BATTERIES FOR ELECTRIC MOBILITY: LIFE CYCLE ASSESSMENT AND PROCESSES FOR RECYCLING OF RAW MATERIALS

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List of Publications

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List of Acronyms[1]

LIB	Lithium-Ion Battery
EV	Electric Vehicle
LCA	Life Cycle Assessment
ROI	Return On Investment
DCF	Discounted Cash Flows
NPV	Net Present Value
LCO	Lithium-cobalt-oxide
LMO	Lithium-manganese-oxide
LNMO	Lithium-nickel-manganese-oxide
LTO	Lithium-titanate-oxide
NCM	Lithium-nickel-cobalt-manganese-oxide
LFP	Lithium-iron-phospate
NCA	Lithium-nickel-cobalt-aluminium
LVP	Lithium-vanadium-phospate
PVDF	Polyvinylidene fluoride
PC	Propylene carbonate
EC	Ethylene carbonate
EMC	Ethylmethyl carbonate
DMC	Dimethyl carbonate
DEC	Diethyl carbonate
HEV	Hybrid electric vehicle
PHEV	Plug-in hybrid electric vehicle
BEV	Battery electric vehicle
GHG	Greenhouse gases
EU	European union
ICEV	Internal combustion engine vehicle
WEEE	Waste electrical and electronic equipment
IE	Industrial ecology
FU	Functional unit
ISO	International standard organization
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
CED	Cumulative energy demand
WTW	Well to wheel
CTG	Cradle to grave
GWP	**Global warming potential**
SOD	**Stratospheric ozone depletion**

[1] In order of occurrence

IR	Ionizing radiation
OF-HH	Ozone formation, human health
FPMF	Fine particulate matter formation
OF-TE	Ozone formation, terrestrial ecosystem
TA	Terrestrial acidification
FE	Freshwater eutrophication
ME	Marine eutrophication
TEcotox	Terrestrial ecotoxicity
Fecotox	Freshwater ecotoxicity
Mecotox	Marine ecotoxicity
HCTox	Human carcinogenic toxicity
HnCTox	Human non-carcinogenic toxicity
LU	Land use
MRS	Mineral resource scarcity
FRS	Fossil resource scarcity
WC	Water consumption

Abstract

Climate change is pushing to rethink the paradigm of anthropic activities. From a society strongly based on fossil fuels and their exploitation, there are many initiatives that aim at a transition towards sustainable energy sources such as the European Green Deal and the European Taxonomy on sustainable finance that aim to promote sustainability and circular economy. Thanks to technological improvements over the past decade in electric storage systems, one of the initiatives is related to the electrification of road transport. It is precisely in this transition that lithium-ion batteries have found ample space thanks to their energy density, which allows the accumulation of energy in weights and volumes that are no longer prohibitive. Looking at global electric vehicle adoption trends, it is possible to identify an exponential growth in battery demand and consequently in the consumption of raw materials such as lithium, cobalt, manganese and nickel. The extraction of these raw materials has strong environmental and social impacts and the availability of these raw materials is limited. To reduce the environmental impacts related to the extraction of raw materials and the production of lithium-ion batteries and to make supply chains more sustainable and circular, it is possible to think of recycling processes that allow the recovery of materials contained within them. The current state of the art in lithium-ion battery recycling technology involves two types of metallurgical processes: pyrometallurgy and hydrometallurgy. The former involves the use of heat to alloy the metals of interest while the latter involves the use of organic/inorganic acids for the selective extraction of those metals. This Ph.D. thesis is one of the first available works on the topic that aims to assess the techno-economic and environmental sustainability of those lithium batteries recycling processes considering different chemistries, plant scales and an Italian scenario. This dissertation is organized in three Chapters.

Chapter I describes the state of the art about the application of LCA to lithium-ion batteries and electric vehicles through a critical review of papers published on the topic. The chapter points out the criticalities of LCA applied to the LIBs and to EVs. Furthermore, to highlight strengths and weaknesses of the assessment procedures adopted, the analysis of the reviewed studies was carried out also from a methodological point of view. From a practical perspective, some LCA results were analysed and discussed. From a methodological point of view, an in-depth analysis of the reviewed papers was carried out to

highlight the different methodological approaches followed as well as the key aspects of variability based on the LCA phases (Goal and scope definition; Inventory Analysis; Life cycle Impact Assessment; Interpretation).
Chapter II deals with the technical confrontation among electric and thermal vehicles and the methodological confrontation between two LCA software tools: Simapro and Greet. The aim of this section is to firstly obtain data regarding the use phase of lithium-ion batteries to be used in the overall life cycle assessment in chapter III. Secondly the comparison aims to understand if the electrification of the mobility is the solution to reduce the environmental impacts related to the transportation sector. The comparison shows that electric vehicles are better than thermal one only when looking at Climate change impact categories. Performing a broader analysis shows that thermal vehicles represent non the worst solution.
Chapter III presents the assessment of the environmental and economic performances of all the lithium-ion batteries chemistries and recycling processes considered. About the environmental evaluation, Chapter III reports a comparative LCA two recycling processes (i.e., pyrometallurgical and hydrometallurgical ones) in Italy focusing on different lithium-ion battery chemistries that are the best solution available nowadays. The main aim of this LCA study was to identify how the recycling processes contribute to reduce environmental burden related to lithium-ion batteries production and exploitation. Both processes shows results in the same order of magnitude in the overall LCA analysis. For the economic analysis Chapter III shows the results in terms of the plant cost and related economic indicators such as the Return on Investment (ROI), the Discounted Cash Flows (DCF) and the Net Present Value (NPV). The choice of the recycling process for lithium-ion batteries, indeed, in a cleaner production and sustainability perspective, should consider not only economical aspects but environmental performances of the whole system. The case study was set in Italy. Data for environmental analysis are obtained thanks to the technical design of battery recycling processes. The process is simulated using a spreadsheet in which adjustable parameters are plant capacity and battery chemistry. From an economic point of view the hydrometallurgical process performs better than the pyrometallurgical one.

Introduction

The human society is urgently in need for green electric energy. Mobile and stationary applications require advanced batteries capable of providing high power and energy density but fulfilling highest safety standards **(Hausbrand et al., 2015)**.
Lithium-ion cells were the best candidate for fulfilling these duties. Lithium-ion cells are electrochemical cells and consists of two electrodes, the anode (the negative electrode) and the cathode (the positive one) and an electrolyte. The two solid electrodes are kept apart from the liquid electrolyte by an electrolyte-permeable separator. This electrolyte conducts the ionic component of the chemical reaction between the anode and the cathode and forces the electronic components to flow through an external circuit **(Goodenough & Park, 2013)**. The material components constituting lithium-ion batteries (LIBs) obviously depends on chemistry and manufacturer. Nevertheless, considering an average of different LIBs chemistries available on the market, the cathode material represents about 50% of mass fraction, the anode about 30%, adhesives and plastics about 7%, the electrolyte about 4% and, finally, the remaining percentage represents other components **(Wang et al., 2014)**. The cathode generally consists of an active material.The most used material are lithium metal oxide, lithium-cobalt-oxide, LCO, lithium-manganese-oxide, LMO, lithium-nickel-manganese-oxide, LNMO, lithium-titanate-oxide, LTO, lithium-nickel-cobalt-manganese-oxide, NCM, lithium-iron-phosphate, LPF, lithium-nickel-cobalt-aluminium, NCA and lithium-vanadium-phosphate, LVP. Above materials are coupled with a 10 microns aluminium foil as current collector. The anode is generally made of graphite in which, recently, additional active components (i.e., SiO_x) are added to improve the capacity. The anode is also coupled with a 10 microns copper foil as current collector. Generally current collector and electrode are held together by PVDF-based adhesive.
The electrolyte consists of lithium salts (i.e., LIPF6, LiBF4, LiClO4) in organic solvents (i.e., propylene carbonate, PC, ethylene carbonate, EC, ethylmethyl carbonate, EMC, dimethyl carbonate, DMC, diethyl

carbonate, DEC) and acts as conductive pathways for li-ions movement. The separator is normally a microporous layer consisting of either polymeric or non-woven matrix, placed between cathode and anode to prevent their physical contact **(Zhu et al., 2021)**.
In a standard arrangement, li-ion battery operates according to the “rocking-chair” principle.
Li-ion are transferred between two materials that can intercalate them into non-occupied lattice site. During the charging phase of the cell the electron flux, coupled to the ion flow, is directed towards the anode while during the discharging phase it is directed to the positive electrode **(Zhao, 2017)**.

Lithium-ion cells are widely used in portable instruments, communication equipment and so forth while in the past few years they became pivotal in the transportation sector electrification process because of their high specific energy and high specific power and long life **(Deng et al., 2021)**. Moreover lithium-ion batteries show low tendency to self-discharge, it is interesting for the absence of heavy metals and for high operating temperature range as well as good longevity in terms number of recharge cycles **(Chandran et al., 2021)**.
Lithium-ion cells, assembled together in modules and battery packs, have been extensively applied in automotive sector for hybrid electric vehicles (HEVs), plug-in hybrid electric vehicles (PHEV) and battery electric vehicles (BEVs) **(Lin et al., 2015)** where the research for renewable energy use has been crucial over the last twenty years. Lithium-ion batteries - coupled with renewable electric energy production - granted the opportunity to contribute to reduction of the dependence on fossil fuels and greenhouse gases (GHGs) emissions.
The transportation sector, in fact, accounts for 23% of global CO_2 emissions related to energy activities (**Helmers et al., 2020**). On average, from 2015 to 2020, transportation-related emissions increased by 2.5% year-over-year (**Helmers et al., 2020**). The other sectors, however, have decreased over time (**Helmers et al., 2020**).
In the European Union (EU), the transport sector is responsible for about one-quarter of greenhouse gas (GHGs) emissions and the share is growing rapidly (Habib et al., 2015). Cars accounted for about 45% of these emissions in 2017 (**Andersson & Börjesson, 2021**). To achieve climate neutrality in 2050, a 90% reduction in transportation sector emissions will be required. The European Commission (EC) plans a

clear path, for cars and vans, to zero-emission mobility starting from 2025 **(Andersson & Börjesson, 2021)**.

The market of electric vehicles has grown exponentially since 2010 and many studies have been performed to evaluate the benefits of electrification of transportation in terms of energy and emission reduction (**Camus et al., 2011**).

Even though the adoption of EVs could have many benefits in terms of energy consumption and reducing local environmental impacts related to climate change depending on renewables share of energy generation mix, most studies doesn't consider other environmental impacts that could be negatively affected from EVs adoption. Moreover the transition from ICEVs to EVs will require a large production of LIBs and thus large quantities of metals such as lithium, cobalt, manganese and nickel (**Dunn et al., 2012**). The European commission forecasted that the demand for lithium would increase by 18 times in 2030 and by 60 times until 2050 compared to the current supply (**Keersemaker, 2020**). To avoid criticality in the lithium supply chain, a great effort should be made because of the low recovering rate of lithium in Europe (**Sun et al., 2019**). **Sonoc et al. (2015)** predicted that to ensure a stable supply chain of lithium in Europe, the minimum recovery rate should be 90%.

Waste electrical and electronic equipment (WEEEs) is one of the potential sources for valuable materials recycling. WEEEs sector is growing quickly and has a serious problem in terms of sustainability (**Menikpura et al., 2014**). At the same time, they have a high economic potential that comes from their high intrinsic value (**Sarath et al., 2015**), that imposes to study, first among all, the processes to make battery material recycle. However, in recent years only 20% of WEEEs is collected and recycled, while the rest is sent to thermal recovery or stored in landfills (**Baldè et al., 2017**).

One of the most important and used tools of Industrial Ecology is the Life Cycle Assessment (LCA) methodology, an environmental accounting and management approach for assessing industrial systems (**Curran, 2008**).

LCA allows to evaluate the environmental performances of alternative systems (products, processes, or services) looking at consumption of resources as well as the emission of pollutants that may occur during their life cycle from cradle to grave. For LIBS this approach includes the extraction of raw materials, the processing and production of

materials, the transportion, the phase of use and, finally, the end of life (**ISO 14040, 2006; ISO 14044, 2006**).
Since the term “life cycle assessment” was coined, in the early 90s, (**Bjørn et al., 2018a**) the methodology was applied to many industrial and economic sectors.

The overall scope of this Ph.D. work is the techno-economic and environmental analysis of recycling processes for LIBs from automotive sector. In order to achieve this overall goal, the first part of my Ph.D. work was focused on a critical review about the application of LCA methodology to the EVs and LIBs, in order to point out the most important results achieved until now as well as strengths and weaknesses of the assessment procedures adopted.

Therefore, the following research activities of this Ph.D. work focused on techno-economic and environmental assessment of the lithium-ion batteries.
In this regard, the specific objectives of these research activities were:
To design recycling process solution for lithium-ion batteries considering the processes family that are already available on pilot or industrial scales (pyrometallurgy and hydrometallurgy).
To perform an economic analysis of both processes to evaluate the most profitable one.
To perform a comparative LCA of recycling processes to assess the most environmentally sound.

