisting on graphene, carbon nanotubes (CNTs) and cobalt (Co) nanoparticles with a 3D structure was obtained via a one-pot pyrolysis process (cathode termed as graphene-CNT-Co-sulfur) (Shaibani et al., 2017). Such cathode could trap lithium polysulfides (thanks to the cobalt nanoparticles) while favorably interacting with electrolyte (because of the highly porous structure), resulting in a capacity fade of 0.05% after 500 cycles at 1C rate (a current that is able to discharge the fully charged battery in 1 h, 1.675 mA h g–1 in this case), Tan et al. designed a composite material consisting on crystalline Li2S nanoparticles wrapped by graphene to obtain a cathode with fast electronic/ionic transport and low volumetric changes (cathode termed as LiS-graphene) (Tan et al., 2017). The group headed by Nazar faced the poor sulfur conductivity and polysulfide shuttling using a lightweight and high-surface area MgB2 metallic cathode (termed as MgB2-sulfur) (Pang et al., 2019). A stable cycling at a sulfur loading of 9.3 mg cm–2 was achieved using a low electrolyte volume. The same group also prepared an electrically conducting CoS8 material interconnected with graphene (sulfur loading of 4.5 mg cm–2) to obtain a Li–S cathode effectively suppressing LiPS shuttling, leading to a capacity fade <0.045% per cycle over 1500 cycles at 2C, which is a 10-fold improvement in comparison with standard porous carbons (cathode termed as CoS8-sulfur) (Pang et al., 2016). Interestingly, CoS8 was synthesized using a fast and scalable microwave solvothermal method, reducing the fabrication impacts in regard with conventional techniques.

By performing the LCA of five high performance Li–S battery cathodes having high sulfur loadings in the range of 1.5–15 mg cm–2, this manuscript closes the existing gap between the quantification of the potential environmental impacts associated with Li–S cathodes and their performance. As the cathode not only determines the associated environmental impact but also the electrochemical performance (delivered capacity, capacity fade, life cycle), obtained results are analyzed within the framework of battery performance.

2. Method

2.1. Goal, scope and inventory analysis

The goal of this research is to compare the associated environmental impacts during the manufacturing process of 5 Li–S batteries. Those Li–S batteries were selected because they rely on different strategies to achieve high sulfur loadings which afford large energy densities: a paramount aspect towards practical implementation (Hu et al., 2020). Moreover, all the selected batteries present acceptable cycle stability which reduces their associated environmental burdens (Peters et al., 2016).
Eleven impact categories (termed as impact indicators from here on) within CML-baseline method have been studied (Acero et al., 2016; Pradhan et al., 2019). The CML-baseline standardized categories are as follows: Ozone Layer Depletion Potential (ODP), Abiotic Depletion, Fresh Water Aquatic Ecotoxicity (FWAET), Photochemical Oxidation, Terrestrial Acidification Potential (TAP), Marine aquatic ecotoxicity (MAETP), Fossil Fuels Abiotic depletion (FDP), Global Warming Potential (GWP), Terrestrial ecotoxicity (TETP), Human Toxicity Potential (HTP) and Eutrophication. The eleven baseline indicators provided by the CML methodology were created by the University of Leiden in 2001 and offer a common ground to compare the different impact categories used in LCA (Acero et al., 2016). All the analyzed indicators are midpoint (directly linked to chemical compounds), presenting a lower uncertainty level in comparison with endpoint indicators (Bare et al., 2000) as they are not linked to cause-effect framework, such as the impacts regarding human health (measured as Disability Adjusted Life Years, DALY) or biodiversity lose (species disappeared per year). Special attention has been paid to the impact in Global Warming Potential (GWP) measured in kg CO₂-eq because it provides a mean for an easy comparison with the environmental impacts associated with other energy storage systems (Jen et al., 2020). EV fabrication doubles the GWP in comparison with combustion engine cars as a result of the environmental burdens associated with battery production. Therefore, we estimate providing the GWP of Li–S batteries results of prime interest. The boundaries of the performed analysis are reflected in Fig. 1.

