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Comparative life cycle assessment of Li-Sulphur and Li-ion batteries for electric vehicles

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ABSTRACT

Nowadays, most of the electric vehicles (EVs) are powered by Lithium-ion (Li-ion) batteries due to their high energy density, higher power density and degree of development relative to other battery technologies. As Li-ion technology evolves and the EVs fleet increases, it is important to understand the environmental impacts of mass-producing the battery packs for EVs. However, with 80-150 Wh/kg energy density, current Li-ion batteries are not able to power the EVs for a comparable driving range with conventional vehicles. Lithium-sulphur (Li-S) batteries have emerged as promising battery technology, with a higher theoretical capacity and energy density than Li-ion batteries used today. Moreover, Li-S batteries presumably present a lower environmental profile due to their chemical composition compared to Li-ion ones. To verify this statement, this study performs a life cycle assessment (LCA) of Li-S battery cells (under industrial development at the moment) that have been scaled up accordingly to estimate their performance as a battery for EVs. This comparison will provide the impact of each battery and the potential benefits in terms of environmental impact indicator values of the Li-S technology. The impacts of the Li-S battery are compared with those of a Nickel-Cobalt-Manganese (NCM) battery under the same driving distance. The environmental impact assessment results show that Li-S batteries present a most favourable environmental profile compared to NCM batteries, especially in the natural resource depletion categories where the Li-S battery has 70%-90% lower values compared to the Li-ion one.

1. Introduction

The continuous and expected increase of electrification in the transport sector, the so-called "electromobility" revolution, is one of the main drivers of progress in energy storage for vehicle propulsion. This change has been promoted due to the necessity of avoiding the use of fossil fuels in the transport sector, that is responsible for two-thirds of greenhouse gas (GHG) emissions worldwide (Kamran et al., 2021; Climate Watch, 2020) being the road transport responsible of the 70% of these emissions alone (Sanguesa et al., 2021).

At the end of the 19th century, the first electric vehicle (EV) was built with electrochemical batteries; however, car manufacturers devoted all their efforts in favour of the Internal Combustion Engine vehicle (ICE) because it was a more efficient and reliable technology at that time (Schiffer, 2016).

As a consequence, it was not until the end of the 20th century that the awareness and concerns of the environmental side effects of ICE emissions were raised about their impact on the environment. In fact, ICEs, and road transport in general, contribute approximately 27% GHG emissions (Deng et al., 2017). Different studies present that the current total vehicle population in 2022 is around 1.4 billion and is expected to rise to around 2 billion by 2040 representing a 60% increase (Sioshansi and Webb, 2019; Briggs et al., 2015) . Thus, the environmental impact resulting from the emissions of these vehicles would inexorably increase accordingly. To counteract this phenomenon, the European Parliament (European Commission - Climate Action) passed a law requiring all vehicles manufactured after 2020 to emit less than 95 g/km of CO_2 into the atmosphere and between 68-78 g/km by 2025 (European Commission, 2017). To meet the requirements of this new law, car manufacturers have focused their production on the development of cleaner alternative vehicles (Hackbarth and Madlener, 2013), putting most of their efforts into EVs.

EVs do not produce tailpipe greenhouse gas (GHG) emissions and do

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Nomenclature

ADP	abiotic depletion potential
AP	acidification potential
BEV	battery electric vehicles
BMS	battery management system
CML	Leiden University Institute of Environmental Sciences
CRM	critical raw material
EoL	end of life
EP	eutrophication potential
EV	electric vehicle
FU	functional unit
GED	gross energy demand
GWP	global warming potential
HEV	hybrid electric vehicles
ICE	internal combustion engine
IEA	International Energy Agency
ISO	International Standard Organization
LCA	life cycle assessment
LCO	lithium cobalt oxide
LFP	lithium phosphate oxide
Li-S	lithium-sulphur
MD	metal depletion potential
NMC	nickel manganese cobalt
ODP	ozone depletion potential
PED	primary energy demand
POCP	photochemical ozone creation potential
SoH	state of health

not rely on fuel oil as combustible. However, EVs require energy for their propulsion and are therefore responsible for GHG emissions resulting from any electricity generation process used to power them.

Today's EVs use Lithium-ion (Li-ion) batteries due to their high energy density compared to other types of batteries. With the introduction of Li-ion batteries, EVs have considerably increased their driving range and cover common urban drivers' daily needs (Fotouhi et al., 2015). Lithium-ion battery technology has evolved considerably since the first commercial unit was produced for portable applications in 1990 (Zakeri and Syri, 2015). However, this chemistry has not yet been able to replace the internal combustion engine vehicle due to its range limitations and high prices (Fang and Peng, 2015). EVs have an average driving distance between 250 and 400 km (Fotouhi et al., 2016, 2017; Bonges and Lusk, 2016), while conventional ICEs can reach 1000 km, by recharging or refuelling respectively.

The development of EV technology and its deployment goes hand in hand with advances in portable energy storage devices such as the battery. This is, in fact, the most important component of EVs as it determines the performance and price of the vehicle. Current rechargeable Li-ion batteries for EVs are capable to store around 180 Wh/kg of energy density at cell level and 120 Wh/kg at battery level, while the typical consumption of one kg of petrol produces 3350 Wh of useful work (Benveniste Pérez, 2021). There is still a factor of 19 between the energy delivered by a kilo of petrol and 1 kg of battery (for example, the autonomy of a car with a similar weight that is powered by batteries is 5-10 times less than with petrol). In fact, the driving range is one of the main issues for vehicles uses and it is determined by the energy density of the battery, which mainly depends on the chemistry as the volume and weight of the battery are a limiting factor (Deng et al., 2020).

For this reason, if the target is to reach, or even approach, the goal of a 500 km of autonomy with battery powered vehicles in the short term, it is necessary to research new materials and battery configurations.

Research to improve Li-ion batteries is very active, but some authors point out that these batteries are reaching their practical specific energy limit (200-250 Wh/kg) (Ding et al., 2019) in 2020 and it is expected to reach 450 Wh/kg by 2030 (Mierlo et al., 2021). Despite the improvements in the energy capacity, it is predicted that it will not be sufficient to meet market demands (Bresser et al., 2013; Deng et al., 2020). In order to achieve a range of 500 km and a consumption of 15kWh/100 km, estimates suggest that batteries should reach a practical specific energy of 550 Wh/kg (Fig. 1).

As a consequence, both industries and research centres are showing interest in studying alternative electrochemical energy storage systems with higher energy density. In this respect, Lithium-Sulphur (Li-S) batteries are the closest battery technology capable of meeting these expectations. Li-S batteries are one of the most promising electrochemical energy storage systems for the next generation of EVs.