This dissertation is organized as following:

- Chapter I describes the state of the art about the Life Cycle Assessment application to the lithium-ion batteries and electric vehicles through a critical review of papers published on the topic**.** Chapter I points out the criticalities of the LCA applied to the LIBs and to EVs. Furthermore, to highlights strengths and weaknesses of the assessment procedures adopted, the analysis of the reviewed studies was carried out also from a methodological point of view. From a practical perspective, some LCA results were analysed and discussed. From a methodological point of view, an in-depth analysis of the reviewed papers was carried out in order to highlight the different methodological approaches followed, as well as the key aspects of variability based on the LCA phases (Goal and scope definition; Inventory Analysis; Life cycle Impact Assessment; Interpretation).

- Chapter II deals with the technical design of battery recycling processes. The process is simulated using a spreadsheet in which adjustable parameters are plant capacity and battery chemistry. The main output of the model is the plant cost and related information (ROI, DCF) The main aim of this design phase was to obtain technical parameters that are going to be used for economic and environmental analysis. Chapter II also reports LCA results applied to battery recovery. In such a manner environmental and economical are evaluated in order to help in the selection of the recycling processes. About the economic evaluation, the analysis was performed based on two main indicators: the return of investment (ROI), return on investment, and the net present value (NPV) calculated according to chemical engineering standards. Sensitivity on plant capacity and LIBs chemistry was performed. The case study was set in Italy.
- Chapter III presents the assessment of the environmental performances of all the lithium-ion batteries chemistries and recycling processes considered. About the environmental evaluation, Chapter III reports a comparative LCA two recycling processes (i.e., pyrometallurgical and hydrometallurgical ones) in Italy focusing on different lithium-ion battery chemistries that are the best solution available nowadays. The choice of recycling process for lithium-ion batteries, indeed, in a cleaner production and sustainability perspective, should consider not only economical aspects but environmental performances of the whole system. The main aim of this LCA study was to identify the way recycling processes contribute to reduce environmental burden related to lithium-ion batteries production and exploitation. The case study was set in Italy.

Chapter I
State of the art of LCA application to li-ion batteries

I.1 Introduction

Stationary and mobile application needs advanced power storage devices. High safety standards and high energy and power density are required for the more recent mobile. The older batteries, on the contrary, that are characterized by long life and reliability are more suitable for energy stationary storage applications (**Hausbrand et al., 2015**). The lithium ion batteries (LIBs) are most promising candidates for both applications, thanks to their power and energy density as well as their low self-discharge rate (**Ioakimidis et al., 2019**).

During the past decade, lithium ion cells have been widely used in the automotive sector as energetic sources for electric vehicles (EV), hybrid electric vehicles (HEV) and plug-in hybrid electric vehicles (PHEV) **(Lin et al., 2015**). Total production LIB has been 103 GWh or 11,400 tons in the year 2017, and a quick increase in the future is expected. It is forecasted an annual demand for LIBs of 1,300 GWh (145,000 tons) in 2030 (**Curry, 2017**).

The annual demand for LIBs and for raw materials as its consequence, is related to the market penetration of EVs, batteries lifespan and recycling capacity. Therefore, considering the importance of electric mobility in economic and social terms, it is important to evaluate it from an environmental point of view.

In this context, the application of basic principles of industrial ecology and the use of its application tools can be a valid help for industries.

Industrial ecology (IE) can be seen as an emerging framework for environmental management, seeking transformation of the industrial system in order to match its inputs and outputs to planetary and local carrying capacity (**Lowe & Evans, 1995**). In the industrial ecology context, the Life Cycle Assessment (LCA) methodology can be used to evaluate the potential environmental impacts. The LCA method grants a complete vision of the life cycle of products, processes, or services during all the life cycle (Cradle to Grave approach) or only some parts of it (Cradle to Gate, Gate to Grave, etc.). The use of a ‘Cradle to Gate’ approach considers all processes from resource extraction to the factory gate. A ‘Cradle to Grave’ approach considers processes of Cradle to Gate approach to which are added use and disposal phases. The ‘Well to wheel’ approach is usually used for fuels. Inputs and outputs of an LCA study must be referred to a Functional Unit (F.U.), which is a measure of the function of the studied system. The International Standards Organization (ISO) provides the general guidelines to perform LCA in the ISO 14000 series **(International Standard Organization, 2006a, 2006b**).

The “Life cycle assessment” was coined, in the early 90s, **(Bjørn et al., 2018**) and starting from that period, the methodology was applied to many industrial and economic sectors.

Being an under-development sector that is undergoing an exponential growth, the electric mobility is starting to be assessed with this method too. Figure 1 shows the shares of batteries produced by countries in the world.

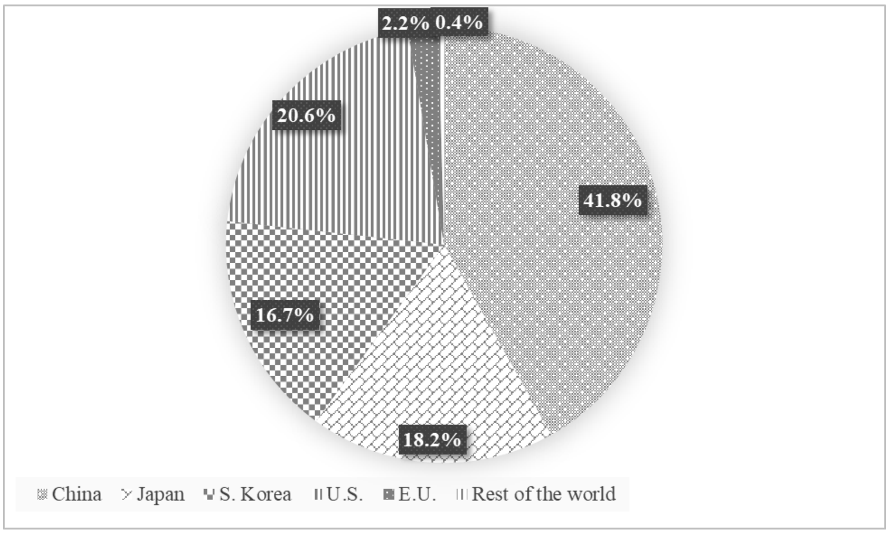


Figure 1: Shares of batteries produced by countries in the world (Mayyas et al., 2019)

Environmental evaluations are crucial to ensure that the battery recycling industries are not only economically but also environmentally sustainable.

Although is still in a developing phase, several studies have been carried out about the application of LCA to the electric mobility and lithium-ion batteries. In December 2020 there were 916,000 papers available on LIBs and the 2% was related to LCA or energetic and environmental analysis.

Therefore, studies published on the topic have been analysed in this Chapter in order to conduct a critical review useful to point out the most important results achieved until now as well as the main environmental hotspots of LCA application to electric mobility and LIBs sector. The analysis of the considered studies was carried out from a methodological point of view (concerning the application of LCA).

I.2 Methodological approach

LCA allows to compare different systems considering the consumption of resources as well as the emission of pollutants that may occur during their life cycle, which include the extraction of raw materials, the production and processing of materials, the transport, the phase of use and, finally, the end of life (**ISO 14040, 2006; ISO 14044, 2006**).

LCA is a good relation tool; this means that its purpose is the realization of comparisons between alternatives, and does not provide absolute evaluations (**Curran, 2008**). Therefore, it can be used to judge which products or systems are better for the environment or to point to the processes that contribute the most to the overall impact and should receive attention **(Bjørn et al., 2018**). LCA results are calculated by mapping all emissions and resources used and, if possible, the geographical locations of these, and use factors derived from mathematical cause/effect models to calculate potential impacts on the environment from these emissions and resource uses **(Bjørn et al., 2018**).

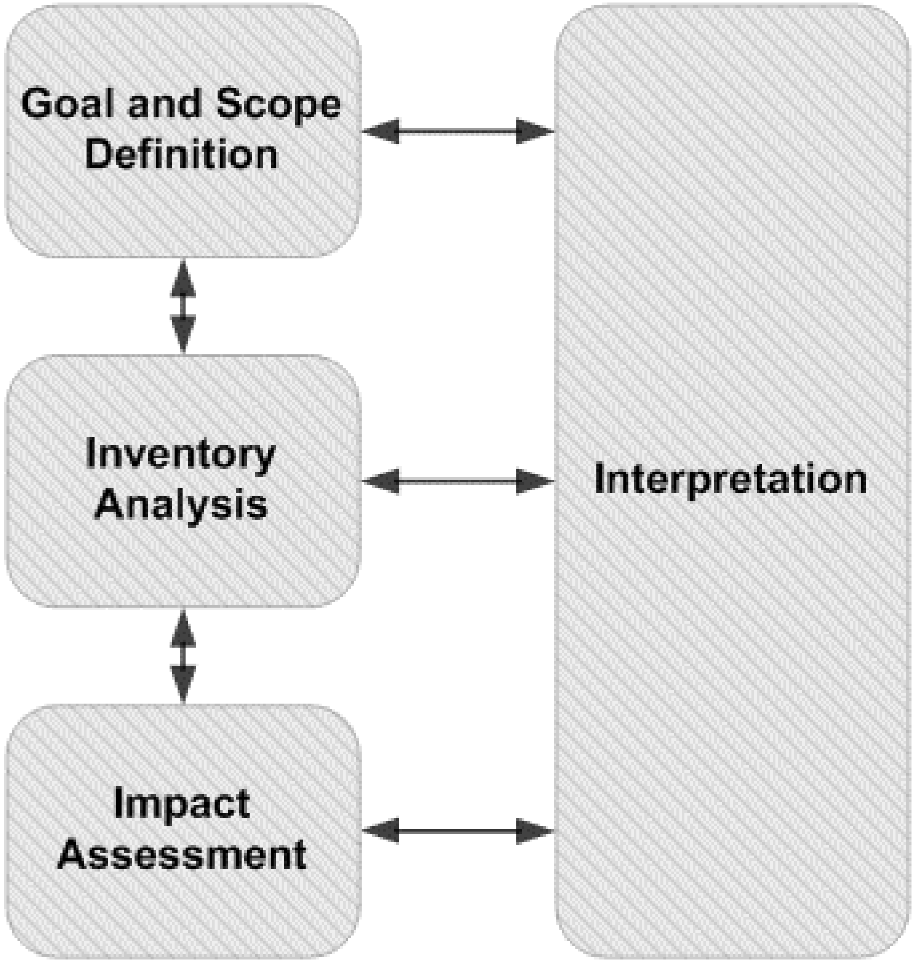


Figure 2: Life Cycle Assessment (International Standards Organization (2006) phases as described in ISO 14040.

Figure 2 shows the framework structure of an LCA study according to ISO 14040 standard composed of four main phases, and its different applications.

An LCA starts with the definition of the goal of the study. The goal definition sets the context of the LCA study and is the basis of the scope definition where the assessment is framed and outlined (**Hauschild, 2018**).

In the goal definition phase, it is necessary to define:

- what activities belong to the life cycle of product that is studied;
- the function unit: a quantitative measure of the function of the studied system;
- the geographical, temporal, and technological boundaries;
- the impact categories that shall be assessed in the study;
- a choice between consequential or attributional approach.

The phase of goal and scope definition is very important since it the choices that are made influence the data collection and the way in which of modelling and assessing the system. This phase, therefore, affects a lot the validity of the conclusions and recommendations that are based on the results of the LCA (**Hauschild, 2018**).

The goal and scope definition phase is followed by the inventory analysis. It is a process of in which energy and raw material requirements, pollutants in air, soil, and water as well as solid wastes production for the entire life cycle of the product system are quantified. The collected data are scaled in accordance with the reference flow and the functional unit.

A list of the material and energy flow is the result of inventory analysis phase. This list will be translated in impacts on the environment, during the impact assessment phase, using a characterization model (**Hauschild, 2018**).

Finally, according to ISO 14040:2006, the last phase of LCA is the the interpretation phase. This phase should include:

- the identification of significant issues based on the results of the inventory analysis and impact assessment phases;
- the evaluation of the study considering completeness, sensitivity, and consistency checks;
- the conclusions, limitations, and recommendations.

To guide the formulation of conclusions from the results sensitivity and uncertainty analysis are applied as part of the interpretation phase **(Hauschild, 2018**).

LCA methodology allows the comparison of environmental impacts of products, process and systems made up of hundreds of processes thanks to elementary processes contained in dataset accounting for thousands of resource uses and emissions that are taking place in different places at different times (**Bjørn et al., 2018**).

The broadness and comprehensiveness are the strength of the LCA methodology but, at the same time, it is also a limitation, as it requires simplifications and generalisations in the modelling of the product, process or service and the environmental impacts. It is more accurate to say that performing an LCA calculates *potential environmental impacts* **(Bjørn et al., 2018**).