Table 1 shows the materials and energy inputs for inventory of the five Li–S batteries. The composition of each cathode has been explained in the Introduction section, and is based on a mixture of sulfur (the active part), carbon (to enhance the poor electronic conductivity of sulfur), a binder which holds together the cathode components and a solvent to allow its processing. Regarding the remaining components, all the batteries are very similar as they comprise metallic lithium as the anode (common in all of them) and use a 25 μm thick Celgard® 2400 monolayer polypropylene membrane with a porosity of 41% as the separator. For the liquid electrolyte, lithium nitrate (LiNO₃) salt is mixed with a lithium bis(trifluoromethanesulfonylimidate (LiTFSI) salt and incorporated into a 50:50 vol mixture of dimethoxyethane (DME) and dioxolane (DOL) liquid electrolyte. Such combination of lithium salts (LiNO₃ and LiTFSI) and aprotic solvents (DME and DOL) yields a protective layer on the surface of the lithium anode which spans Li–S battery life cycle, limiting the self-discharge from 10% day⁻¹ to 2% month⁻¹ (Wang et al., 2016; Zhang et al., 2015). Cell container, module packaging, cooling system and the pack packaging are similar for all the batteries, and their amount differences arise from the different size of the final batteries. Although all cathode current collectors consist of aluminum ingot, their quantity differs substantially between cathodes due to differences on their sulfur mass loading (the greater the mass loadings, the smaller the cell and the lower the amount of required aluminum).

The model includes the use of a glove-box filled with argon to provide a moisture free inert atmosphere (H₂O and O₂ < 1 ppm) during Li–S cell assembly process. Furthermore, the impacts of metal and chemical factories have been included in the assembly process of the cell, using “item” as unit. Finally, the consumed electricity during battery production has been calculated according to the batteries weight. Taking as reference (Deng et al., 2017), the value of 17,204.32 Wh kg⁻¹ has been used in this study.

2.2. Comparison between Li–S with lithium ion and sodium ion batteries

This research uses OpenLCA software (Kadhum et al., 2018), and EcoInvent 3.6 database (released on September 12, 2019) (Moreno Ruiz et al., 2019; Bora et al., 2020), to model the impacts of five different Li–S batteries with potential application in the transportation field. EcoInvent database provides a reference compact electric passenger car of 920 kg without the battery and a rechargeable prismatic LIB of 262 kg. “LIB reference case” from now on. The average life time of the car has been parameterized to be 150,000 km, while the life expectancy of the battery is set at 100,000 km. The battery pack contains 14 single cells and provides an electric supply of 29.87 kWh with 48 V (the life cycle inventory of this battery has been included in Table S3). The energy density of the battery is 114 Wh kg⁻¹ with a 120 km driving range. It has been previously estimated that the battery causes 15% of the impacts of the total car (Notter et al., 2010). The reference battery dataset includes the production of 14 single cells, a battery management system, a steel box, and cables. Furthermore, in order to perform this analysis regarding LIB reference case, has been followed EcoInvent inventory: the cathode (LiMn₂O₄), separator (polyethylene floss processed with liquid acetone, hexafluoroethane, phthalic anhydride, PVDF and silica sand), anode (graphite) and the electrolyte (LiPF₆ in ethylene carbonate) (Moreno Ruiz et al., 2019) (Table S3). The LIB reference case has been originally modeled following a cradle-to-grave approach, thus the data sheet has been modeled into cradle-to-gate for the sake of comparison. Table S4 provides the differences in impacts between both analyses, where an increase of 18% of CO₂-eq from gate to grave impacts and an increase of 38% in ozone layer depletion impact are observed.

Obtained results have also been compared with the environmental impacts arising from a sodium ion battery (NIB) comprising a layered transition metal oxide in combination with a PVD binder as a cathode and hard carbon with NaCMC as an anode (the most extensively studied combination in NIBs) (Peters et al., 2016). An aluminum current collector and a sodium hexafluorophosphate (NaPF₆) salt in an organic solvent together with a polyethylene/polypropylene membrane is used as electrolyte/separator pair. The energy density of the battery is 128 Wh kg⁻¹. Similarly to this work, the impacts for the NIB have been obtained using a cradle-to-gate perspective, making the comparison meaningful.

Our battery packs have been modeled assuming that each cell has to generate 60 A h, using the approximate reference of Audi e-tron (Audi, 2020) and following the models reproduced by Deng et al. (2017). The cells are able to perform about 132–144 Wh, and the modeled battery packs contains 432 cells (Table S1) with a power capacity of 57 kWh − 62 kWh.