Li-S batteries compared to Li-ion batteries offer a higher theoretical

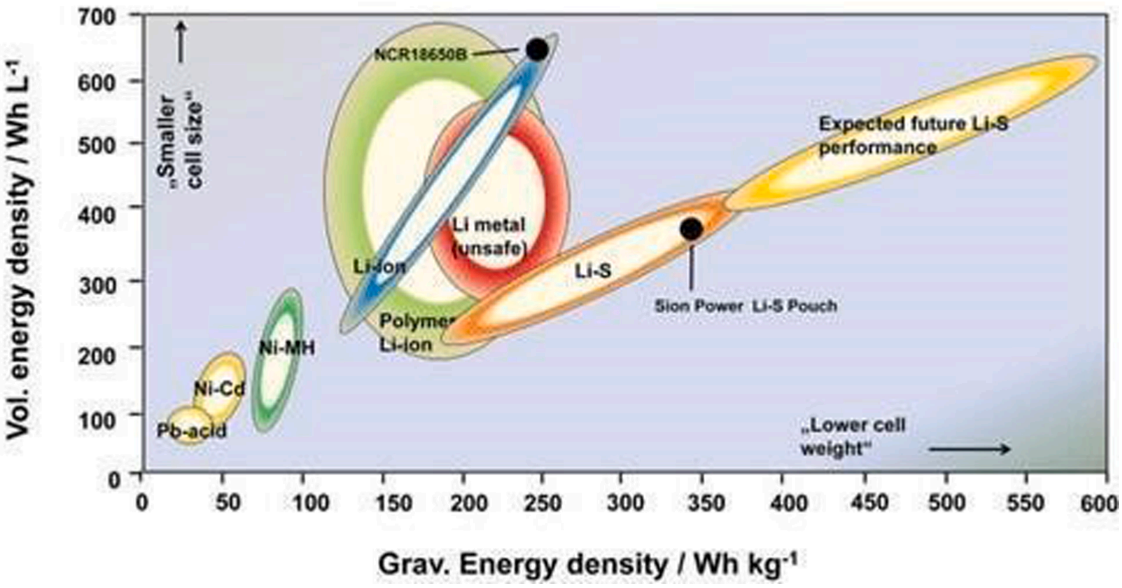

Fig. 1. Energy densities of different batteries chemistry Source (Hagen et al., 2015)

energy density (2600 Wh/kg) (Benveniste et al., 2018). In addition, the materials used present a theoretical lower environmental profile compared to the heavy metals used in Li-ion batteries (Wolff et al., 2019) Over the past years, various research projects have been developed such as EUROLIS "Advance European Lithium Sulphur cells for automotive applications'' (Eurolis, 2022) and ECLIPSE "European Consortium for Lithium-Sulphur Power for Space Environments" (ECLIPSE, 2022), to manufacturing Li-S batteries competitive enough to replace Li-ion batteries and more recently the creation of and European platform to discuss about the anode stability in Li-S batteries as a result of an homologous European project (HELIS, n.d.). HELIS managed to manufacture a prototype Li-S battery capable of reaching 500 Wh/kg of energy capacity, 1000 W/kg of power capacity, and a life longer than 1000 cycles. Although Li-S batteries offer good features to take over Li-ion batteries in EVs applications, some problems need to be solved, such as self-discharge and short cycle life.

Li-S technology is not just another modification of Li-ion chemistry. The replacement of metals by Sulphur at the cathode causes them to behave differently. Thus, many concepts learned from Li-ion cannot be implemented in Li-S batteries because of the different chain of chemical reactions that take place in one and the other (Benveniste et al., 2018). Li-S technology is however still far from commercialization, as the following key points need to be solved first:

- Solving the shuttle effect that leads to low columbic efficiency.
- Increasing power density at the cell level
- Reducing self-discharge of cells
- Increasing the lifetime of the cells

In addition to the technical challenges presented by Li-S batteries, the economic and environmental feasibility of large-scale production of Li-S batteries needs to be addressed in order to have a comprehensive knowledge about this chemistry and be capable to demonstrate that they are a real alternative to Li-ion chemistry.

Generally speaking, batteries have traditionally been considered as environmental hazards due to their toxic materials content and their availability, which necessitates their recovery through recycling. To date, only a few industrial recycling processes for Li-ion batteries are available. Moreover, these cannot be directly applied to Li-S batteries, mainly due to the low material recovery due to the high percentage of organic components (plastic, electrolyte, separators, etc.) and the unsatisfactory economic performance due to the low number, grade and value of the recovered products (Chen et al., 2015; Deng et al., 2017). Thus, it has to be checked whether the choice of theoretically low impact materials for Li-S batteries can provide and advantage over Li-ion batteries despite their low rates of recoverability.

Life Cycle Assessment (LCA) studies are thought to be valuable for assessing the potential impact of moving towards an electrified transportation infrastructure (Commission, 2022). LCA methodology has been appointed to be the most suitable approach to understand in detail the environmental performance of services and products throughout their lifecycle, and it is the basis to provide eco-designed products (Manhart et al., 2016) and improve their overall environmental profile.

LCA studies of EVs have focused on impact categories, such as climate change and energy demand. This is because variation in the electricity grid mix has a large influence on the overall result, thus decarbonisation of the grid mix will lead to further improvements in environmental impact of the EV. Along the same line, improvements in driving range and efficiency of the battery will also lead to a lower environmental impact. Despite that many efforts have focused on the analysis of the use phase of the EV, components such as their batteries, that generally use scarce and precious materials, present environmental concerns that also need to be assessed. Therefore, using an LCA approach will give a full picture of the environmental burdens caused by EVs, and more specifically, by their batteries and associated systems.

Furthermore, when dealing with EVs impacts, it is a matter of fact that batteries play a relevant role in the environmental impact of the vehicles. As a consequence, their use in the vehicle and potential uses after their "first" life is a challenge (and an opportunity) that car manufacturers should face in order to accomplish with circular economy principles and European directives regarding Circular Economy (European Commission, 2017).

In relation to studies analysing the environmental performance of batteries using LCA methodology, the literature shows a large number of publications, including some that focus on the manufacturing phase of the materials used for their different components. In particular, for producing cathode materials for Li-ion batteries, Dunn et al. (2014) indicated that the energy consumption was considerably different depending on the Li-ion battery technology. In their study, Lithium-Cobalt-Oxide (LCO) cathodes had the highest energy consumption during the manufacturing phase, followed by Lithium Iron Phosphate (LFP) cathodes which consumed half the energy required for the production of Nickel-Cobalt-Manganese (NCM) cathodes (Dunn et al., 2014).

Moreover, while in some years ago studies it was common to analyse environmental impact only in relation to greenhouse gas (GHG) emissions by taking as a key indicator the kg CO_2 equivalent emitted (Arvidsson et al., 2018; Messagie et al., 2015), nowadays there is a trend to include additional environmental impact categories, such as material depletion potential, toxicity and fossil resource depletion, among others (Bonsu, 2020), that provide a wider view on the environmental performance of the batteries.

Within these recent approaches (Dai et al., 2019), the literature agrees that LFP and NMC batteries have a lower environmental impact than other Li-ion technologies (Deng et al., 2017a). In fact, NMC performs better in some of the environmental impact categories while LFP performs better in others, having a similar overall total impact (Arvidsson et al., 2018; Messagie et al., 2015).

Therefore, NMC Li-ion batteries could be considered as the least polluting Li-ion technology (Hawkins et al., 2012, 2013).

As it has been pointed out, the environmental impact of Li-S batteries is considered to be lower than that of Li-ion batteries as Sulphur is an abundant element in nature and a waste in many industrial processes. As these batteries are still in development, scientific literature regarding the environmental performance of Li-S batteries are scares (Zhao et al., 2018; Deng et al., 2017; Benveniste et al., 2018), and more studies are required in order to have more information about their performance in comparison to Li-ion ones.