I. 3 Practical aspects resulting from the LCA application to electric vehicles and lithium-ion batteries

Fifty-nine papers and studies on the topic, among the papers available in literature have been considered in this study. As a rule, papers with no original results as well as papers with no faithfully proved results have not been considered. The research of these papers has been conducted on the main available scientific databases (Scholar, Scopus, Science Direct, Web of Knowledge, etc.) using different key words such as LCA, life cycle assessment, electric vehicle, battery, lithium-ion battery, hydrometallurgy, pyrometallurgy, recycling, second life, end of life, etc. as well as combining them with the ‘and’ boolean operator.

The main characteristics of the studies considered in this review are reported in Table I. In the Table I the analysed papers are divided into three categories: vehicle studies, battery studies and other studies.

Table I: Main characteristics of the studies considered

LCA APPROACH	FUNCTIONAL UNIT (F.U.)	MAIN TOPIC	LOCATION	REFERENCES
		BATTERY		
CRADLE TO CRADLE	1 kWh	LCA analysis on recycling and reuse of lead acid, LIBs, and vanadium redox flow batteries	Germany	(Unterreiner et al., 2016)
CRADLE TO GATE	1 kg of cathode	Different impact assessment methods comparison	-	(L. Wang et al., 2019)
	1 kg of raw material	Real energy demand and GHGs emission comparison to GREET	China	(Yin et al., 2019)
	1 kWh	LCA on NMC battery	US	(Dai et al., 2019)
	100 kg of batteries	Production environmental impact of different batteries (LIBs and non-LIBs)	UK	(Mcmanus, 2012)
	1000 kWh	LCA comparison among LIBs, NMHs and solar cells	China	(Liang et al., 2016)
	Local energy demand [MWh]	Optimized consequential LCA for ESS application	FR	(Elzein et al., 2019)
CRADLE TO GATE + END OF LIFE	17 kWh	LCA on innovative LIB chemistry	-	(Raugei & Winfield, 2018)

CRADLE TO GATE, CRADLE TO GRAVE	1 battery pack (250 kg)	LCA approach and EIO-LCA	UK	(S. Zhao & You, 2019)
CRADLE TO GRAVE	1 battery	Complete LCI for NCM battery	-	(Ellingsen et al., 2014)
	1 battery pack (346 kg)	LCA on LithoRec project for LIBs recycling	Germany	(Cerdas et al., 2018)
	1 battery pack (43,2 kWh)	LCA on battery with silicon nanowire anode	-	(Li et al., 2014)
	1 battery pack (43,75 mAh)	LCA on SSBs manufacturing	Germany	(Troy et al., 2016)
	1 kWh	LCA on different battery types	-	(Hammond & Hazeldine, 2015)
		LCA on battery reused for ESS application	Canada	(Ahmadi et al., 2015)
		LCA on new advanced material for LIBs	EU	(Kushnir & Sandén, 2011)
		Environmental burdens of used batteries	JP	(Ishihara et al., 2002)
	1 MWh	LCA comparison between LIBs and LMPs	Canada (Quebec)	(Vandepaer et al., 2017)
	50 kW power / 450 kWh capacity	Energy analysis on eight battery technologies	-	(Rydh & Sandén, 2005)

	Average yearly energy balance	LCA of repurposed EV batteries in ESS applications	Netherlands	(Bobba et al., 2018)
END OF LIFE (RECYCLING PHASE)	1 ton of batteries	LIBS recycling processes investigation and LCA	Australia	(Boyden et al., 2016)
WELL TO WHEEL	-	Different battery chemistries assessment	EU	(Gerssen-Gondelach, 2012)
	1 km	LCA study	Sweden	(Zackrisson, 2016)
VEHICLE				
CRADLE TO GATE	-	Energy consumption and GHGs emissions evaluation of ICEVs and BEVs production	China	(Qiao et al., 2017)
	100,000, 150,000 and 200,000 km	Impact of driving patterns, geographic locations, and heating/cooling use on energy consumption of EV. LCA approach	Germany, Brazil and Spain	(Egede et al., 2015)
	150,000 km	LCA study on BEV and ICEV	EU	(Pero et al., 2018)
CRADLE TO GRAVE	-	LCA on BEV, HEV and PHEV with LMO batteries	US	(Dunn et al., 2012)
		LCA study for different LIB chemistry in PHEV	US	(Gaines et al., 2011)

		Energy use and GHGs emission of vehicle	US	(Sullivan et al., 2010)
	1 km	LCA on five different powertrain scenarios	Brazil	(Souza et al., 2018)
		LCA comparison between ICEVs and BEVs	EU	(Tagliaferri et al., 2016)
		LCA on different EV types and LIB chemistries	US	(Ambrose & Kendall, 2016)
	1 kWh	Different impact methods sensitivity on electric powertrains LCA	EU	(Hernandez et al., 2017)
	1 mile	LCA analysis on different advanced powertrains	US	(Mayyas et al., 2019)
	100 km	Carbon and water footprint analysis	Czech	(Jursova et al., 2019)
	120,000 km	Batteries production impact on EV life cycle	US	(Lastoskie & Dai, 2015)
	150,000 km	LCA with present and future energy mixes	Czech and Poland	(Burchart-Korol et al., 2018)
	160,000 km	LCA on electric vehicle and LIBs second life scenarios	Canada	(Ahmadi et al., 2014)
	200,000 km	Comparative LCA on two batteries type: LFP and LMO	China, Germany, France, Portugal	(Marques et al., 2019)

	200,000 km	CO_2 emissions comparison between BEVs and ICEVs	US, EU, JP, China, Australia	(Kawamoto et al., 2019)
	4,000 days	Second life scenarios LCA	Spain	(Ioakimidis et al., 2019)
GATE TO GATE	1 km	Impact of different vehicle technologies on online shopping using LCA	Thailand	(Koiwanit & Hamontree, 2018)
WELL TO WHEEL	-	LCA study on different size segments EVs	EU	(Ellingsen et al., 2016)
	-	Environmental profile for EV	Germany	(Held et al., 2011)
	1 kg of battery	Exergetic efficiency analysis on thermal management system for EV an ICEV	EU	(Hamut et al., 2014)
		LCA on ICEV and BEV	-	(Frischknech, 2011)
		ICEVs and BEVs environmental comparison using LCA	Switzerland	(Notter et al., 2010)
	1 km	LCA comparison between EVs and ICEVs	EU	(Hawkins et al., 2013)
		Evaluation of GHGs emission for PHEV	US	(Samaras & Meisterling, 2008)
	10 kWh	LCA on LFP batteries in PHEV application	EU	(Zackrisson et al., 2010)

	180,000 km	LCA on electric, hybrid and fuel cell vehicles	EU	(Van den Bossche et al., 2006)
	180,000 miles	Energy inputs and GHGs emission for ICEVs, HEVs and BEVs	US (California)	(Aguirre et al., n.d.)
	200,000 km	LCA on automotive and second life for LIBs and sensitivity on energy mixes.	Portugal, France and Poland	(Faria et al., 2014)
	230c,500 km	Environmental impact of conventional and electric vehicles.	-	(Van Mierlo et al., 2011)
	50 MJ (equivalent to 100 km)	Environmental comparison between LIBs and NMHs in EV applications.	EU	(Majeau-Bettez et al., 2011)
		OTHERS		
CRADLE TO GRAVE	kg of batteries	Material and energy flows for different LIB chemistries.	US	(J. B. Dunn et al., 2014)
	20 kWh	Material and energy flow for NMC production	-	(Simon & Weil, 2013)
	-	Energy and mass flow	EU	(Hischier et al., 2007)
	-	Cost analysis for LIBs	US	(Gaines & Cuenca, 2000)

Seventeen percent of the reviewed studies considered a Cradle to Gate approach. For example, **Liang et al. (2016)** focused on the evaluation of emissions related to the batteries production. The last 7% is made of different approaches such as Gate to Gate, Cradle to Cradle and End of Life only. The impact categories of all the studies in this review have been collected and are shown in Figure 3.

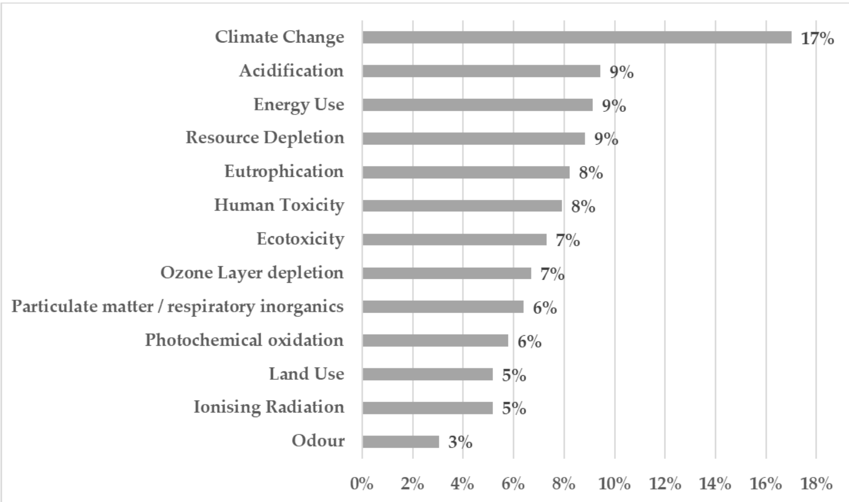


Figure 3:Most used impact categories.

Climate change and energy use are the most common impact categories in the LCA studies taken into consideration. They are useful to evaluate technical and environmental performances of products and processes. The climate change category is related to the Greenhouses Gas Emissions (GHGs), whilst the energy use category gives an indication of the harvested energy across the life cycle of the process considered. There is a strong relevance between climate change impact category and energy use; therefore, it is possible to speculate that energy intensive processes are likely to be more impacting from a climate change perspective (**Yin et al., 2019**) and the magnitude of this correlation is related to the energy mix composition selected to perform the study. The more the energy mix relates to non-renewable sources, the higher is supposed to be the climate change category.

The LCA studies on LIBs appeared in the early 2000s and their number increased over time. Among the considered studies, 38% is an LCA study on lithium-ion battery, while 55% is an assessment on the electric vehicle powered by a lithium-ion battery pack. The first noticeable fact that appears from the studies considered is the geographic location in which the LCA is performed. As can be seen in Figure 4, more than half (57%) of the studies are conducted considering a European energy mix, 21% of the studies considered a US energy mix and about 6% used a Chinese energy mix. Other small contributions come from Canada, Brazil, Japan, and Australia.

Figure 4: Geographic location considered in the LCA studies.

A second important aspect concerns the cathode chemistry of the lithium-ion batteries. The LIBs are classified on the base of chemical composition: lithium iron phosphate (LFP), lithium nickel cobalt manganese (NCM), lithium manganese oxide (LMO), lithium cobalt oxide (LCO), lithium nickel cobalt aluminum (NCA); lithium cobalt phosphate) (LCP); lithium iron manganese phosphate (LFMP); lithium metal polymer (LMP); lithium cobalt nickel (LCN). In about 6% of the papers, the chemistry of the battery is not specified. The range of chemistry considered in the remaining papers is shown in Figure 5.

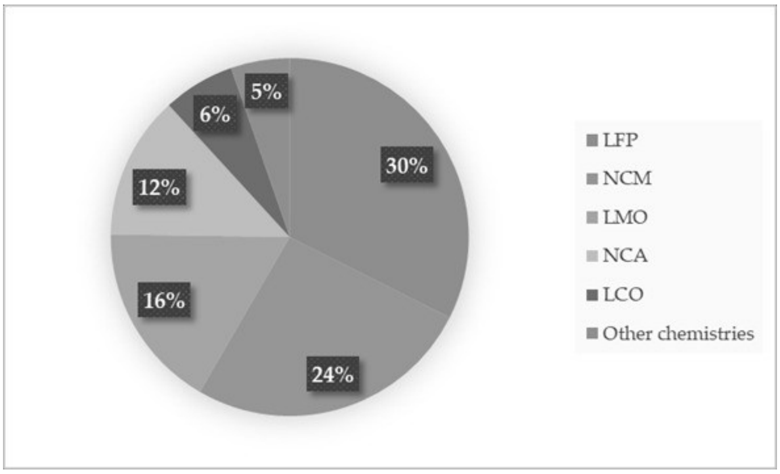


Figure 5: Considered LIBs chemistries: LFP (lithium iron phosphate); NCM (lithium nickel cobalt manganese); LMO (lithium manganese oxide); LCO (lithium cobalt oxide); NCA (lithium nickel cobalt aluminum); LCP (lithium cobalt phosphate); LFMP (lithium iron manganese phosphate); LMP (lithium metal polymer); LCN (lithium cobalt nickel).

Lithium iron phosphate (LFP) and nickel cobalt manganese (NCM) are the most analyzed chemistries. LFP batteries are made of cheap, nontoxic and easily accessible materials like iron and phosphorus (**Wang et al., 2019**). The cobalt based chemistry batteries show high environmental impacts to human and ecosystems because of the metal extraction process (**Kallitsis et al., 2020**). Furthermore, LFP batteries are safer than cobalt based batteries **(Sun et al., 2019**). For these reasons, LFPs are the best candidates to be used for next generation green LIBs. NCM is the most diffuse cathode chemistry in the EVs sold in Europe, Japan and United States (**Dai et al., 2019**).