2.3. Sensitivity analysis

In batteries having an electrolyte/sulfur (E/S) ratio above 10 mL g⁻¹, the electrolyte accounts for more than the 50% of the weight of the whole cell. An excessive amount of electrolyte may thus increase the whole battery weight (and volume), reducing the energy density. Therefore, several works have highlighted that lowering the E/S ratio to values below 5 results a boundary condition to obtain high-performance Li–S batteries (Zhao et al., 2020). Accordingly, the environmental impacts of the analyzed Li–S batteries were calculated again lowering the E/S ratio to 4.

2.4. Life cycle interpretation

LCA analyses have been carried out with OpenLCA software, and the EcoInvent 3.6 Database (Moreno Ruiz et al., 2019). Our research compares the production of five Li–S batteries within a cradle-to-gate perspective. All the five batteries have been modeled as full battery packs, but the analysis of the Battery Management Electronic System (BMS) has been maintained out of scope of the study. This choice has been made on the basis that its incorporation may
obstruct a proper comparison of the environmental analysis of Li–S batteries (BMS is not itself an energy storage system). Regarding the Functional Unit (FU), to compare the results between the analyzed Li–S batteries, reference LIB and NIB, all the modeled batteries are assumed to perform 100,000 km during their life. This threshold has been settled down with the reference of Ecoinvent database. The boundary system has been defined as cradle-to-gate in all the batteries, due to the current lack of data when defining the recycling processes of Li–S and NIB batteries (Peters et al., 2016). Furthermore, to standardize different storage capacity batteries (from 29.9 to 62.0 kWh), a FU of 1 kWh of storage capacity has been used. This standardization allows us to compare environmental impacts of Li–S, LIB and NIB batteries. When defining the system boundary, it must be specified that the contribution of the battery to the global emissions of the selected electric car ranges from 9 to 21% (Table S1) depending on the fed (national electric mix, fully solar photovoltaic electricity). The 9–21% emissions corresponding to the battery could be identified in their totality (100%) by a cradle-to-grave analysis (Table S4). Instead, cradle-to-gate approach allows detecting nearly ~82% of the eqCO₂ emitted by the battery.
during its life (Table S4), and 62–99% in the rest of the eleven impact categories.

3. Results and discussion

3.1. Environmental impact of Li–S batteries

The environmental impacts of Li–S batteries per kWh of storage capacity are displayed in Fig. 2. Results are shown in 11 impact indicators extracted of the life cycle impact assessment (LCIA) for all the studied five Li–S batteries, which can be easily distinguished in different colors. At first sight, the NaCMC-sulfur battery having a cathode composed by 70% colloidal sulfur, 20% activated carbon and 10% NaCMC shows the lower environmental impact in all the studied categories, while the graphene-CNT-Co-sulfur battery comprising a cathode based on graphene nanosheets, CNTs, Co nanoparticles, sulfur, super P carbon and PVDF presents the largest environmental burdens. The second battery with lower environmental impact in the analyzed impact indicators is the MgB2-sulfur cathode comprising the metallic MgB2 (Pang et al., 2019). In this battery, a lightweight magnesium diboride (density of 2.57 g cm$^{-3}$) is used as the host for sulfur to reach high areal sulfur loadings with a low electrolyte amount, allowing the implementation of lighter batteries and thus reducing its associated environmental impacts. These differences are larger in impact indicators such as ODP, acidification potential (AP) and GWP, where increases up to a factor of 4.5 are observed. It should be taken into account that typically Li–S cathodes are designed to provide an enhanced performance in terms of energy density, so strategies which trap lithium polysulfides are commonly pursued. This performance optimization is carried out at expenses of process simplicity and resource/energy efficiency, so little attention is paid on green chemistry principles.