Taking into account what it has been mentioned above, the present study considers NMC technology as it is a good representative (and conservative) choice for Li-ion batteries when comparing it to Li-S technology from an environmental point of view within an EV perspective. In Deng's study, the results clearly showed that the Li-S battery was a more environmentally friendly technology than, for example, a conventional NCM-Graphite battery with 9% and 90% less impacts in most impact categories. Specifically, the Li-S battery of the study emitted 158 g CO_2 eq/km while the NCM battery emitted 174 g CO_2 eq/km. It should be noted that these values were obtained using a particularly unfavorable electricity mix, such as the Chinese electricity mix (Deng et al., 2017). In fact, these values would be clearly lower if an average European electricity mix were considered, where the prevalence of renewable energies is higher. Though this study is very interesting as it was the first one addressing the comparison of these 2 technologies, Deng's must be contrasted by a scenario assessment using different electricity mixes that could occur if the batteries or their components were manufactured in countries with a more favorable electricity mix from an environmental point of view together with a real analysis of the performance of the battery in the use phase. More recently, a first attempt to provide more insights on the environmental aspects related to Li-S battery for EV, together including real data concerning its performance in a first life in the vehicle and potential second life in stationary use was studied by Wolff et al. (2019). In this paper, we proceed to continue the research presented by Wolff and perform a comparative LCA of Li-S and Li-ion batteries for EVs applications using the scaling of Li-S coin cells produced in a laboratory and literature data collected from an industrial manufacturer of a Nickel-Cobalt-Manganese (NCM) battery, to understand the relative significance of the life cycle environmental impacts of both batteries.

The present study is motivated by the need for quantitative information on the environmental performance of Li-S technology for energy storage systems for EVs and complete the studies that have been presented before. Furthermore, this environmental performance shall be compared with the most competitive Li-ion technology for EV application. In this way, useful information will be made available to the scientific community for decision-making regarding the choice of the most suitable storage system, from an environmental point of view, for EVs.

2. Material and methods

The environmental assessment of both Li-S and Li-ion batteries has been carried out using LCA methodology. LCA methodology applied to these products is considered to be of great value in analysing the potential impact of moving towards electrified transport infrastructure (Drabik and Rizos, 2018). In fact, LCA methodology has been designated as the most suitable approach to understand in detail the environmental performance of services and products and is the basis for delivering ecologically designed products (Manhart et al., 2016).

The LCA provides an analysis of the potential environmental impacts of a product or service throughout its life cycle, including the extraction of raw materials and the production, use and final disposal of the product. The implementation of the LCA is regulated by the International Organization for Standardization (ISO): ISO 14040 Environmental management. Life cycle assessment. Principles and framework and ISO 14044 Environmental management. Life cycle assessment. Requirements and guidelines (ISO 14040:2006, 2006 (ISO, 2006)) (ISO 14044:2006, 2006 ISO, 2006).

According to ISO 14040, LCA is divided into 4 phases: Definition of the objective and scope, analysis of the life cycle inventory, evaluation of the environmental impact of the life cycle and interpretation.

2.1. Goal and scope, boundaries and functional unit definitions

In this phase of the LCA, the goal and scope of the study shall be defined. The scope definition entails the clear definition of:

- The system of the product to study
- The function of the system
- The functional unit
- The limits of the system
- Data source and data quality requirements
- The hypotheses and limitations
- The procedure for assigning environmental loads, the selected impact categories, the impact assessment methodology and the interpretation.

As stated before, the purpose of this study is to compare the environmental impacts of a 50 kW Li-S battery (virtually built from the scaling up of Li-S coin cells) with a 35,8 kW Li-ion battery used in a Volkswagen e-Golf. An LCA with a ‘cradle to grave’ scope was implemented to analyse the impact of the cathode, anode and housing of the battery pack of each one of the chemical batteries. It is important to remark that data for the NMC battery was obtained from industry and therefore corresponds to a commercialized battery, while data for the Li-S corresponds to experimental conditions.

The scope of this LCA is defined as "from the cradle to the grave", i.e. the environmental impact will be assessed from the extraction of raw materials, through the manufacturing phase, the phase of use in an EV, until its final management, including the recycling operations and potential materials recovery.

Fig. 2 shows the system boundary of the battery life cycle, from the extraction of raw materials, the manufacture of components and the battery assembly, the battery usage in the EV where the energy is evaluated and the end of life that includes the possible operations of recycling and raw materials recovery. The transport operations, the production of other components of the EV and the option to give a second life to the battery are outside the limits of the system.

The functional unit (FU) is a key element of LCA and should be clearly defined as it gives a relationship between inputs and outputs and allows comparison between different systems. The function of the system is to provide enough energy for the proper operation of the electric vehicle for 150,000 km.

Therefore, the FU chosen for this study has considered the energy that is delivered by the battery to enable 150,000 km vehicle drive, the number of charging-discharging cycles and the capability to enable the comparison between the Li-ion and the Li-Si battery. Since both batteries have a different energy capacity and size, it has been decided to use 1 kWh as a FU, to allow a simple comparison between the 2 batteries technologies.

For the assessment of the LCA impact categories, the midpoint approach has been used, calculated with the CML 2011 (CML Institute of Environmental Science at Leiden University, 2022) and ReCiPe 2008 (Huijbregts et al., 2017) methods in the GaBi software (Software, n.d.). The impact categories evaluated are Abiotic Depletion (ADP), Acidification Potential (AP), Eutrophication Potential (EP), Global Warming Potential (GWP), Photochemical Ozone Creation Potential (POCP), Mineral Resource Scarcity (MD) and Primary Energy Demand (renewable and non-renewable, PED).

The selection of these impact categories responds to the necessity of using the most appropriate and comprehensive indicators to characterise the potential environmental impacts of the battery analysed. For this reason, on the one hand, it has been decided to include the indicators that measure the depletion of mineral resources (ADP elements) and the potential for scarcity of mineral resources (MD) and energy demand (PED) to measure the consumption of raw materials and energy resources. The choice of two apparently similar indicators such as ADP elements and MD is due to the fact that the former considers the total depletion of resources, while the latter places more emphasis on their scarcity and is in line with the problems described in the introductory chapter on the use of critical raw materials. On the other hand, impact categories have been chosen that characterise effects on air (e.g. climate change category - GWP -, acidification - AP - or photochemical ozone formation - POCP -) and on land and water (eutrophication potential - EP and acidification - AP), which give a comprehensive picture of environmental effects on all media.

The data referring to the materials used for the manufacture of the NMC Li-ion battery and its properties, have been provided by a company in the automotive sector, specifically SEAT, SA for the primary data. Primary data for the Li-S coin cells where obtained from experimental processes at laboratory scale. In both cases, secondary data was obtained from the database GaBi Professional (Software, 2022) and EcoInvent 3.5 (Ecoinvent Centre, 2020) included in the database of the GaBi software.

2.2. Life cycle inventory

Table 1 presents the main characteristics of the two types of batteries. Production data of the Li-ion battery was given by the manufacturer while the production data of the Li-S battery was based on the scaling of Li-S coin cells produced in a laboratory. The details of the Li-S coin cells and the scaling up procedure were already explained in other publications from the same authors (Benveniste Pérez, 2021; Wolff et al., 2019) and it is briefly introduced as follows.