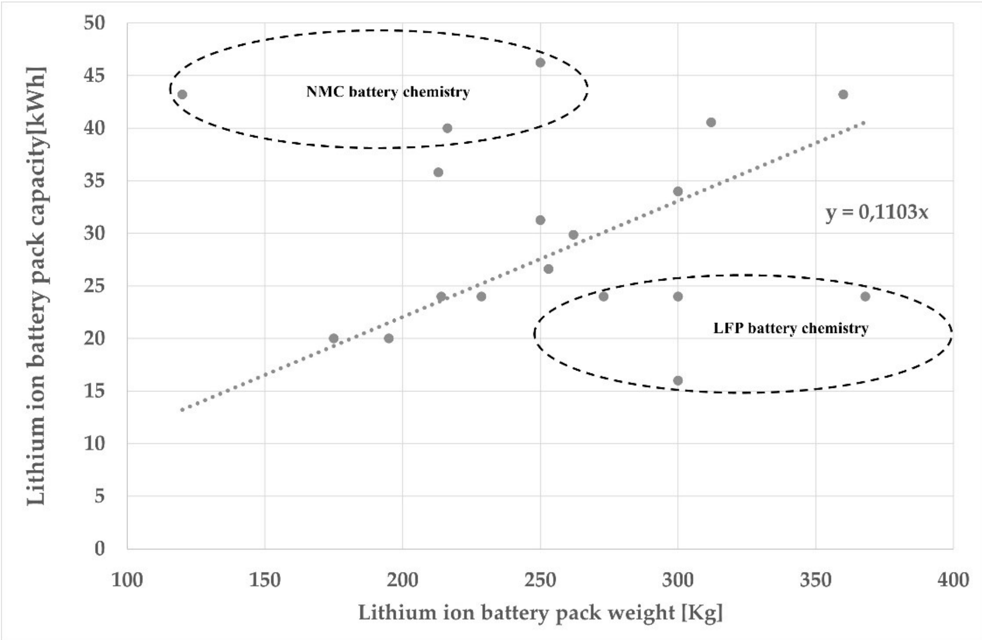


Figure 6: Lithium-ion battery pack capacity versus the battery pack weight.

Figure 6 show the LIB and its capacity as a function of their weight. On the above data it is possible to assess an average value of the ratio capacity/weight [kWh/kg] equal to 0.1103. The inverse value is equal to 9.066 kg/kWh and gives an indication of the average energy density. The batteries with NCM and LMO cathode chemistry have values of the weight/capacity ratio lower than the average. Other chemistries, such as LFP, have higher values than the average one, and the reason for this is related to the different energy densities among different chemistries. It is possible to state that, from a size point of view, batteries with a lower value of weight/capacity are preferable for automotive applications. Lighter batteries are less impacting on vehicle electrical consumption.

I. 4 Methodological aspects resulting from the LCA application to electric vehicles and lithium-ion batteries

In this section, the reviewed studies are presented from a methodological point of view. The approaches adopted in the LCA main phases (Goal and scope definition, Inventory Analysis, Life Cycle Impact Assessment, Interpretation) are going to be underlined

The most common Functional Unit in the cited literature (33.90% of the studies) is the distance travelled by the vehicles. It is a common choice when comparing the environmental behaviour of internal combustion engine vehicle and electric vehicle) (**Tagliaferri et al., 2016**). The second most common FU (30.51%) is the total amount of energy provided to the vehicle by the batteries. It is a correct choice when it is necessary to consider the influence of parameters such as lifetime, efficiency and Depth of Discharge (DOD) on the output delivered by the batteries (**Vandepaer et al., 2017**). The third most common FU (16.95%) is the battery pack mass. This type of FU is commonly used when it is necessary to compare different cathode materials (**Wang et al., 2019**) or when the work mainly relies on LIBs production phases focusing on raw materials acquisition, transportation, production (**Yin et al., 2019**) and EoL recycling phases.

There are other possibilities for choosing a FU to perform an LCA study. Ioakimidis et al. (2019), for example, have chosen an amount of time (4,000 days) as FU. In their work, they considered a second life repurposing for batteries as energy storage units in building. They were able to compare the two scenarios (base case and repurposing scenario) on a time basis. All the FU definitions are valid because the choice depends on which is the focus of the analysis.

The inventory analysis phase (LCI) is one of the most crucial in performing an LCA study. The study performed must afford the problems of data quality and their availability. The primary data, collected from stakeholder of lithium-ion batteries supply chain, can be subject to non-disclosure agreements and cannot be easily accessed. The recycling of lithium-ion batteries is a novel problem, and it is likely to experience lack of data. Another way to obtain primary data is to perform experimental procedures. Databases on which LCA software tools are built can be considered a sort of benchmark for data. These are the so-called secondary data. It would be preferable to perform LCA studies using inventories only made of primary data because they are more reliable since their operational origin. Secondary data present a valid substitute if based on robust models and assumptions.

Among the reviewed papers, 83% used a mix of primary and secondary data, 12% used only primary data and 5% only secondary data, as shown in Figure 9.

The most cited articles are Majeau-Bettez et al. (2011) (in 19% of the papers), Hischier et al. (2007) (in 13% of the papers) and Zackrisson et al., (2010) (in 10% of the papers).

Around 41% of the reviewed papers (i.e., 24) do not precisely state which database is used to perform the LCI phase. Among the remaining ones, Ecoinvent is chosen in 66% of the studies, while BatPac is preferred in 17% of the studies. Ecoinvent is developed by the Swiss Centre for Life Cycle Inventories. It contains around 17,000 LCI datasets in many areas such energy supply (*Swiss Centre for Life Cycle Inventories. Ecoinvent 2017—Ecoinvent Database v3.6, 2019*).

Finally, another important aspect that can be underlined is related to the software tool used to perform the LCA. In 49% of the reviewed studies, it is not specified. In these cases, it would be useful to know why it was chosen not to use any software and what calculations were made. Among the remaining 51%, SimaPro is used in 38% of the cases, GREET in 31%, GaBi in 25%, and OpenLCA in 6%.

In the Life cycle impact assessment phase (LCIA), the potential environmental impacts are calculated in relation to the LCI phase results. In the Life cycle impact assessment phase (LCIA), the potential environmental impacts are calculated in relation to the LCI phase results.

In 23% of the reviewed papers, it is not clearly expressed which LCIA method is used. Other than these studies, the most used LCIA method is ReCiPe (19%) developed by PRè Sustainability in collaboration with Dutch National Institute for Public Health and the Environment (RIVM), Radboud University Nijmegen and Norwegian University of Science and Technology (*ReCiPe | PRé Sustainability*). The ReCiPe method can determine environmental impacts category on two levels: Midpoint and Endpoint. Midpoints indicators focus on an environmental problem while Endpoint show the impact from a higher aggregation level. The passage from Midpoint to Endpoint simplifies the LCIA phase but the uncertainty increases due to the aggregation process. Other LCIA methods used are IPCC (9%) and CML-IA (9%).

Among the reviewed studies, 22.8% consider only one impact category (i.e., global warming, potential or cumulative energy demand, CED, etc.). The remaining 77.2% consider more than one impact category.

The representation used to show the statistical data is the named block box, that provides a synthetic description of a data distribution and are based on 5 numbers: minimum, first quartile (q1), median, third quartile (q3) and maximum. The light grey rectangle represents the distance between q3 and median, while the dark grey rectangle is the distance between q1 and median. The interquartile range (q3–q1) is a measure of the distribution dispersion. 50% of the observations lie between these two values. The bars ("whiskers") above and below the box show the locations of the minimum and maximum. The lengths of the two bars and the heights of the two rectangles provide information on the symmetry of the distribution: this is more symmetrical as the lengths of the bars are like each other and the heights of the two rectangles are similar to each other.

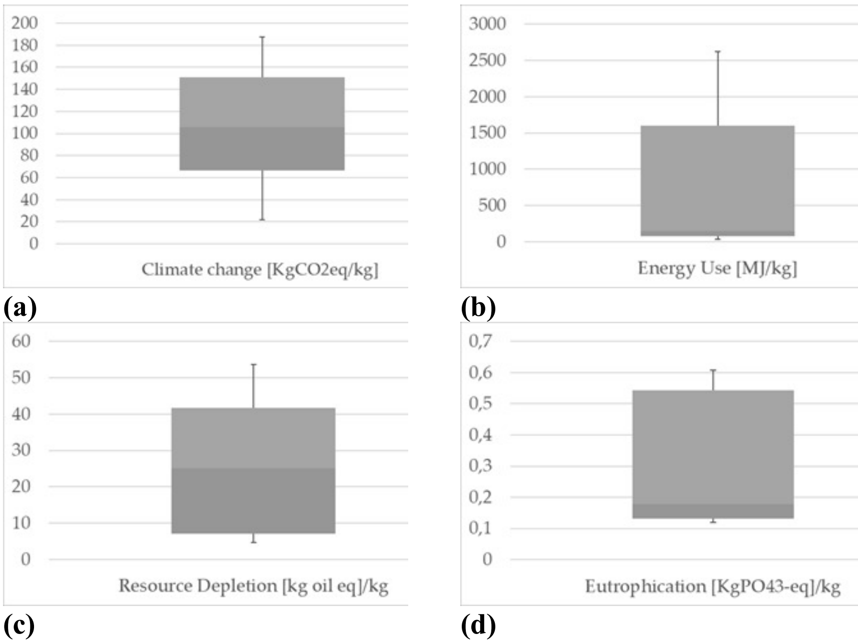


Figure 7: Some results derived from papers in terms of impact categories per kg of LIBs: (a) Climate change value, , per kg of batteries; (b); Energy use, per kg of batteries; (c) resource depletion, per kg of batteries; (d) Eutrophication, per kg of batteries.

The types of graphs in Figure 7 are box plots.

The process of data obtained from the reviewed papers was not straightforward and the value gained shows a lot of variability. This variability can be related to the different system boundaries among the different studies and to the lack of precise numerical data.

The interpretation phase is the last phase of an LCA study. Usually, it includes a sensitivity analysis and a discussion on the results reliability. Most of the reviewed papers did not provide the numerical values of the environmental results. This makes the results comparison difficult to perform. When hypotheses are made, it is important to check the influence of the input parameters on the obtained results. To quantify this influence, a sensitivity analysis is conducted. As matter of fact, it is noticeable that in 51% of the reviewed studies a sensitivity analysis is performed while in the remaining 49% it is not.

From a practical point of view, the**Errore. L'origine riferimento non è stata trovata.** Table II shows the main outcomes of this study.

Table II: main outcomes of the literature review

LITERATURE ANALYSIS HIGHLIGHTS	
1	No environmental impacts analysis in countries where batteries manufacturing takes place
2	Increasing awareness of lithium-ion batteries environmental burdens
3	Usually the environmental assessment of lithium-ion batteries is performed looking at climate change impact category
4	Distance travelled by electric vehicles, mass or capacity of the battery pack are the most used FU
5	Lack of primary data
6	Many of the LCA study are not showing numerical results and this make it difficult to perform a complete and reliable meta analysis

More than half of the studies considered a European energy mix. Other studies are necessary to quantify the LIBs environmental impacts in countries in which battery production takes place. Lithium iron phosphate (LFP) and nickel cobalt manganese (NCM) are the most analysed chemistries for electric batteries, but it is likely to happen that in the future different chemistries will be analysed as long as they become more widespread. In fact, as emerged from the analysis, batteries with a lower value of weight/capacity are preferable for automotive applications, while lighter batteries are less impacting on vehicle electrical consumption. New chemistries are necessary to meet these requirements. The Cradle to Grave approach was adopted in around 50% of the case studies, while climate change and energy use were the most used impact categories. The performed review emphasizes the potentiality of EVs and LIBs to reduce the overall contribution of the transportation sector to the GHGs emissions.
From a methodological point of view, the followings are the main outcomes of the review. In the Goal and scope definition, the distance travelled by the vehicles is the most used FU because the studies focused mainly on comparing the different vehicles. In the Inventory Analysis, Ecoinvent is the preferred database, while SimaPro is the preferred software tool. The lack of primary data is a crucial concern. It is likely that the more the EVs will be widespread

the more data will be accessible. In the Life Cycle Impact Assessment, ReCiPe is the most used method. In the Interpretation, many articles do not provide all the numerical values, thus not allowing an easy comparison of the environmental results.

Chapter II
Comparative LCA between BEV and ICEV in Italy

II.1 Introduction

Climate change is more than ever a guiding factor on several aspects of society these days. People are becoming more conscious of the real impacts of this change and want to play an active role in mitigating this issue. Seeking more energy efficient and environmentally friendly products can also result in lower energy bills (**Faria et al., 2013**).