We explain this enlarged environmental impact in terms complex multicomponent character and multiple synthetic approaches followed to obtain the cathode, which include calcinations at 900 °C under argon atmosphere and the use of strong hydrochloric acid. These results highlight the pivotal role of walking away from cathodes comprising many components which are synthesized

| Table 1 | Material and energy input inventory for all the studied Li–S cathodes for a standardized 60 kWh EV battery pack of 100,000 km life as Functional Unit (depending the used technology the capacity varies from 57 to 62 kWh, Table S2). NaCMC: sodium carboxymethylcellulose; PVDF: polyvinylidene fluoride; DMF: dimethylformamide; NMP: N-Methyl-2-pyrrolidone; LiTFSI: lithium bis(trifluoromethanesulfonylimide); DOL: dioctane; DME: dimethoxyethane; LiNO3: lithium nitrate; PP: polypropylene; PE: polyethylene; ABS: acrylonitrile butadiene styrene. |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Cathode** | **Cathode** | **Cathode** | **Cathode** | **Cathode** | **Cathode** | **Cathode** | **Cathode** |
| NaCMC-sulfur | Graphene-CNT-Co-sulfur | LiS-graphene | MgB2-sulfur | Code-sulfur |
| Ch (kg) | Ch (kg) | Ch (kg) | Ch (kg) | Ch (kg) | Ch (kg) |
| Colloidal sulfur | Graphene-Sulfur | LiS/Li-graphene | Sulfur | Graphene-MgB2 host | Sulfur |
| 21.60 | 24.69 | 22.34 | 20.41 | 12.85 | 22.94 |
| Carbon black | Carbon | Binder [PVDF] | Binder (PVDF) | Binder (PVDF) | Binder (PVDF) |
| 6.17 | 7.05 | 9.07 | 1.24 | 2.72 | 5.73 |
| Binder [NaCMC] | | Solvent [PVDF] | Solvent [PVDF] | Solvent (DMF) | Solvent (DMF) |
| 3.09 | | 2.30 | 1.24 | 3.36 | 2.87 |
| Aluminum ingot | Aluminum ingot | Aluminum ingot | Aluminum ingot | Aluminum ingot | Aluminum ingot |
| 2.60 | 26.66 | 2.69 | 3.98 | 8.26 |
| Anode | Anode | Anode | Anode | Anode |
| Lithium billet | Lithium billet | Lithium billet | Lithium billet | Lithium billet |
| 7.04 | 7.04 | 7.04 | 7.04 | 7.04 |
| Electrolyte | Electrolyte | Electrolyte | Electrolyte | Electrolyte |
| LiTFSI | LiTFSI | LiTFSI | LiTFSI | LiTFSI |
| 59.05 | 122.17 | 90.85 | 156.76 |
| DOL | DOL | DOL | DOL | DOL |
| 13.59 | 28.12 | 20.87 | 36.68 |
| DME | 107.56 | 51.52 | 38.33 | 66.63 |
| LiNO3 | 5.55 | 4.22 | 3.00 | 5.30 |
| Separator | Separator | Separator | Separator | Separator |
| PP | PP | PP | PP | PP |
| 1.96 | 15.81 | 1.96 | 2.69 | 6.03 |
| Cell container & Assembly | Cell container & Assembly | Cell container & Assembly | Cell container & Assembly | Cell container & Assembly |
| Aluminum ingot | Aluminum ingot | Aluminum ingot | Aluminum ingot | Aluminum ingot |
| 5.98 | 5.98 | 7.90 | 18.15 |
| PP | PP | PP | PP | PP |
| 1.83 | 1.83 | 2.41 | 5.60 |
| PC | PC | PC | PC | PC |
| 1.19 | 1.19 | 1.19 | 1.19 |
| Argon | Argon | Argon | Argon | Argon |
| 0.64 | 0.64 | 0.64 | 0.64 |
| Chemical Factory (units) | Chemical Factory (units) | Chemical Factory (units) | Chemical Factory (units) | Chemical Factory (units) |
| 87,994 | 1.37 | 3,387 | 3,387 |
| Metal Factory | Metal Factory | Metal Factory | Metal Factory | Metal Factory |
| 3,132 | 1,837 | 3,813 | 3,813 |
| (units) | (units) | (units) | (units) | (units) |
| Module packaging | Module packaging | Module packaging | Module packaging | Module packaging |
through energy-intensive multistage synthetic procedures. Moreover, for the binder choice, the use of renewable materials such as those derived from cellulose is preferred against petroleum-based PVDF. Therefore, in order to lessen the environmental impact associated with Li$_2$S battery production, simple one-pot batch syntheses and the use of renewable resources are recommended.