The inventory has been split into the 3 main life cycle stages of the batteries: materials and manufacturing, use phase and end of life. Since the study is focused on analysing the first life of these batteries in the

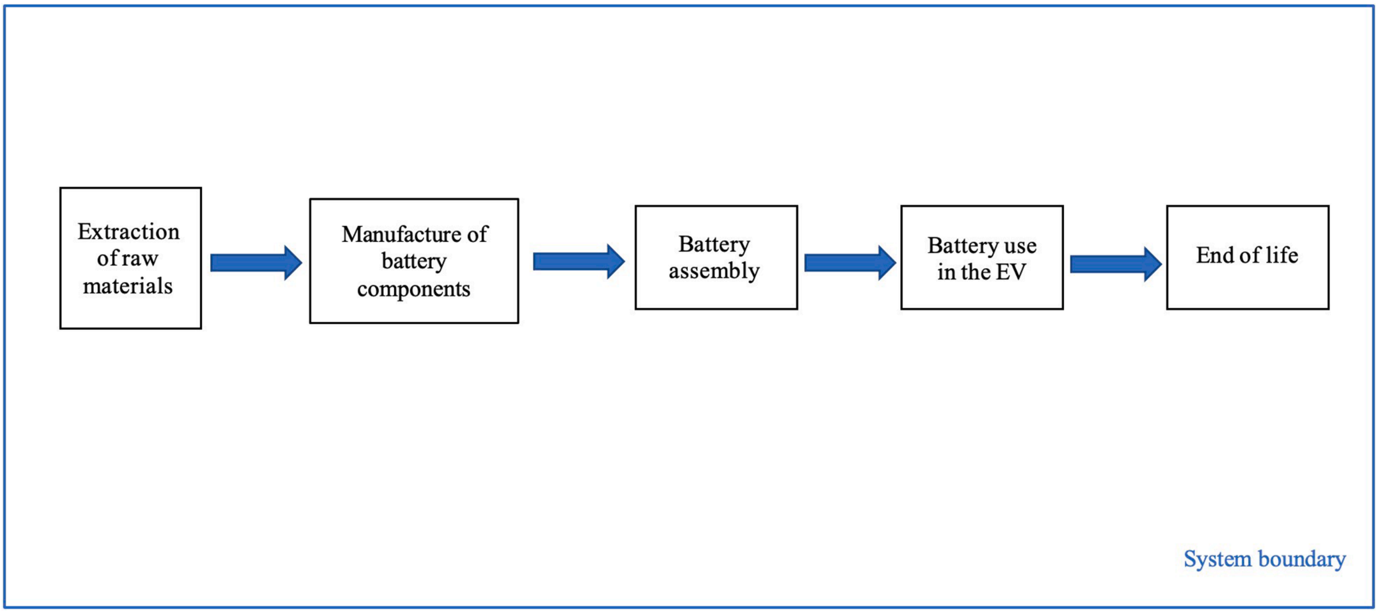

Fig. 2. System boundary of the battery life cycle

Table 1
Characteristics of the two types of batteries

Battery chemistry	Li-S	Li-ion
Capacity (kWh)	50	35.8
Energy for driving (kWh/km)	0.2	0.154
Cycles	1000	850
Total driving distance (km)	150000	150000

Table 3
Li-ion battery mass breakdown

Component	Mass	Units
Anode	68.04	kg
Li Cu		
Cathode	120.96	kg
Ni Mn Co Al		
Battery pack	123	kg
Steel Al Cu Polymer Others		

vehicle, the life cycle stages here reported do not include any operation related to the collection of exhausted batteries from the vehicle to be reconditioned for a potential second life in other applications.

2.2.1. *Batteries materials and manufacturing*

This phase includes the inventory of all the raw materials, materials and components production processes, considering the different chemistries of Li-ion and Li-S battery here studied.

The Li-ion battery consists of 288 cells, and each cell weighs 0.656 kg. As it is an NCM battery, the anode is composed of Li and Cu and the cathode is composed of Ni, Mn, Co and Al (Table 2). The mass breakdown of the battery among active material and mechanical components is depicted in Table 3.

Concerning the Li-S battery, it has been modelled after the scaling up of Li-S coin cells to obtain the 50 kWh battery.

As presented by Wolff et al. (2019), the gravimetric energy density (GED) and the ratio of total mass to active mass employed in the coin cells were used to quantify the amount of active material (electrolyte, anode and cathode) required for a 50kWh Li-S battery. The GED and active mass have been calculated using Eqs. (1) and (2), respectively. Laboratory data as primary data has been used to quantified the electrolyte amount. Similarly, the quantity of lithium anode in the experimental coin cells was calculated considering that the diameter of the Lithium ribbon is the same as the cathode one. A ratio of 1:1 of carbon to sulfur has been considered to the cathode's active material. The mass of the different components of the coin cells are provided in Table 4. It should be noted that Eqs. (3) and (4) have been used to calculate the mass the mass of the active material, as depicted in **Error! Reference source not found.**.

$$GED = \frac{Ccc}{M_{AM}} \; x \; Vcc \tag{1}$$

Table 4
Inventory of the coin cell (Wolff et al., 2019).

Component	Mass	Unit
Anode (*MA_{CC}*)	0,0064	g
Cathode (*MC_{CC}*)	0,0078	g
Electrolyte (*MEl_{CC}*)	9,11	µl
Mass active material (*MAM*)	0,026	g
Mass coin cell (*TMCC*)	3,59	g
Capacity (*CCC*)	3,3	mAh
Voltage (*VCC*)	2,3	V
Capacity Density (*CCC / MAM*)	128,4	Ah/kg
Gravimetric Energy Density (GED)	295,4	Wh/kg
Energy Li-S Battery (EB)	50	kWh

Table 2
Composition of the Li-ion battery

Composition housing of battery pack [%]	Cell composition [%]	Cell mass [kg]	Total mass [kg]						
Steel	Al	Cu	Plastic	Others	Ni Mn Co	Al	Cu	189	312
68	2	10	19	1	15	19	13		

$$M_{AM} = M_{El\ cc} + M_{Acc} + M_{Ccc} \tag{2}$$

$$Scaling\ Factor = \frac{1x10^6}{M_{AM}} x \frac{E_B}{GED} \tag{3}$$

$$M_{X_B} = M_{X_{CC}}\ x\ \frac{Scaling\ Factor}{1000} \tag{4}$$

where *CCC* is the capacity (mAh) of the coin cell, *VCC* is the voltage of the coin cell, *MAM* is the mass (g) of the active material in the coin cell, *MElCC, MACC* and *MCCC* are the masses (g) of electrolyte, anode and cathode in the coin cell, *EB* is the energy (kWh) of the battery, and GED is the gravitational energy density (Wh/kg) *MXB* (kg) and *MXCC* (g) are the masses in kilograms and grams for X (electrolyte, anode or cathode) in the battery (B) and coin cell (CC), respectively.

The quantification of the mass of the cell container, separator, module and pack packaging, and cooling system has been done considering literature data extracted from the LCA study carried out Li-S batteries (Deng et al., 2017a). This LCA study considered the Argonne National Laboratory BatPac software (Argonne, 2022). The BatPac software was employed to adapt the values calculated for a Li-ion battery to a Li-S one. Similarly, the data regarding the energy consumption during the manufacturing of the components and battery assembly was obtained from the estimations included in Deng et al. (2017a) (Table 5).

2.2.2. *Use phase in the vehicle*

For the use phase, the energy consumed by the battery during its useful life in the vehicle was evaluated twofold: regarding the Li-ion battery, the data provided by the vehicle manufacturer has been used to quantify the electricity consumption during battery usage; regarding the Li-S battery, the average energy requirements were given as a project target by the automotive industry in the HELIS project (HELIS, 2022; Wolff et al., 2019) and data corresponds to the cycling tests that were carried out for the Li-S cells. In both cases, the A European electric mix available in the GaBi database has been used to model the energy consumption during the manufacturing and use phase Fig. 3.