The transportation sector accounts for 23% of global CO_2 emissions related to energy activities. On average, from 2015 to 2020, transportation-related emissions increased by 2.5% year-over-year. While, the other sectors have decreased over time (**Helmers et al., 2020**).

In the European Union (EU), the transport sector is responsible for about one-quarter of greenhouse gas (GHG) emissions and the share is growing rapidly (**Faria et al., 2012; Habib et al., 2015**). Road transportation accounted for about 71% of these emissions in 2018 (*Greenhouse Gas Emissions from Transport in Europe — European Environment Agency*)

To achieve climate neutrality in 2050, a 90% reduction in transportation sector emissions will be required. The European Commission (EC) plans a clear path, for cars and vans, toward a zero-emission mobility starting from 2025 (**Andersson & Börjesson, 2021**).

In recent decades, energy uses for electricity generation and transportation have more than doubled and now face several challenges related to reliability, safety, and environmental sustainability. Scientific evidence on climate change has called for urgent cross-sector emission reductions **(Camus et al., 2011**).

II.1.1 Electric vehicles

The categories of electric vehicles that are widely commercialized are as follows: Battery Electric Vehicles (BEVs), Hybrid Electric Vehicles (HEVs) and Plug-in Hybrid Electric Vehicles (PHEVs). Electric vehicles use electric motors driven by rechargeable batteries and inverters (**Rezaee et al., 2013**). These technological solutions have advantages over conventional ones such as being noiseless, requiring less maintenance, and not emitting harmful substances (**Ma et al., 2012**). Electric vehicles can be powered by battery packs, some acquire power from a combustion engine (ICE), while some employ both (**Un-Noor et al., 2017**).

Battery electric vehicles (BEVs) are one of the most promising vehicle types for reducing greenhouse gases (GHGs) emissions in the transportation sector. Only recently, with the development of battery technology, electric cars have entered the scene again and many studies have been done to evaluate the benefits of electrification of transportation in terms of energy and emissions reduction (**Wang et al., 2020**). In fact, BEVs are particularly beneficial because they are highly efficient and do not produce local combustion-related emissions. However, electricity, which is the fuel for BEVs, is generated from various primary energy sources (coal, natural gas, crude oil, uranium, etc.), and thus, driving BEVs still causes emissions related to the conversion of primary energy to electricity (**Choi & Song, 2018**). Series or parallel HEVs are powered by an electric motor and an ICE, both of which are connected to the wheels via a torque coupling and complement each other when needed.

Series or parallel PHEVs have a small battery that provides a range of between 30 and 80 km (**Onwunta, 2021**). When the battery charge is depleted, the propulsion system uses an ICE motor to recharge it and extend the range (**Tran et al., 2021**). PHEVs are an evolution of HEVs and can be connected to the grid to recharge the batteries, increasing the overall energy efficiency. BEVs are powered only by an electric motor and a battery pack, providing an autonomy range of up to 450 km (**Sagaria et al., 2021**).

Numerous studies have demonstrated the potential environmental benefits of electric vehicles. For instance, the potential reduction in GHGs through the introduction of PHEVs in the U.S. was already in 2011 was evaluated of 42% per mile driven on the average (**Brady & O'Mahony, 2011**) More recently Burchart-Korol et al. (2018) showed that in terms of GHGs emissions and fossil resources depletion, electric vehicles perform better than the thermic counterpart. In addition, Onat et al. (2018) showed that when the energy mix used to recharge the batteries is highly composed of renewable energy, the environmental impact of the electric vehicle are better performing with respect to thermic vehicles in all the environmental impact categories.

The European Commission is currently working on proposals regarding electric vehicles, which are described as a "very important" part of its green strategy, and the European Parliament has launched a resolution supporting development and innovation on this issue (**Baptista et al., 2013**). The European Union aims to halve vehicles with combustion engines by 2030 and phase them out in cities by 2050. Sales of electric vehicles are increasing worldwide, with China and Norway leading the technology changes main drivers. Over the next few years, electric vehicles are expected to increase tremendously to reach about 18 million in 2025, and 21 million in 2030 (**Dolganova et al., 2020**).

II.2 Literature review

A sample of 35 papers published between 2009 and 2021 has been considered and among them only 19 are LCA studies with well-to-wheel (WTW) or cradle-to-grave (CTG) approaches and only 9 papers of them presented a multicategory LCA analysis. The other 10 papers analysed only the Climate Change impact category which tends to favour the results of analysis of the electric vehicles.

Petrauskienė et al. (2020) stated in their work that the climate change category must not be the only one to be considered when assessing the environmental sustainability of electrical and internal combustion engines vehicles (**Petrauskienė et al., 2020**). In fact, when two or more categories are considered, there is not a precise winner in the comparison between ICEVs and BEVs. The parameters influencing the preferable vehicle category are the energy mix, the driver behaviour, the driving cycle, and the vehicle segment.

influences the BEVs emissions (**Weldon et al., 2016**).

Over the 60% of the analysed studies states that the energy mix is the most important variable that influence the results of the LCA. For example, Faria et al., 2013 highlighted that the fundamental variable to be considered for determining the environmental performance is the national energy mix **(Faria et al., 2013**) and the study confirmed that BEVs can be compared with ICEVs when the energy mix is based mainly on solar energy. Choma & Ugaya (2017) as well as Rupp et al. (2019) stated that to compare BEVs and ICEVs the functional unit to be chosen is the “transportation of a passenger on a certain distance”. Hofmann et al. (2016) highlighted that the mere introduction of electric vehicles in the transportation sector do not produce positive results reducing GHGs emissions if the energy mix is not sufficiently decarbonized. Woo et al. (2017) analysed over 70 countries showing that BEVs emissions can be higher than ICEVs emissions when the energy mix is based on fossil fuels energy generation systems. These results are also confirmed by Burchart-Korol et al. (2018) and Wilken et al. (2020). In any case, the results of comparison are not far. Weldon et al. (2016) in their paper demonstrated in fact that the driving style of the driver influences the BEVs emissions.

Girardi et al. (2015) showed that BEVs have better environmental profile than ICEVs in some of impact categories regarding “in air emissions” as acidification, particulate matter formation potential and photochemical oxidant formation because they do not have direct emissions deriving from combustion. Pero et al. (2018) proposed a study in which the complete life cycle of vehicles was analysed. In their study, BEVs were better than ICEVs only in climate change category while ICEVs performed better in acidification, human toxicity, particulate matter, photochemical ozone formation and resource depletion.

On the base of the above literature, it is possible to conclude that BEVs are a valid alternative to ICEVs to reach the goal of achieving a sustainable mobility with respect the climate change. Nevertheless, to assess the effective environmental sustainability it is important to include in the input data set for analysis also other parameters.

II.2 Methodology

II.2.1 Life Cycle Assessment

As stated before, LCA is a standardized methodology that allows to evaluate potential environmental impacts of products, processes, or services. To perform an LCA, it is necessary to follow the framework of ISO 14040 and ISO 14044.

According to the literature, SimaPro and GREET are the software tools most used (60% of the studies) in comparative LCA analysis between BEVs and ICEVs. Among the reviewed papers, 21% used the ReCiPe 2016 method in the Life Cycle Impact Assessment phase (LCIA).

In this study the life cycle impacts of the vehicles are estimated using the ReCiPe 2016 evaluation method with the hierarchist perspective (H) (**Huijbregts et al., 2017**).

This method allows for the consideration of two evaluation approaches: a problem-oriented approach (midpoint level) that contains 18 impact categories and a damage-oriented approach (endpoint level) that considers 3 damage macro-categories (**Huijbregts et al., 2017**). The midpoint impact categories are the following: Global Warming Potential (GWP); Stratospheric Ozone Depletion (SOD); Ionizing Radiation (IR); Ozone Formation, Human Health (OF-HH); Fine Particulate Matter Formation (FPMF); Ozone Formation Terrestrial Ecosystems (OF-TE); Terrestrial Acidification (TA); Freshwater Eutrophication (FE); Marine Eutrophication (ME); Terrestrial Ecotoxicity (TEcotox); Freshwater Ecotoxicity (FEcotox); Marine Ecotoxicity (MEcotox); Human Carcinogenic Toxicity (HCTox); Human non-Carcinogenic Toxicity (HnCTox); Land Use (LU); Mineral Resource Scarcity (MRS); Fossil Resource Scarcity (FRS); Water Consumption (WC). The three endpoint damage macro-categories are Human health, Ecosystems, and Resources (**Huijbregts et al., 2017**). Both approaches are used in the study.

The last phase is the Life Cycle Interpretation in which conclusions and recommendations are produced based on the obtained results.

II.2.2 Software tools

The two software tools used in this work are SimaPro and GREET.

SimaPro is a professional software tool released in 1990 by PRè Sustainability based in Netherlands which allow to collect, analyse, and model the sustainability performance of product and service. It is a generalist software suitable for modelling every type of system. The software version used in this study is the SimaPro 9.1.1.

The GREET software tool (Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model) made by Argonne is specific for the transportation sector. It permits to model different vehicles and fuels. It is largely used because it is sponsored by the US Department of Energy (**Anderson et al., 2018**). GREET 2020 is the software version used in this study.

II.2.2.1 Assumptions and inventories

In this study, ten BEVs and ten ICEVs were modelled for different market car segments. According to the European Commission, the main categories of vehicles are M (vehicles carrying passengers), N (vehicle carrying goods), L (2- and 3-wheel vehicles and quadricycles), T (agricultural and forestry tractors and their trailers). Vehicles that belong to M or N categories are classified as light-duty vehicles (passenger cars and vans) or heavy-duty vehicles (trucks, buses and coaches) (**European Commission, 2016**).

According to the UNECE standard, vehicles carrying passengers are M category vehicles. The M category is divided in nine categories named A, B, C, D, E, F, S, J and M. The differences between vehicles in these categories are typically in term of power and weight (*Classification and Definition of Vehicles | UNECE*).

The electric vehicle data were obtained from the EV database (ev-database.org). The internal combustion engine vehicle data are obtained from the auto-data database, which is a daily updated car specifications database with more than 41,000 automobiles (www.auto-data.net).

Table III and Table IV show the electric and thermic vehicles considered and their main parameters. Where possible, the same vehicle with different powertrain system have been selected and chosen. Where some correspondence was not found, a vehicle with similar characteristics has been selected to obtain the comparison.

Table III: Selected BEV models and their main characteristics

SEGMENT	VEHICLE NAME	POWER (HP)	WEIGHT (KG)	BATTERY CAPACITY (KWH)
A	2014 Smart Fortwo III cabrio Brabus 17.6 kWh (82 CV) electric drive	82	945	17.6
A	2020 Renault Twingo III (facelift 2019) Z.E.	81	1208	22
B	2020 Opel Mokka-e B	136	1523	50
B	2019 Peugeot 208 II e-208	136	1530	50
C	2017 Volkswagen Golf VII (facelift 2017) e-Golf	136	1540	35.8
C	2019 Nissan Leaf II (ZE1) e+	218	1756	62
D	2020 BMW iX3 (G08) 80 kWh (286 CV)	286	2185	80
E	2018 Audi E-tron Quattro	408	2400	95
L	2020 Porsche Taycan Turbo	680	2305	93.4
N	Opel Zafira-e Life L	136	2167	75

Table IV: Selected ICEV models and their main characteristics

SEGMENT	VEHICLE NAME	POWER (HP)	WEIGHT (KG)	FUEL
A	2014 Smart Fortwo III cabrio 0.9	90	920	Petrol
A	2019 Renault Twingo III (facelift 2019) 0.9 TCe EDC	92	999	Petrol
B	2020 Opel Mokka B 1.2 Turbo	130	1200	Petrol
B	2019 Peugeot 208 II 1.2 PureTech Stop&Start Automatic	130	1158	Petrol

C	2017 Volkswagen Golf VII (facelift 2017) 1.4 TSI	147	1336	Petrol
C	2015 Nissan Pulsar (C13) 1.6 DIG-T	190	1363	Petrol
D	2017 BMW X3 (G01) 30i xDrive Steptronic	252	1790	Petrol
E	2019 Audi Q5 II 50 TDI quattro Tiptronic	286	1860	Diesel
L	2014 Smart Fortwo III cabrio 0.9	630	2185	Petrol
N	2019 Renault Twingo III (facelift 2019) 0.9 TCe EDC	120	1735	Diesel

The weight distribution of the components is calculated according to total weight reported in Table III and percentage given in Table V. The lithium-ion battery weight is calculated from the capacity parameter using the energy density value defined in (**Tolomeo et al., 2020**).

Table V: Weight percentual distribution of components in BEVs.

COMPONENT	%
VEHICLE BODY	49.9
CHASSIS	24.0
POWERTRAIN SYSTEM	4.6
TRANSMISSION SYSTEM/GEARBOX	5.5
VEHICLE TIRE REPLACEMENT	0.7
ELECTRONIC CONTROLLER	5.7
BRAKE FLUID	0.1
TRANSMISSION FLUID	0.1
POWERTRAIN COOLANT	0.5
WINDSHIELD FLUID	0.2
ADHESIVES	1.0
LEAD ACID BATTERY (ASSEMBLY)	0.8
TRACTION MOTOR	6.9

The values in Table V are the same for all the BEVs and are not related to the vehicle segment. When modelling ICEVs, weight distribution changes as a function of market segment parameter. The values of percentage of weight are shown in Table VI, whereas total weight is reported in Table IV.