With the aim of providing cues to reduce the environmental impacts during Li$_2$S battery design, as depicted in Fig. 3, the distribution of GWP for each battery component has been calculated (the impact has been normalized so in all the cases the GWP is 100%). The electrolyte presents a dominant contribution to the GWP for all the five batteries studied (from 35% for MgB$_2$-sulfur battery to 47% for Li$_2$S-graphene battery). This marked contribution arises from the large amount of electrolyte used in those batteries, which rely on an E/S ratio of 4–15 mL g$^{-1}$, representing a weight contribution of 73±6% to the total battery cell. The electricity required for material processing and cell-pack manufacturing is also a relevant driver for the GWP as it accounts for the 20–25% of the whole impact in this category (arising from the large share of non-renewable energy in the European electricity mix). Pack packaging and cell container and assembly also present an important role as they are based on aluminum cell containers, providing 8–10% of the total impact. On average, the cathode only accounts for 2.3% of the total GWP, which is considerably lower than the contribution of 20% reported for NIBs (Peters et al., 2016).

### 3.2. Comparison with lithium ion and sodium ion batteries

To shed more light on the potential of Li$_2$S batteries for more sustainable energy storage applications, obtained LCA results are contrasted with the LIB reference case using the same 1 kWh of storage capacity and 100,000 km battery life as a FU (see Table 2). Environmental impacts are also compared with NIBs because they are foreseen as a sustainable and cost-competitive alternative to lithium batteries. Sodium is abundant (2.36 wt% of Earth’s crust), low cost and presents a redox potential of −2.71 V vs. E°, which yields NIBs good energy storage ability (translated into energy densities of 90–120 Wh kg$^{-1}$) (Casas et al., 2020). As a consequence, NIBs are useful for large-scale energy storage systems where weight requirements are not severe.

Studied Li$_2$S batteries present an improved GWP of 53–247 kg CO$_2$-eq (mean value 127) in comparison with the 140 kg CO$_2$-eq of a NIB comprising a layered oxide cathode and an anode composed by a styrene-butadiene rubber, NaCMC binder and hard carbon (with an electrolyte based on a porous polyethylene/polypropylene soaked into an organic solvent having sodium hexafluorophosphate salt) (Peters et al., 2016), and 57 kg CO$_2$-eq of the LIB reference case (Moreno Ruiz et al., 2019). From the other side, the human toxicity potential (HTP) of Li$_2$S batteries lies within the range of 40–180 kg 1,4-DB-eq (mean value 92), which equals the results obtained for NIBs (168.15 kg 1,4-DB-eq) in the worst-case scenario (battery cell 2) and remains well-below the 1326.5 kg 1,4-DB-eq of the LIB reference case. Concerning the TAP, impacts of 40–185 kg SO$_2$-eq 10$^{-2}$ (mean value 96) are achieved in contrast to the 151 kg SO$_2$-eq 10$^{-2}$ of NIBs or the 109 kg SO$_2$-eq 10$^{-2}$ of the LIB reference case.

Overall, it is seen that Li$_2$S batteries have the potential to outperform or at least match both LIBs and NIBs for most of the environmental impacts (1 kWh of storage capacity as FU). These results can be explained as follows. The amount of hard carbon used in Li$_2$S batteries is lower than that used in LIBs and NIBs. Typically, petroleum coke or sugars are used as precursors for carbonaceous materials, increasing the overall environmental impact due to the use of non-renewable petroleum or the need for sugarcane/sugar beet for sugar extraction, which enhances water consumption and eutrophication (Chauban et al., 2011). Moreover, up to 11 kg of sugar are required to obtain 1 kg of hard carbon (Peters et al., 2019), where the energy-demanding (electricity and heat) carbonization process and the nitrogen required for maintaining an inert atmosphere...
during production contribute to environmental impacts. Additionally, cathode active materials for LiBs comprise cobalt and lithium (lithium cobalt oxide LiCoO₂, lithium manganese oxide LiMn₂O₄, lithium iron phosphate LiFePO₄) (Manthiram, 2020), while NiBs require nickel hexacyanoferrates (N₉[Fe(CN)₆]₃) or sodium vanadium phosphates (Na₃V₂(PO₄)₃) (Shen et al., 2020; Zhang et al., 2019). Contrarily, the use of heavy metals such as cobalt, manganese or nickel in Li–S batteries is not required, where cathodes use sulfur quantities as large as 70% by weight. This also leads to lower fresh water aquatic ecotoxicity and human toxicity potentials associated with Li–S batteries because LiBs rely on the use of toxic cathodes (Chen et al., 2019). It should be also taken into account that usually, Li–S cathodes are obtained after the introduction of sulfur into a carbonaceous structure by efficient thermal infusion (Pang et al., 2019), avoiding complex, expensive and environmentally harmful synthetic procedures typically of layered oxides used for LiBs and NiBs. As a substantial reduction on the use of toxic materials in achieved using Li–S batteries in comparison with LIBs (Ellingsen et al., 2014), Li–S batteries may represent a sustainable energy storage alternative. However, Li–S batteries also display several drawbacks such as larger ozone layer depletion potential in comparison with the LIB reference case. This could be ascribed to the fact that at cold temperatures (in the atmosphere) sulfur provides a reacting medium for chlorine gases to destroy ozone, where one chlorine atom has the potential of destroying over 100,000 ozone molecules (Varotsos, 1997).