Based on manufacturers' descriptions and existing literature, when EV batteries reach 70-80% of their rated capacity, it is recommended to replace them (Canals Casals et al., 2017). This percentage of residual battery capacity is called "State of Health" (SoH) in Table 6.

2.2.3. *End of life*

At the end of life, the battery is removed from the EV when its SoH is 70% in the case of Li-ion and 60% in case of the Li-S, (difference is due to the fact that Li-S battery has a larger energy capacity) and is recycled to recover as much material as possible and avoid hazardous waste production. In the end-of-life phase of the Li-ion battery, it has been considered to be removed from the vehicle at the end of its useful life and recycled with the aim of recovering as many materials as possible, as well as avoiding the production of hazardous waste. The information used to calculate the environmental impact of the Li-ion battery recycling is provided in Costa and Dewulf studies (Costa et al., 2021; Dewulf et al., 2010) which explains that battery recycling can lead to a reduction of up to 50% of battery life impacts on the battery manufacturing process. Thus, it is considered that the environmental impact generated by the recycling of batteries in this paper it's -50% of the impact of the production of the battery as the recovered materials are assumed to save its equivalent in virgin material Fig. 4.

Table 5
Li-S battery mass breakdown

Li-S battery composition	Mass [kg]
Anode	42.0
Cathode	51.4
Electrolyte	75.9
Separator	6.9
Cells container	19.6
Module pack	22.6
Cooling system	27
Battery pack	41.8

For the end of life of the Li-S battery, it has been considered that the battery is removed from the vehicle at the end of its useful life and is managed in order to be treated with the aim of recovering as much material as possible and avoiding the production of hazardous waste. Data for the treatment of Li-S batteries has been provided by the partners of the HELIS project (HELIS, 2022) which has developed a treatment process for Li-S cells that complies with the European Union (EU) directive 66/2006 (Parlamento Europeo y Consejo de la Unión Europea, 2006). This directive sets a minimum recycling efficiency requirement for batteries of 50% of the battery mass.

The recycling process, developed by ACCUREC as partner of the HELIS project (Accurec, 2022), are described as follows:

In battery bulk discharging:

- Electric conductive solution (in this study, KOH solution) was used to discharge the cylindrical Li-S batteries. This is relatively long period, compared with Li-ion batteries who needs <2 weeks.

In vacuum thermal treatment:

- Due to the enormous pressure released from the batteries during thermal treatment, strong pressure shock needed to be considered in the construction of thermal treatment equipment (e.g. using robust container).
- Due to their similar vapour pressures in the same temperature range, electrolytes and sulphur could evaporate together out from Li-S cells. The sulphur weight share and species in off-gas were identified to determine the loss of sulphur in off-gas.
- In order to completely pyrolyze the organics, the vacuum treatment operating parameters were: temperature >400°C, holding time > 1h, pressure < 400 mbar.
- After vacuum thermal treatment, the solid materials were still reactive, possibly due to the excess reserve and incomplete reacted metallic lithium in the battery. Therefore, an oxidation step was added after vacuum thermal treatment to completely deactivate the material.
- The weight loss of Li-S cylindrical batteries in thermal treatment is ca. 37wt%, which is higher than that of Li-ion battery cells (10-25% wt), due to the high share of electrolytes.

In mechanical treatment:

- By shredding, magnetic separation and sieving, three main outputs were resulted: steel flakes, aluminium flakes and active mass powder.
- The steel flakes have high purity and could be fed to steel making plants to produce new steel. The active mass powder will be conducted to subsequent hydro-treatment for recovery of lithium and other elements (Al, C). The aluminium flakes have relatively low purity. It could be fed to Al-remelter to produce new aluminium metal or integrated in the active mass fraction and recovered as Al (OH)3 in hydro-treatment step.

In hydro-treatment of active mass:

- From the developed hydro-treatment process, three main products were obtained, namely 1) lithium carbonate, 2) S-impregnated carbon powder and 3) Al-hydroxide. By-products such as copper sulphides, sodium sulphate were obtained.
- The purity of lithium carbonate product did not yet reach the battery grade (>99.0% Li_2CO_3), mainly due to the sodium residue. But it was appropriate to be used as an additive in glass and ceramics industries.

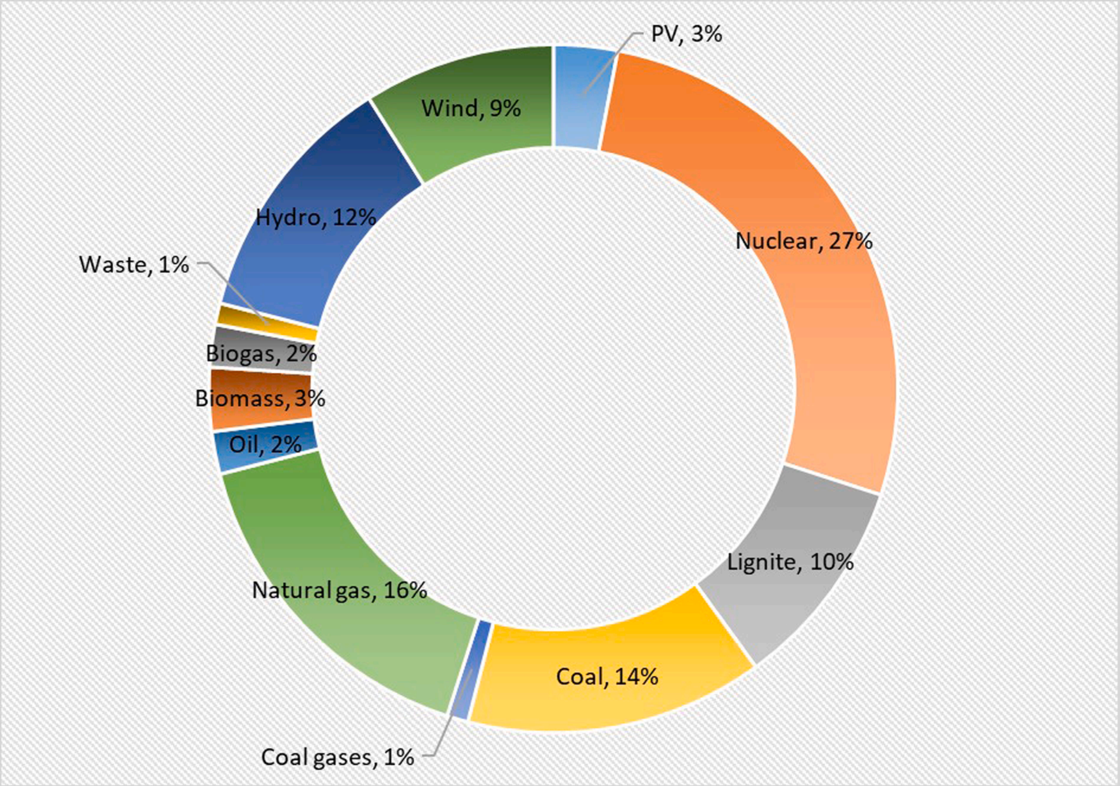

Fig. 3. European Energy mix (source:(Software, n.d.), 2020)

Table 6
Use phase inventory

Parameter	Quantity Li-ion	Quantity Li-S	Units
Storage energy capacity	35.8	50	kWh
Energy required per km	0.154	0.2	kWh/km
Expected cycles	1000	1000	Cycles
Distance driven	150000	150000	km
SoH at the end of first life	70-80%	60%	-

Otherwise, additional re-crystallization step could be followed to lower the impurity content to reach battery grade Li_2CO_3.