Table VI: Weight percentual distribution in ICEVs

COMPONENT	**MARKET SEGMENTS**		
	A, B, C	N, D	E, L
VEHICLE BODY	50.5%	50.1%	40.5 %
CHASSIS	24.2%	25.4%	34.9 %
POWERTRAIN SYSTEM	14.6%	13.6%	14.7 %
TRANSMISSION SYSTEM/GEARBOX	6.0%	5.6%	5.3%
VEHICLE TIRE REPLACEMENT	0.6%	0.8%	0.7%
LEAD ACID BATTERY (ASSEMBLY)	1.1%	1.4%	1.2%
ENGINE OIL	0.3%	0.3%	0.2%
POWER STEERING FLUID	0.0%	0.0%	0.0%
BRAKE FLUID	0.1%	0.1%	0.0%
TRANSMISSION FLUID	0.8%	0.8%	0.7%
POWERTRAIN COOLANT	0.7%	0.7%	0.6%
WINDSHIELD FLUID	0.2%	0.3%	0.2%
ADHESIVES	0.9%	1.0%	0.9%

The aim of this part of study was to perform a comparative LCA between electric and thermic vehicles. A cradle-to-gate plus use phase approach was used and the chosen functional unit (FU) is 150,000 km. The driving cycle considered is mixed: urban for the 50% and extra-urban the remaining 50%.

In the GREET software tool the confrontation between BEVs and ICEVs was performed considering the climate change category that is the only one that can be evaluated. In the software tool SimaPro the confrontation between BEVs and ICEVs was performed considering the ReCiPe 2016 method for the impact assessment. The comparison between GREET and SimaPro was performed only using the climate change category.

II.3 Results and discussion

II.3.1 SimaPro results

Table VII summarizes the results of the one-to-one comparison between the midpoint impacts of BEV and ICEV. The ratio between the impact value for BEV and ICEV for all the eighteenth midpoint categories of ReCiPe 2016 Midpoint (H) was calculated. If the ratio is less than one, the BEV vehicle is the most environmentally sound for that midpoint impact category, otherwise it is the ICEV the preferable option for the environmental problem considered.

Table VII: Ratio between the impact value for BEV and ICEV for all the eighteenth midpoint categories of ReCiPe 2016 (H). Global Warming Potential (GWP); Stratospheric Ozone Depletion (SOD); Ionizing Radiation (IR); Ozone Formation, Human Health (OF-HH); Fine Particulate Matter Formation (FPMF); Ozone Formation Terrestrial Ecosystems (OF-TE); Terrestrial Acidification (TA); Freshwater Eutrophication (FE); Marine Eutrophication (ME); Terrestrial Ecotoxicity (TEcotox); Freshwater Ecotoxicity (FEcotox); Marine Ecotoxicity (MEcotox); Human Carcinogenic Toxicity (HCTox); Human non-Carcinogenic Toxicity (HnCTox); Land Use (LU); Mineral Resource Scarcity (MRS); Fossil Resource Scarcity (FRS); Water Consumption (WC).

MIDPOINT IMPACT VALUE (BEV)/MIDPOINT IMPACT VALUE(ICEV)

Segment	A	A	B	B	C	C	D	E	L	N
GWP	0,63	0.58	0.75	0.72	0.69	0.61	0.78	0.9	0.62	0.82
SOD	1.28	1.25	1.37	1.39	1.31	0.97	1.48	1.19	1.27	1.16
IR	1.85	1.79	2.04	1.97	2.08	1.52	3.36	1.94	2.00	2.44
OF-HH	1.18	1.13	1.23	1.25	1.21	1.02	1.42	0.72	1.19	0.81
FPMF	1.14	1.06	1.16	1.18	1.12	0.95	2.10	0.98	1.04	1.21
OF-TE	1.25	1.21	1.30	1.32	1.27	1.09	1.46	0.78	1.23	0.88
TA	1.19	1.14	1.22	1.24	1.15	0.94	4.24	0.95	1.08	1.23
FE	1.82	1.63	1.54	1.51	1.46	1.43	1.62	1.31	1.22	1.65
ME	1.80	1.68	1.75	1.74	1.70	1.51	1.96	1.54	1.42	1.76
Tecotox	0.56	0.45	0.19	0.18	0.12	0.35	0.09	0.00	0.01	0.22
Fecotox	1.14	0.95	0.66	0.64	0.53	0.80	0.50	0.38	0.36	0.61
Mecotox	1.15	0.95	0.67	0.66	0.55	0.81	0.52	0.40	0.37	0.63
HCTox	1.31	1.16	1.27	1.29	1.22	1.15	0.89	1.16	1.04	1.29
HnCTox	1.33	1.11	0.83	0.82	0.69	0.99	0.76	0.55	0.50	0.83
LU	2.02	1.92	1.72	1.64	1.65	1.50	1.46	1.26	1.31	1.63
MRS	0.89	0.77	0.91	0.94	0.89	0.86	1.62	0.87	0.80	0.91
FRS	0.77	0.77	0.84	0.90	0.76	0.57	0.82	0.66	0.74	1.05
WC	14.68	15.32	31.93	32.64	35.38	21.21	56.99	38.94	35.92	33.51

Ten market segments for eighteen impact categories corresponds to 180 comparisons. In 74 cases, corresponding to 41.1%, the ratio was less than one and, therefore, BEV was the best option. Consequently, in the other 106 cases, corresponding to 59.9%, the ratio was more than one and, therefore, ICEV was the best option. In terms of Global warming and Terrestrial ecotoxicity, BEV was the best option for all the market segments. In fact, when the energy mix has an high share of renewable energy generation, as showed by Hofmann et al. (2016), the global warming performance of the electric vehicles are better than the thermic

one. Woo et al. (2017), conducted an analysis over 70 countries and showed that non decarbonized energy mixes led to thermic vehicles to perform better than electric in the GWP impact category.
On the contrary, in terms of Ionizing Radiation, Freshwater eutrophication, Marine eutrophication, Land use, and Water consumption, ICEV was the best option for all the market segments. As reported in the study of Burchart-Korol et al. (2018), the electric vehicles, in fact, seems to perform worser that the thermic ones when looking at acidification, eutrophication and human toxicity categories.
Therefore, only in numerical terms, by assigning the same importance to each impact category, it should be concluded that ICEV vehicles are better than BEVs in environmental terms. However, such an argument cannot be made since some environmental problems have greater priority than others. To overcome this issue, the comparison was repeated at the endpoint level, in terms of environmental damages.
Therefore, the comparison was performed using the ReCiPe 2016 Endpoint (H) method and Table VIII shows the obtained results. In this case there is a perfect balance between ICEV and BEV. The BEVs win all the one-to-one confrontations in the damage category Resources because they do not need fossil fuels. The ICEVs win all the one-to-one confrontations in the Ecosystem category because of the environmental burden related to battery production for BEVs. In the Human health category, there is a tie. Therefore, overall, in terms of damages, there is a perfect balance between the two vehicle categories in terms of damages of their life cycles.

Table VIII: Ratio between the damage value for BEV and ICEV for the endpoint categories of ReCiPe 2016 (H).

	ENDPOINT DAGAME VALUE (BEV)/ENDPOINT DAMAGE VALUE(ICEV)		
Segment	**HUMAN HEALTH**	**ECOSYSTEMS**	**RESOURCES**
A	0.97	1.17	0.57
A	0.88	1.10	0.57
B	1.05	1.32	0.62
B	1.04	1.30	0.67
C	0.98	1.26	0.55
C	0.89	1.08	0.41
D	1.22	1.94	0.60
E	1.12	1.39	0.64
L	0.88	1.12	0.54
N	1.11	1.35	0.78

Figure 8 makes clearer the comparison between BEVs and ICEVs in terms of impact on climate change. As it can be seen, for the same vehicle segment, thermal vehicles emit more than their electric counterparts. Therefore, if the choice only depended on the global warming issue, there is no doubt that the best choice would be that of electric vehicles.

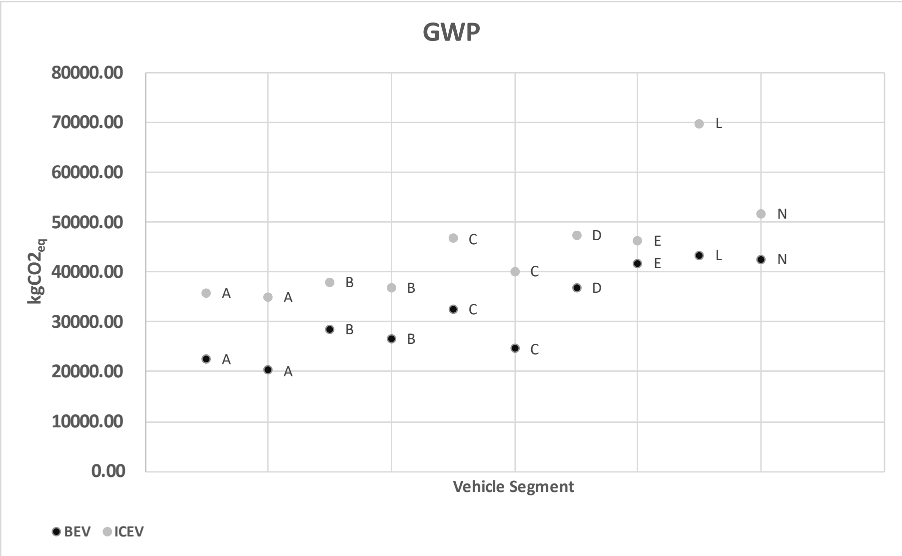
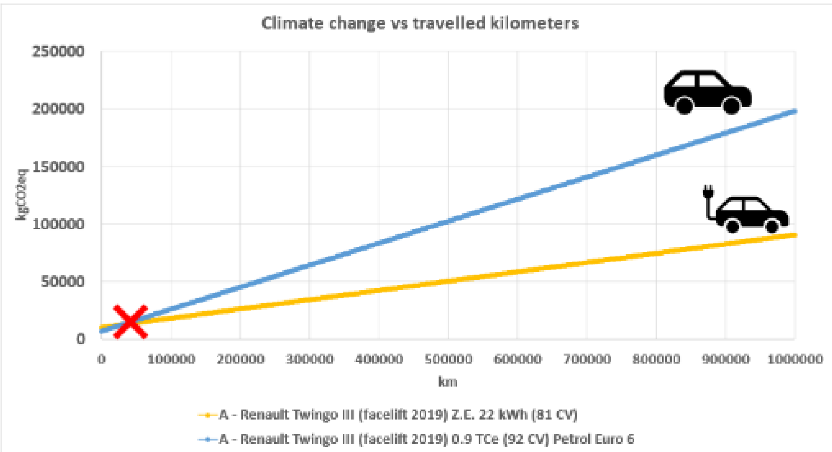


Figure 8: GWP comparison between electric and thermic vehicles in SimaPro.

As last step of the analysis performed with the software tool SimaPro, the FU was varied from 150,000 km up to 1,000,000 km, according to recent results on BEVs batteries lifespan by Tesla, Toyota and CATL, to evaluate the impact of this parameter on the GHGs emissions confrontation with respect to ICEVs. From Figure 9 it can be deduced that an increase of the life cycle of the BEVs increases their potential to reduce GHG emissions.

Figure 9: Global warming potential (expressed as kgCO₂eq vs travelled kilometers) trend for electric and thermal vehicles.

By interpolating the values obtained in Figure 9, equations can be derived to calculate CO2eq emissions as a function of vehicle kilometres travelled. By doing so, it is possible to identify the crossover point between the line for ICEV and the line for BEV for the same vehicle segment.