In comparison with the petroleum-based PVDF which is the dominating binder in LIBs and NiBs (accounting for nearly 20% of the cathode and notably contributing to greenhouse gas emissions) (Ludwig et al., 2016), Li–S batteries can use cellulose-derived binders, leading to favorable results. Additionally, the use of aluminum as cathode current collector instead of copper enables the reduction of freshwater eutrophication potential (FEP) and HTP results while reducing the metal depletion caused by copper mining (aluminum is 1200 times more abundant than copper on Earth’s crust). These results are in line with the investigations of Ellingsen et al., who reported that copper current collectors have a contribution above 60% in the impact indicators of freshwater ecotoxicity, marine ecotoxicity, terrestrial ecotoxicity and human toxicity (Ellingsen et al., 2014).

3.3. Sensitivity analysis

To achieve good electrochemical sulfur utilization and obtain high-performance Li–S batteries, most of the papers report the use of large electrolyte volumes, typically expressed as E/S ratios of 1.5–15 mL g⁻¹ (Hagen et al., 2014) (see Fig. 1). However, the electrolyte contains a high passive weight, which increases the environmental burdens of the battery with no positive electrochemical contribution. Based on this assumption and according to our results showing that most of the environmental impact of Li–S batteries arises from the electrolyte use, a sensitivity analysis has been carried out to quantify the influence of the amount of liquid electrolyte (Yue et al., 2012). In this regard, the cathodes have been standardized with a E/S value of 4, assuming that long-cycling can still be maintained ensuring a proper wetting of electrodes (Zhu et al., 2019) and assuming the future predictions for the electrochemical stability (Manthiram et al., 2015). The effect of the electrolyte reduction on the 11 impact indicators in a cradle-to-gate LCA for the studied five Li–S batteries is shown in Fig. 4, where the vertical axis accounts for the relative reduction on each impact indicator. It is clearly observed that an E/S reduction up to 4 dramatically reduces the environmental impact of Li–S batteries (see Fig. S1, Fig. S2 and Table S5 for the variations in individual batteries). When compared, the life cycle impacts of Li–S batteries with an E/S ratio of 4 are 11–71% lower than those using the large electrolyte volumes reported in the analyzed batteries (Table S5). Especially relevant are the improvements obtained in ozone layer depletion, abiotic depletion, photochemical oxidation, acidification and human toxicity, where reductions over 40% are achieved.

This is ascribed to several factors. From one side, lithium is a relatively scarce element found in nature at low concentrations, so its mining requires large quantity of soil processing. Therefore, reducing the amount of lithium salts within the liquid electrolyte (LiTFSI and LiNO₃) may lower the waste generation, water contamination and transport issues produced during brine treatment, limiting the impacts related to ozone layer depletion, abiotic depletion or human toxicity (Flexer et al., 2018). From the other side, large amounts of methylchloride solvent are needed for the synthesis of LiTFSI, encompassing a significant ozone depletion potential (Deng et al., 2017). Additionally, dioxolane and dimethoxymethane solvents used as liquid electrolyte present a NFPA 704 (Standard System for the Identification of the Hazards of Materials for Emergency Response) rating of 1-3-2 and 2-2-0, respectively for health, flammability and instability (NFPA 704, 2017), so their use can present associated health issues. Moreover, dioxolane and dimethoxymethane present a vapor pressure of 79 and 48 mm Hg at 20 °C, respectively, meaning that they can be volatilized easily, and their release to soil or water presents a high reactivity based on a high estimated KOC (organic carbon-water partition co-efficient) of 15 and 18, respectively. Finally, during the production of dioxolane and dimethoxymethane ammonia and nitrate are emitted (Deng et al., 2017).