- The Al-hydroxide can be re-integrated into standard Al production route to produce virgin Al metal.
- The carbon powder product still contains considerable Sulphur content (approx. 9% S). It can be pelletized to make sulphur-impregnated activated carbon pellets for cleaning/adsorption of mercury-containing off-gas. Or, it might be re-used for Li-S battery electrode, which likely needs further refining and tests in the future.

In summary, the following five main products can be obtained from the battery treatment process Fig. 5, that corresponds approximately to the 50% of the total weight of the battery:

- Steel flakes
- Aluminium flakes
- Lithium carbonate (Li2CO3)
- Carbon powder (sulphur impregnated)
- Aluminium hydroxide

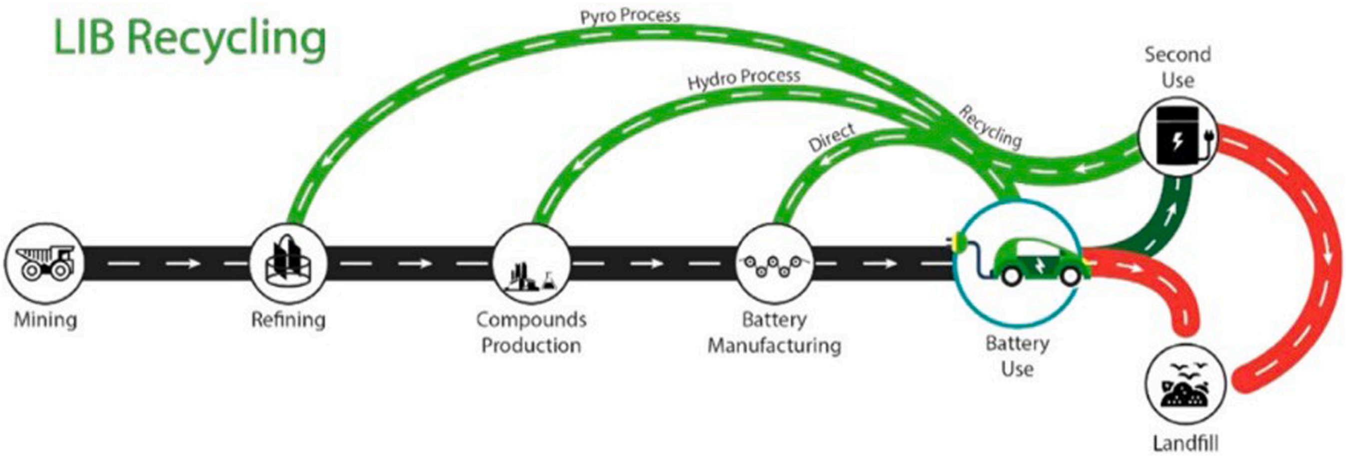

Fig. 4. Li-ion battery end of life management scheme (source: (Korthauer, 2018))

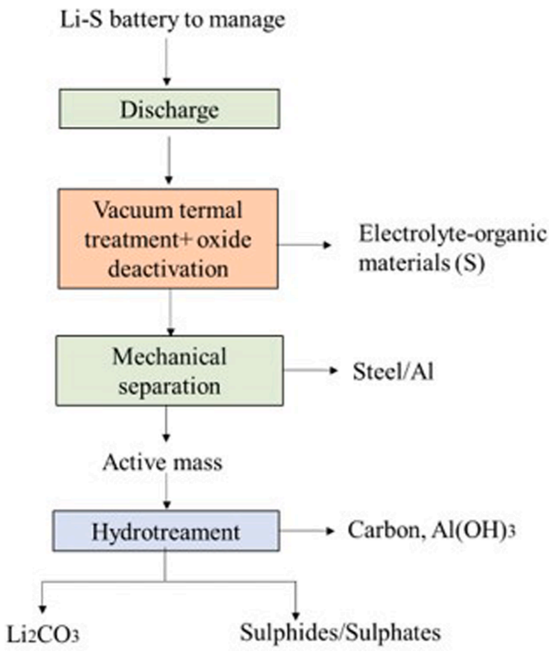

Fig. 5. Li-S battery recycling operations (source: (Accurec, 2022; Benveniste Pérez, 2021))

3. Results and discussion

The LCA results for the cradle to grave analysis of the two types of batteries are shown in Table 7, calculated for the selected FU to enable a proper comparison.

Comparing the overall performance of both batteries (total results) for the different impact categories, as expected, the Li-S battery presents a most favourable environmental profile compared to the NMC Li-ion battery, as shown in Table 7 and Fig. 6. It should be noted that the environmental impacts of the Li-S battery have been evaluated from a coin cell scaling as these batteries are still under development, while the Li-ion battery has been evaluated with a full production capacity at an industrial scale.

More specifically, differences are significant for those impact categories that deal with the assessment of the use of resources, which is the case of ADP or MD. Concerning the ADP impact category, Li-S battery presents a value of 0.0005 kg Sb eq/kWh, for the manufacturing stage, which is 88% lower than the value presented by the Li-ion battery. Equivalent trend is found for the MD impact category, where Li-S value is 91% lower compared to the Li-ion battery value in the manufacturing stage. The reason of these differences can be found if considering that the Li-S battery does not use metals such as nickel, cobalt and manganese, and uses much less copper than a Li-ion battery. These metals are found in the cathode composition. Therefore, this component has been analysed in depth.

Regarding the GWP indicator and therefore the GHG emissions, the manufacture of the Li-S battery generates 89.8 kg CO_2 eq./kWh, 35% lower than the emission of 139 kg CO_2 eq./kWh of Li-ion battery manufacturing. This GWP impact reduction can be mainly attributed to low-impact materials such as sodium thiosulfate used in the manufacturing of Li-S batteries. The GHG emissions in the battery usage phase can be reduced from 256 kg CO_2 eq./kWh to 183 kg CO_2 eq./kWh. In general, a 31% reduction in the life cycle in GHG emissions can be achieved by using a Li-S battery instead of a Li-ion battery.

In terms of the EoL of the Li-S battery that includes the components management and potential recycling and recovery of materials has a negligible impact when considering the overall life cycle impacts. However, the materials recovery potential benefits are not sufficient to negativize the value of the EoL as happens for the Li-ion battery EoL. The environmental impacts associated to the recycling process (energy consumption) are high enough to balance the potential benefits in terms of negative impacts due to the materials recovery. It should be stated that for the Li-S battery experimental recycling process were used and therefore they were fare from being optimized and be effective as, it can happen, for Li-ion battery.

In contrast, regarding the Li-ion EoL, negative results are explained as potential avoided impacts due to the materials recovery in the recycling operations, following the assumptions stated in the methodology section and they can reach up to 48-49% in relation to the manufacturing stage.

As stated before, it has been interesting to consider in detail the impacts associated to the manufacturing stage since the LCA assessment has led to identify this stage as the one with a major contribution to the overall results for both batteries (with exception of the use phase, that for the GWP potential has a major contribution). The impacts associated with the manufacturing stage deal with the materials selection and the manufacturing process of the batteries, and, more precisely, to the production of the cathode.