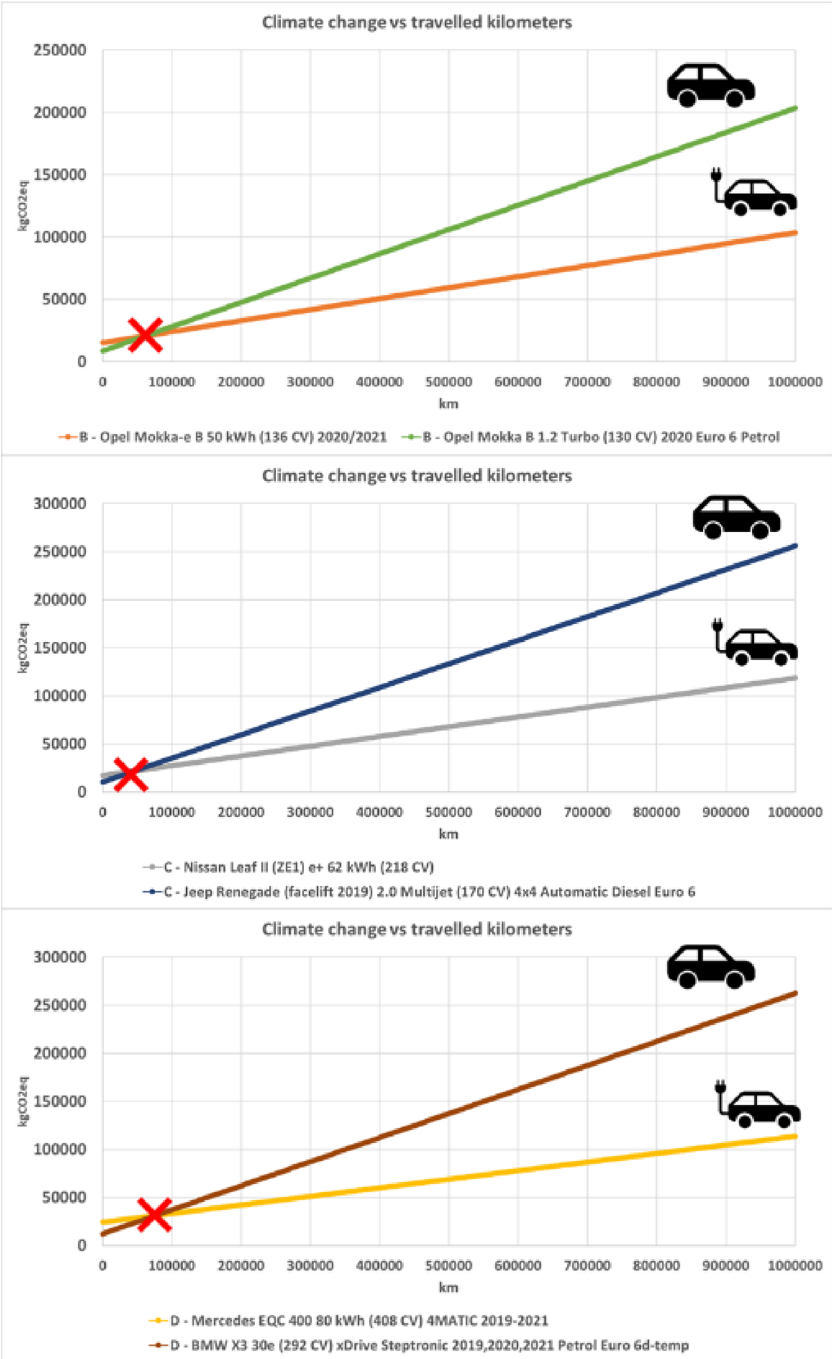


Figure 10: Climate change (espressed as kg CO_2 equivalent) vs travelled kilometers for ICEV and BEV comparison. Crossover point individuation.

The crossover point, highlighted with a red cross in Figure 10, is found for all the confrontation between 35000 km and 76000 km. Just recently, Volvo published a paper comparing its electric and thermal vehicles using primary CO_2 emission data (*Volvo Recharge – Our Range of Pure Electric and Plug-in Hybrid Cars*, 2021).

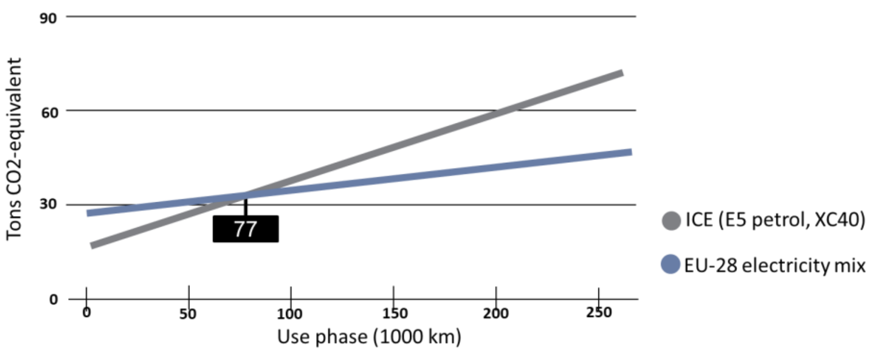


Figure 11: Tons of CO_2 equivalent as a function of travelled thousands kilometer, from Volvo LCA results

As can be seen in Figure 11, Volvo found that the crossover point at which the BEV are environmentally sounder than ICEV is at 77000 km using an EU-28 electricity mix. That result isn't too different from the one obtained by SimaPro modeling.

II.3.2 GREET results

With GREET it is possible to evaluate only the climate change impact category in terms of $kgCO_2eq$. Figure 12 shows the results obtained by using this software tool. Using the GREET software tool the ICEVs emission of greenhouse gases are higher than their electric counterparts.

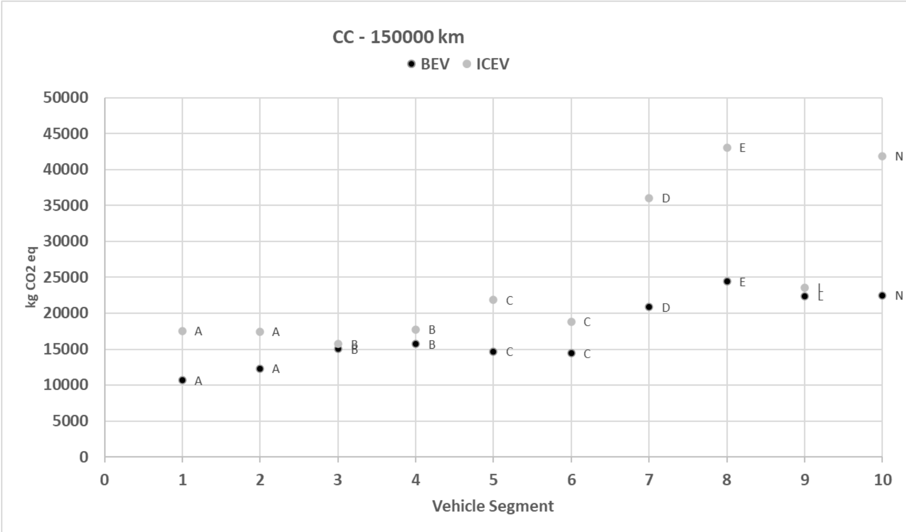


Figure 12: Climate change (expressed as kg CO_2 equivalent) as a function of vehicle segment, comparison between BEV and ICEV in GREET.

In addition, the energy mix in the GREET software tool has been changed from 41% of renewables to 55%. This value of renewables share is expected by 2030 for Italy. The modification to the energy mix was performed increasing the share of renewable that are not in saturation condition. Table IX shows the variation of the energy mix.

Table IX: Percentage contribution of the various sources in the two energy mixes considered in the study.

ENERGY SOURCE	MIX WITH 41% OF RENEWABLES	MIX WITH 55% OF RENEWABLES
COAL	6%	5%
OIL	4%	3%
NATURAL GAS	49%	37%
BIOFUELS	6%	6%

WASTE	2%	2%
HYDRO	16%	16%
GEOTHERMAL	2%	2%
SOLAR PV	8%	15%
WIND	7%	13%
OTHER SOURCES	0%	1%

The results in terms of kgCO2eq shows that an increase of the renewable share generation in energy mix from 41% to 55% reduces the GHGs emissions between the 10% and 15% as can be seen in Figure 13.

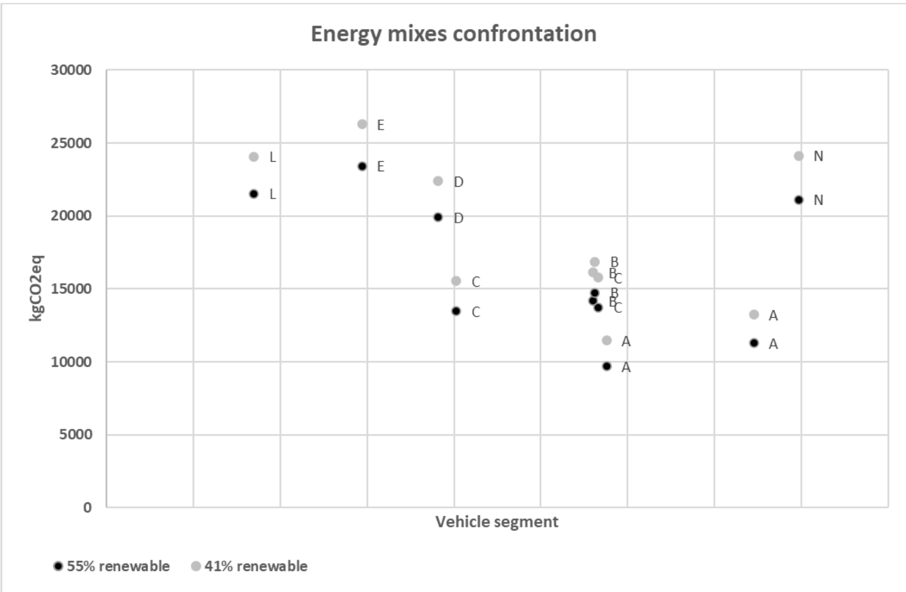


Figure 13: Climate change (expressed as kg CO_2 equivalent) as a function of vehicle segment for a BEV vehicle. The parameter is the effect of different electric generation energy mixes.

II.3.3 Confrontation among Greet and SimaPro results

To perform a methodological confrontation between the two software tools SimaPro and GREET the climate change/global warming potential category has been used. The FU is 150000 km and the energy mix considered is the one with 41% of renewable energy. The confrontation results are shown in Figure 14 and Figure 15. The two figures show the one-to-one confrontation between BEVs in GREET and SimaPro and ICEVs in GREET and SimaPro.

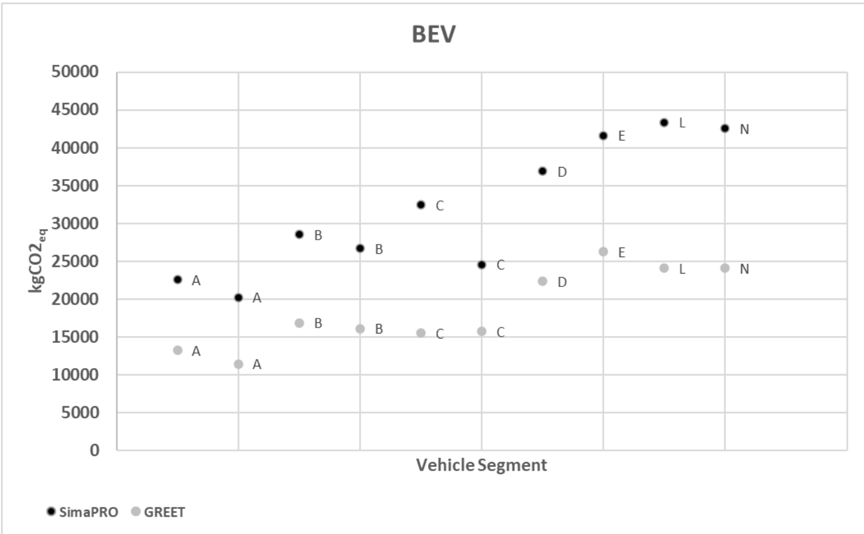


Figure 14: Climate change (expressed as kg CO_2 equivalent) as a function of vehicle segment. Results comparison for BEV vehicle, calculated by SimaPro and GREET.

The difference in value between BEVs in the two software tools varies from 36% to 52% but the order of magnitude of the results is the same.

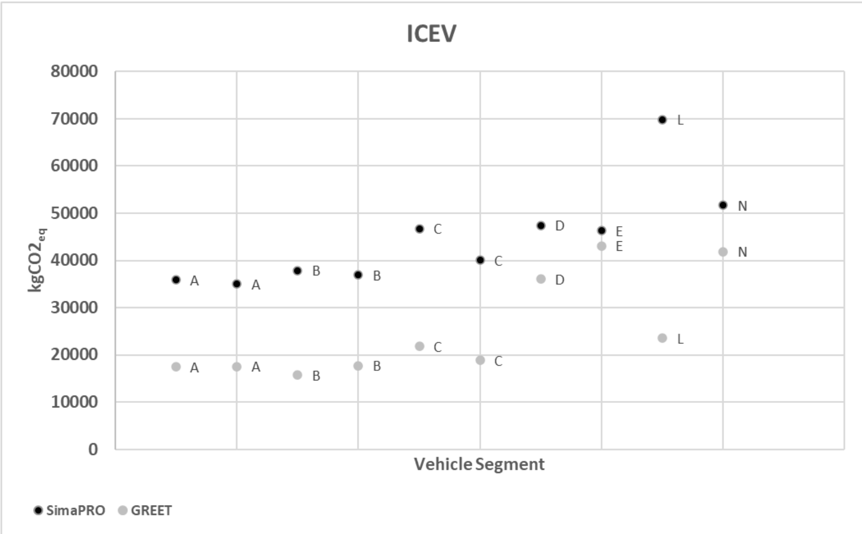


Figure 15: Climate change (expressed as kg CO_2 equivalent) as a function of vehicle segment. Results comparison for ICEV vehicle, calculated by SimaPro and GREET.