It may be reasonable to assume that lowering the E/S ratio up to 4 may decrease the delivered capacity of batteries, which in turn would decrease the energy density and should bring an increased environmental impact per kWh of storage capacity (as all the other components are not changed). However, we here hypothesize that lowering the E/S ratio up to 4 is not associated with a lower electrochemical performance because we would like to emphasize the effect of electrolyte on the overall environmental impact. Moreover, very few research manuscripts carefully analyze the loss of electrochemical performance upon electrolyte reduction, so obtaining such information results somehow complex.

3.4. Future improvement potential

As demonstrated by the drawbacks that still exist on Li–S batteries, their research is still in its infancy. This can be seen as an advantage since still a lot of room exists for improvement, not only in terms of electrochemical performance but also in terms of sustainability. For example, renewable materials such as cellulose, the most abundant organic compound on Earth, could potentially be used to fabricate highly porous and electrically conducting cathode materials (Nguyen et al., 2019a, 2019b), limiting the use of toxic or scarce elements. Scaling up the manufacturing of Li–S batteries from laboratory-scale to market (into pouch cells for example) should be a priority towards lowering their associated environmental impact. Moreover, very few research manuscripts carefully analyze the loss of electrochemical performance upon electrolyte reduction, so obtaining such information results somehow complex.
is highly stable chemically and thermally, allowing its recovery and providing an additional economic revenue (Elibama, 2014). A further progress in this area may come from the recycling of the anode, as the pure lithium metal used in Li-S batteries (unlike conventional LIBs) can be recovered via automated methods such as Foucault’s current and density separators (Deng et al., 2017).

Recent LCA studies carried out in the field of thermal energy storage have shown that hybridization of different energy storage systems can reduce greenhouse gas emissions and improve reliability and economic viability of energy sources (Rashid, 2019a; Rashid et al., 2019b). Although the maximum benefits from hybridization are achieved at plant-level (Peterseim et al., 2014), hybridization can also be implemented at light-level, allowing its use in EVs powered by Li–S batteries where space limitations exist. The combination of Li–S

![Graph showing environmental impacts of Li–S batteries](image)

**Fig. 2.** Environmental impacts of the selected five Li–S original batteries (with different E/S factors) corresponding to 1 kWh of storage capacity and 300,000 km battery life (FU).

![Diagram showing battery component contribution](image)

**Fig. 3.** Relative contribution of each battery component to the GWP, the comparison respects the FU of the model.
batteries with thermal energy storage should be explored in the near future as a strategy to obtain synergistic benefits towards energy storage systems with lowered environmental impact.

4. Conclusion

A comparative cradle-to-gate life cycle assessment study on the environmental impacts of five Li–S batteries comprising cathodes with high sulfur loadings in the range of 1.5–15 mg cm⁻² has been performed. Life cycle inventories of studied batteries containing component production and battery assembly are disclosed, enabling future comparisons. Environmental impacts are classified into 11 standardized CML-baseline indicators such as ozone depletion potential, global warming potential or acidification potential as they provide more detailed information linked to specific chemical compounds and respective environmental burdens associated with the use of Li–S batteries. Results reveal that the cathode choice and respective changes in the rest of components can lower the environmental impact of the whole Li–S battery by a factor up to 5 (for instance, global warming potential ranges between 53 and 248 kg CO₂-eq per kWh). Interestingly, a cathode composed by 70% colloidal sulfur, 20% activated carbon and 10% NaCMC yielded the lowest environmental impact, indicating the pivotal role of obtaining high sulfur loadings and renewable-based binders. When comparing with conventional lithium-ion batteries, Li–S batteries can present lower environmental impact per kWh of storage capacity. Moreover, although a liquid electrolyte excess is generally seen as beneficial for the electrochemical performance, obtained LCA results clearly indicate a negative role of large electrolyte volumes on the environmental burdens associated with the use of Li–S batteries. Accordingly, a sensitivity analysis in which the amount of electrolyte is lowered was performed, revealing that environmental burdens can be reduced by up to 70% by solely limiting the amount of used electrolyte.