This is why a detailed comparison of the cathode performances has been carried out, focusing on the manufacturing stage, obtaining the results shown in Fig. 7.

In all categories, except eutrophication (EP), the cathode environmental performance of the Li-S battery is significantly lower compared to the cathode impact of the Li-ion battery. The impact categories where we can observe a most significant reduction are abiotic depletion (ADP) with a 94% reduction, acidification (AP) with an 89% reduction, photochemical ozone creation potential (POCP) with an 80% reduction and scarcity of natural resources (MD) with a 99% reduction. This high difference in impact between the cathode of the two batteries is due to the absence of nickel, cobalt and manganese in the Li-S battery. In terms of the global warming potential (GWP) and primary energy demand (PED) impact categories related to the emission of pollutants into the air during electricity generation, the use of a sulphur cathode can be reduced by 22–43%. The eutrophication impact is higher in the Li-S battery due to the SOx emissions produced by the extraction of sulphur to manufacture the cathode.

4. Conclusions

This study has performed an LCA of two battery chemistries to identify the main hotspots and potential benefits due to the use of

Table 7
LCA comparative results

		Li-ion	Li-S						
Impact categories	Units	Manufacture	Use	EoL	Total Li-ion	Manufacture	Use	EoL	Total Li-S
ADP	kg Sb eq./kWh	4.05E-03	8.52E-05	-2.00E-03	4.14E-03	5.00E-04	6.10E-05	6.04E-09	5.37E-04
AP	kg SO_2^- eq./kWh	2.34E+00	5.30E-01	-1.17E+00	2.87E+00	5.00E-01	3.80E-01	2.76E-04	8.78E-01
EP	kg PO_4^{-3} eq./kWh	2.90E-01	6.00E-02	-1.50E-01	3.53E-01	2.20E-01	4.00E-02	4.47E-05	2.63E-01
GWP	kg CO_2 eq./kWh	1.39E+02	2.56E+02	-6.93E+01	3.94E+02	8.98E+01	1.83E+02	2.60E-01	2.73E+02
POCP	kg $C_2H_4^-$ eq./kWh	1.10E-01	4.00E-02	-5.00E-02	1.46E-01	3.00E-02	3.00E-02	-1.08E-05	5.93E-02
MD	kg Cu eq./kWh	8.24E+01	3.40E-01	-4.12E+01	8.28E+01	7.27E+00	2.40E-01	6.72E-05	7.51E+00
PED	MJ/kWh	2.82E+03	6.59E+03	-1.41E+03	9.41E+03	1.59E+03	4.72E+03	2.50E-01	6.31E+03

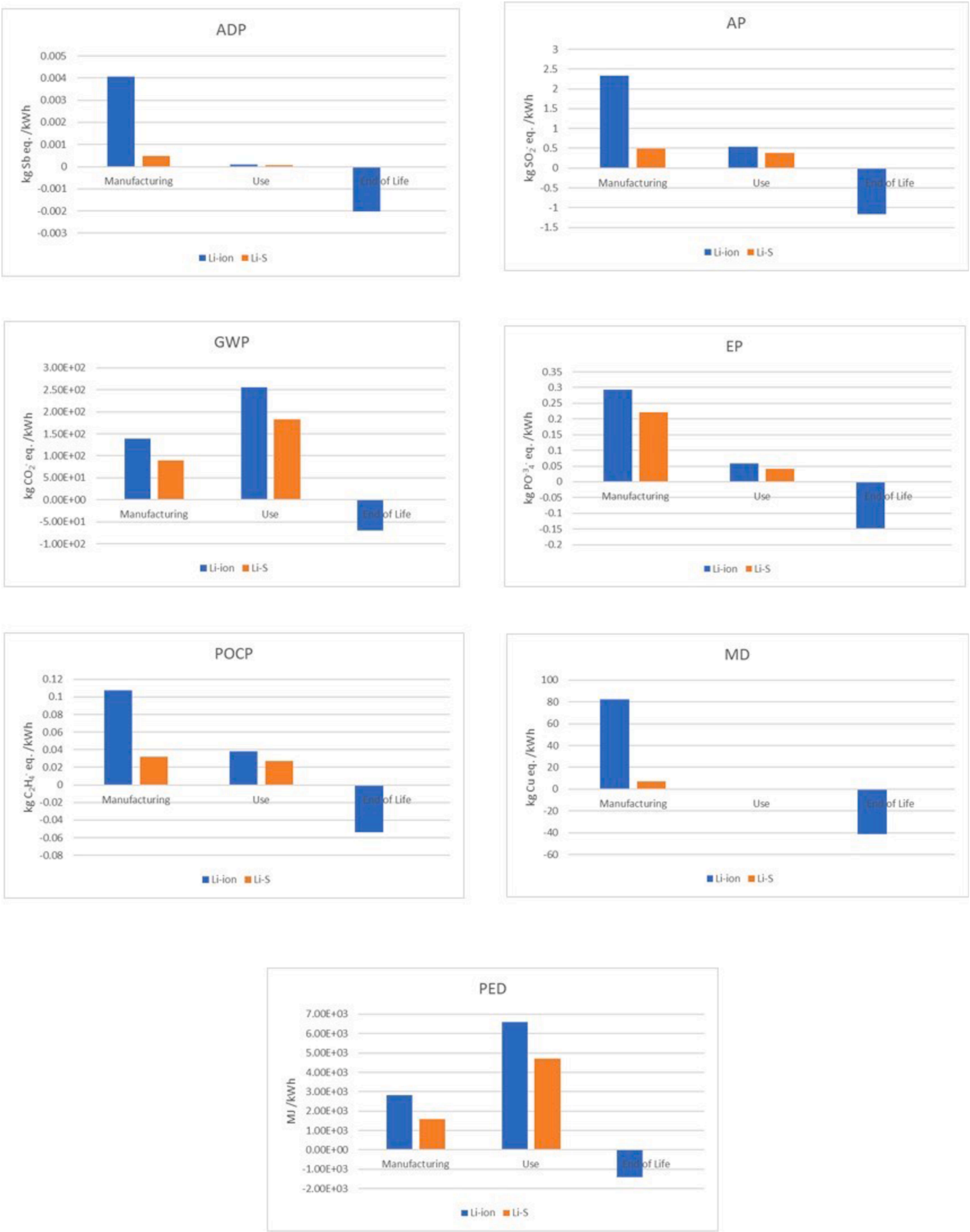

Fig. 6. Life cycle impact benchmarking between the Li-ion and Li-S battery packs for the different impact categories