Table 2

Environmental impacts for all the studied Li–S cathodes. Data corresponding to LIB reference battery were extracted from Ecoinvent database, while NIB cell impacts are taken from reference (Peters et al., 2016). In the case of NIB some impact categories cannot be obtained due to the differences on the LCA analysis. In all the batteries FU have been maintained and all the analyses have been performed with cradle-to-gate boundary system.

| Impact category                          | NaCMC-sulfur | Graphene-CNT-Co-sulfur | Li₁₅S-graphene | MgB₂-graphene | Co₂S₈-graphene | Average Li–S | Ecoinvent Data LIB | NIB | Unit      |
|----------------------------------------|-------------|------------------------|---------------|---------------|---------------|--------------|------------------|-----|-----------|
| Ozone Layer Depletion Potential (ODP)   | 66.29       | 293.50                 | 135.97        | 111.75        | 175.97        | 156.70       | 4.29             | 0   | kg CFC11-eq 10⁻⁶ |
| Abiotic Depletion                       | 41.94       | 186.67                 | 83.87         | 70.18         | 114.52        | 99.43        | 5.27             | 0   | kg Sb-eq 10⁻⁷  |
| Fresh Water Aquatic Ecotoxicity (FAET)  | 36.99       | 165.35                 | 66.27         | 67.91         | 84.56         | 84.21        | 829.24           | 0   | kg 1,4-DB-eq  |
| Photochemical Oxidation                 | 26.56       | 135.03                 | 51.24         | 51.99         | 67.22         | 66.41        | 48.42            | 0   | kg C₃H₆-eq 10⁻³ |
| Terrestrial Acidification Potential (TAP)| 40.41       | 184.97                 | 76.04         | 77.88         | 99.35         | 95.73        | 109.32           | 151 | kg SO₂-eq 10⁻² |
| Marine aquatic ecotoxicity (MAETP)      | 10.54       | 48.33                  | 18.62         | 19.14         | 24.86         | 24.30        | 106.44           | 0   | kg 1,4-DB-eq 10⁴ |
| Fossil Fuels Abiotic depletion (FDP)     | 40.14       | 121.30                 | 45.07         | 48.18         | 62.72         | 63.48        | 24.98            | 4.34 | kWh 10⁻²     |
| Global Warming Potential (GWP100a)      | 53.33       | 247.58                 | 99.84         | 107.01        | 129.11        | 127.37       | 57.07            | 140.33 | kg CO₂-eq  |
| Terrestrial ecotoxicity (TETP)          | 15.21       | 67.85                  | 29.07         | 28.07         | 37.15         | 35.47        | 65.76            | 0   | kg 1,4-DB-eq 10⁻² |
| Human Toxicity Potential (HTP)          | 40.29       | 180.37                 | 73.64         | 80.72         | 88.72         | 92.75        | 1326.50          | 168.15 | kg 1,4-DB-eq  |
| Eutrophication                          | 94.60       | 404.24                 | 170.76        | 198.21        | 218.08        | 217.18       | 489.79           | 0   | kg PO₄-eq 10⁻³ |

Fig. 4. Sensitivity analysis of life cycle impacts in each of the 11 impact indicators from using the original E/S ratio (0%) to using a reduced E/S = 4 ratio.
Obtained results are particularly promising taking into account the low technical maturity of Li–S batteries in comparison with LIBs, which leaves room for great improvements regarding environmental performance. To fully understand the environmental impacts of Li–S batteries, cradle-to-grave LCA studies should be carried out as the data becomes available; to increase the capacity of modeling the environmental impacts of the batteries up to 38% according to LIB batteries (depending the impact category). This would shed light on the increased recycling potential of Li–S batteries in comparison to LIBs as the used salts and anodes can be easily recovered through several automated separation methods. Overall, this research would support further follow-up research focusing on the key aspects that need to be considered for low environmental impact Li–S batteries, providing a guide to promote commercialization of greener Li–S batteries.

Credit author contribution statement

Sergio Lopez: Data curation, Formal analysis, Investigation, Methodology, Validation. Ortizzi Akizu-Gardoki: Formal analysis, Investigation, Methodology, Validation, Software, Figures, Writing - original draft, Writing - review & editing. Erlantz Lizundia: Conceptualization, Formal analysis, Investigation, Methodology, Project administration, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.124528.

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