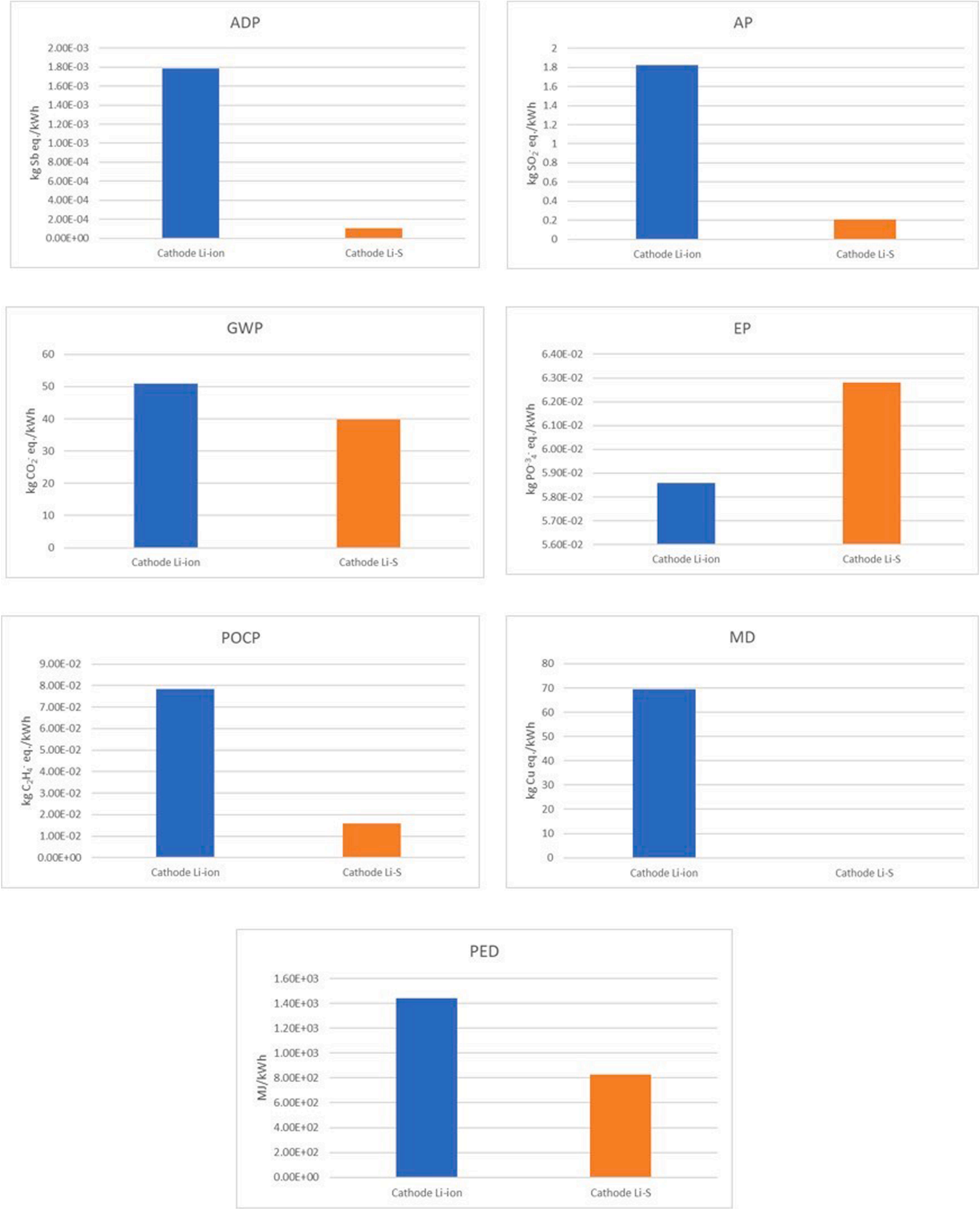

Fig. 7. Cathode impact benchmarking between the Li-ion and Li-S battery packs for the different impact categories

Sulphur based chemistry.

The results of the LCA of the 35.8 kWh Li-ion battery have enable the identification of influence of each stage of the battery on the environmental profile. These results indicate that the environmental impacts of the battery vary significantly depending on the phases of its life cycle. In particular, all the results of the impact categories associated with the manufacturing phase are considerably higher than the other phases, except the impact of global warming potential (GWP) (394kg CO_2 equivalent/kWh) and primary energy demand (PED), that present higher values in the use phase. The production phase, which includes the extraction of materials and the manufacture of components, represents the main contributor (up to 70-90%) to the categories of impact related to the consumption of raw materials such as the depletion of abiotic resources (ADP) and the scarcity of natural resources (MD).

The in-vehicle battery phase contributes the most in the categories of global warming potential (GWP) and primary energy demand (PED). Regarding the category of primary energy demand (PED), the value obtained is so high due to the consumption of electricity during the 150,000km of driving the vehicle (0.154 kWh / km) since it has been used a European electric mix that is largely made up of fossil fuel sources.

With this additional analysis, it has been observed that the cathode of the battery is the component that influences the most with 51% of the total environmental impact of the manufacture, since it is made up of metals such as nickel, cobalt and manganese.

The materials recovery in the recycling process included in the end of life can lead to a reduction around 48-49% of the values of the impact categories related to the natural resources depletion, such as ADP or MD.

The LCA results for the Li-S technology have included the evaluation of one battery capable to meet the 150000 km and arriving at a SoH of 60%. results showed that the GWP of the battery is 273 kg of CO_2 equivalent/kWh, being the use phase the life stage that contributes up to 73% of the overall result. The contribution of the battery's components and the consumption of raw materials such as steel, aluminium copper and electronic boards contribute significantly to the ADP and MD indicators (86% and 96% respectively on the overall result). As a general conclusion, according to the LCA results, the introduction of Sulphur in the composition of the cathode, in replacement to lithium metals, contributes to improve the environmental profile of the Li-S battery compared to Li-ion. However, the data in this study has been scaled from Li-S coin cells, and thus must be updated when data on larger Li-S batteries is available. Indeed, results here reported, Li-S batteries are thought to be a promising technology for use in electric vehicles in order to reach higher practical specific energies than Li-ion batteries and reduce the quantity of raw materials required. However, as more data is obtained and more tests are done on larger Li-S batteries, the results in the LCA studies should be updated to improve the quality of the data.

Considering than one of the objectives of the LCA was to compare the environmental performance of these two batteries, from the results it can be stated that Li-S batteries (and specifically, the battery object of the study) present a most favourable environmental profile compared to conventional Li-ion batteries.

In fact, the use of a Li-S battery instead of a Li-ion battery can lead to a reduction of up to 31% in GHG emissions. With the LCA results of the Li-ion battery, we acknowledge that the component with the highest contribution in the manufacturing phase was the cathode, so it was decided to make an additional analysis to go into detail about the impact of the cathode in both batteries. The results show that the impacts of cathode production of a Li-S battery in the categories of abiotic depletion (ADP), acidification (AP), photochemical ozone creation potential (POCP) and natural resource scarcity (MD) can be reduced between 80% and 99% compared to a Li-ion battery.

The results here provided are aligned to what it was presented in previous studies that were mentioned in the literature review in section 1 (Zhao et al., 2018; Deng et al., 2017b), where Li-S batteries' environmental profile is more favorable compared to NMC Li-ion batteries and complete these studies as this time real data coming from battery testing performance have been employed.

It can be stated that Li-S batteries are a good energy storage alternative for EV in terms of sustainability compared to current Li-ion batteries, mostly by the absence of toxic metals such as nickel, cobalt and manganese. However, it is important to note that Li-S batteries are still under development, and the LCA results have been obtained by evaluating a coin cell scaling, while the Li-ion battery has been assessed with a full production capacity at industry scale.

CRediT authorship contribution statement

Gabriela Benveniste: Conceptualization, Data curation, Methodology, Software, Writing – original draft. **Anna Sánchez:** Data curation, Writing – original draft. **Hector Rallo:** Data curation, Investigation. **Cristina Corchero:** Software, Supervision, Validation. **Beatriz Amante:** Writing – review & editing, Supervision.

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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