The difference in value between ICEVs in the two software tools varies from the 20% to the 60% but the order of magnitude of the results is the same. The results obtained with GREET are always lower than the results obtained with the SimaPro.

II.4 Conclusion

The aim of this study was to perform a comparison between electric and thermic vehicles and a methodological confrontation between two different software tools as SimaPro and GREET.

Looking at the comparison between ICEVs and BEVs analysing only the $kgCO_{2eq}$ both the software tools showed that the BEVs performed better than ICEVs. Moreover, the results obtained from the two software tools are of the same order of magnitude and differences are related to the different grade of details that is possible to model. These results are confirmed varying the energy mix and the travelled distance.

Looking at this result only, however, may lead to inaccurate conclusions. The SimaPro software tool permits to analyse all the midpoint impact categories. Looking at these results the ICEVs performs better than the BEVs in over 60% of the considered categories.

Chapter III
Economic and environmental assessment of lithium-ion battery recycling processes for electric vehicles

III.1 Introduction

LIBs made their name in the electric vehicles sector **(Sambamurthy et al., 2021**), where the research for renewable energy use, instead of the traditional thermic engine, has been crucial over the last twenty years. Thanks to the high potential energy storage in lithium-ion batteries, they can also be used as energy storage systems (ESS) to store energy when available and to distribute it when not available, overcoming an intrinsic limitation of the renewable sources due to their intermittent availability (**Zheng et al., 2018**). This opportunity can even contribute to reduce the dependence on fossil fuels and greenhouse gas (GHG) emissions.

Against the growing fossil fuel consumption in the transport sector, a pivotal role in the global fight against climate change (CC) is taken by the electric vehicles (EVs). EVs appear to be a possible solution to the problem of fossil fuel consumption and GHGs emissions from internal combustion vehicles (ICEVs) (**Lutsey & Hall, 2018**).

The market for electric vehicles is growing exponentially since 2010. Considering the average life and the recent extended life for newer batteries (now it is usual an eight-year warranty on LIBs), it is expected to soon dispose of a large quantity of retired LIBs **(Ciez & Whitacre, 2019; Luo & Zeng, 2017**). Despite the fact that the adoption of EVs has many benefits in terms of energy consumption and reduced local environmental impacts, the transition from ICEVs to EVs will require a large production of LIBs and thus large quantities of metals such as lithium, cobalt, manganese and nickel **(Dunn et al., 2012**). Availability of natural sources of lithium and cobalt deserve to be briefly discussed.

There are two main natural sources for lithium: brines and hard rock. The most important source of lithium is the brines, through the evaporation of saline waters containing lithium; a lithium precipitate is obtained when the concentration overcomes saturation. The second lithium source is inside the rock, where, in geological eras, sea or lake water containing lithium has deposited (**Tadesse et al., 2019**). The recovery of lithium by this way has a high environmental impact, especially due to the energy consumption (**Kushnir & Sandén, 2011**). Considering both the sources, lithium reserves do not show geopolitical criticism and then no availability problems are expected soon.

On the contrary, availability potential risks are related to the extraction of cobalt. In fact, the sources of this metal are located in a restricted area, in politically unstable countries (**Schmuch et al., 2018**). For this reason, cobalt extraction has already suffered many ethical, social and environmental concerns (**Banza Lubaba Nkulu et al., 2018**).

Therefore, in addition to the increased awareness of environmental sustainability and the forecast of an increase in battery production in the near future, there is concern about the scarcity of raw materials that pushes the reuse of materials present in batteries at the end of their life.

Moreover, the United States of America (USA) and the European Union (EU) have defined lithium and cobalt as critical materials for their availability as a resource as well as for the criticality of the supply chain. The concept of "critical material" was first introduced in the USA in 1939. This concept aims to identify those materials that are of critical importance and that are not produced in the USA in sufficient quantities to meet requirements. In 2011, the European Commission identified 14 critical raw materials (CRMs) (Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on Waste and Repealing Certain Directives (Text with EEA Relevance), 2008) comparing two parameters: the economic importance and the supply chain risk. The most recent list has been published in 2020 and has finally included lithium among the CRMs because of the increasing demand for vehicle batteries and energy storage systems. The European commission forecasted that the demand would increase by 18 times in 2030 and by 60 times until 2050 compared to the current supply (**Keersemaker, 2020**). Lithium should be recovered to close the loop for a circular economy in Europe and prevent supply chain problem related to the aspect that lithium resources are mainly located in South America. To avoid criticality in the lithium supply chain, a great effort should be made because of the low recovering rate of lithium in Europe (**Sun et al., 2019**). Sonoc et al. (2015) predicted that to ensure a stable supply chain of lithium in Europe, the minimum recovery rate should be 90%.

Waste electrical and electronic equipment (WEEEs) is one of the potential sources for valuable materials recycling. WEEEs sector is growing quickly and has a serious problem in terms of sustainability (**Menikpura et al., 2014**). At the same time, they have a high economic potential that comes from their high intrinsic value (**Sarath et al., 2015**), that imposes to study, first among all, the processes to make battery material recycle. However, in recent years only 20% of WEEEs is collected and recycled, while the rest is incinerated or stored in landfills (**Baldè et al., 2017**).

Nowadays, the techniques for recovering valuable metals from WEEEs can be divided into three different categories. The first one is the category of pyrometallurgical processes. They are conventional processes that have been used over the last two decades for recovering valuable metals from e-wastes **(Iannicelli-Zubiani et al., 2017**). The second technique is the category of hydrometallurgical processes. These processes are becoming widespread due to their high material recovery efficiency (**Dutta et al., 2018**). The latter one is a combination of pyrometallurgical and hydrometallurgical processes.

In the light of the above introduction, WEEEs experience in recovery of valuable material recovery from the end-of-life of battery and their reuse is essential also for LIBs batteries. It is also important, due to the absence of similar studies in literature, to identify the best process to recycle LIBs, considering both economic and environmental approach.

For this purpose, the methodological section of the chapter is organized in three main subsections: 1) LIBs recycling processes; 2) economic model; 3) Life Cycle Assessment (LCA). In particular, the pyrometallurgical and hydrometallurgical processes are described to be subsequently analysed and compared both in economic and environmental terms by means of the LCA methodology. This approach was applied to four different nickel-manganese-cobalt (NMC) and lithium-iron-phosphate (LFP) chemistries. For the NMC batteries, the chemistries NMC(111), NMC(622) and NMC(811) were chosen. The numbers in brackets indicate the molar ratio for cathodic metals. For example, in NMC(111) the molar ratio among nickel, manganese and cobalt is 1:1:1 whereas in NMC(811) is 8:1:1.

III.1 Methodology

III.1.1 LIBs Recycling processes

The main target of recycling technologies is to recover the valuable materials present in the end-of-life of LIBs batteries. Recovery can be obtained by using alternative processes (**Alessia et al., 2021**): hydrometallurgy, pyrometallurgy and combined processes. The first step of a recycling process is always a full electric discharge, necessary to avoid possible explosions of combustible gases and material triggered by electric short-circuiting during dismantling (**Sun & Qiu, 2011**). To discharge batteries, the most used techniques are the cryogenic freezing **(Dorella & Mansur, 2007**) and the chemical discharging where LIBs are soaked in conductor solution where NaCl and Na_2SO_4 are used as salts **(Pinna et al.,**

2017). After discharge, LIBs are submitted to some preliminary treatments to increase the recovery efficiency of valuable materials.

The biggest problem in the pre-treatment phase is the electrolyte disposal because it contains lithium compounds and upon degradation could generate harmful gases (**Cao et al., 2019; Cheng et al., 2017; Tornheim et al., 2019**).

The most common pre-treatments are crushing, shredding, sieving, air magnetic separations and manual dismantling, which generally separate the material on the basis of physical properties (density, magnetic behaviour and conductivity) (**Golmohammadzadeh et al., 2018; Hu et al., 2017**). Thermal pre-treatments, applying high temperatures (150-500 °C), are mostly used for removing binders, solvent and electrolyte solvents (**Diekmann et al., 2016**).

III.1.2 Pyrometallurgical process

The pyrometallurgical processes are based on the thermal degradation of the battery components occurring at high temperatures. The first phase can be either an incineration pre-treatment or a pyrolytic pre-treatment. Incineration is the phase in which plastics and organic compounds are burnt at high temperature (700-800°C) in air or oxygen. This phase is effective in reducing the volume the material to be treated. The pyrolytic pre-treatment focuses on the thermal degradation of organic compounds in low molecular weight products to be used as fuel or chemical feedstock. It is carried out in an inert atmosphere or under vacuum condition. The main phase is roasting/calcination and smelting and it is used to recover active cathode material using a reducing agent as carbon, charcoal, or coke. The result is a mixture of alloys containing copper, cobalt, nickel and iron and a slug phase where lithium is discarded (**Makuza et al., 2021**). Lithium and aluminium, although 100% recoverable from a technical point of view, are generally not recovered because they are not economically viable and are left in the slag phase and used as an aggregate in the cement industry (Rahman, 2017). The economic feasibility of the pyrometallurgical process strongly depends on the chemistry of LIBs, mainly in terms of cobalt content.

The economic and environmental impacts related to the treatment of lithium-manganese-oxide (LMO), lithium-iron-phosphate (LFP) and nickel-manganese-cobalt-oxide (NMC) with low cobalt grade is not better than mining the necessary resources (Winslow et al., 2018).

The LIBs recovery plants based on pyrometallurgic technology show very flexible processes, able to treat all LIBs chemistries without critical parameter adjustment (**Jha et al., 2013**). Nevertheless, pyrometallurgic plants have high capital cost, high energy consumption and significant emissions of hazardous gases. Figure 16 shows a generic pyrometallurgical process layout in which the mass balances are highlighted on the black connection arrows, where the actual mass flows are reported; the boxes represent unit operations. The feed is considered 10,000 kg (corresponding to 20 conventional battery packs, according to our hypothesis of 500 kg/battery pack). The result of mass balance is different considering different chemistries. In Figure 16 it is shown the process for NMC111 chemistry. The furnace is the core unit operation of the process. It is an electrical furnace designed to consume 30 kW of power. Another important phase is the recovery one in which Fe and Cu are separated from the "matte" phase.

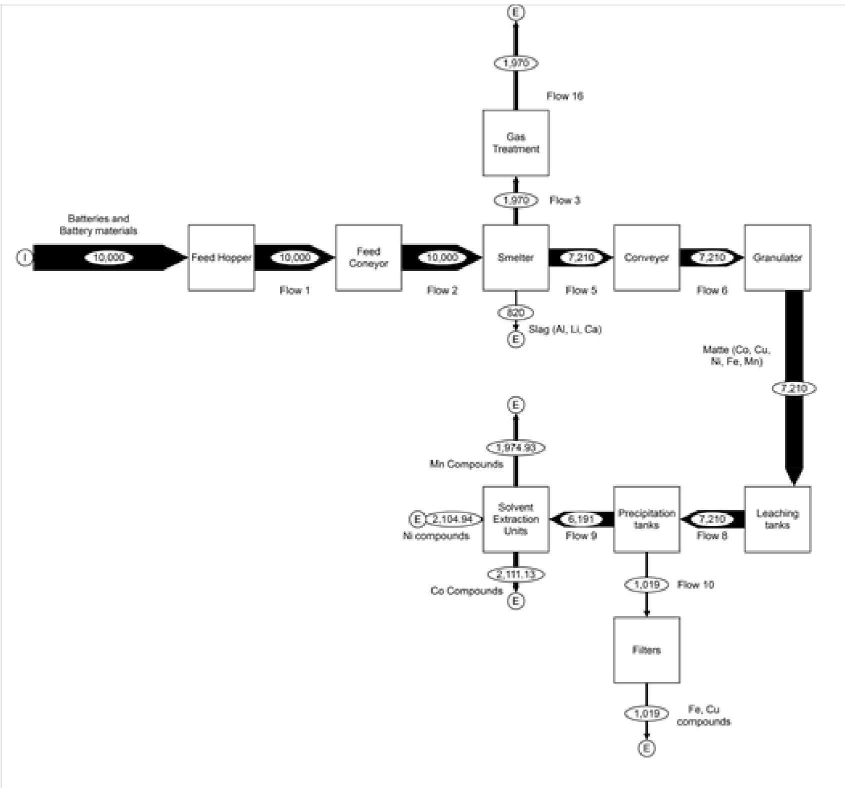


Figure 16: Generic pyrometallurgical process: flow sheet.

III.1.3 Hydrometallurgical process

After the pre-treatment already described, an alternative route to the pyrometallurgy is the hydrometallurgy, based on chemical leaching and separation as main phases (**Gao et al., 2017**). The active powder of the cathode, obtained from the pre-treatment, is leached to separate and purify LIBs metals (**Gaines, 2018**). The leaching phase involves the use of an aqueous acid solution to extract the desired metal. H_2SO_4/H_2O_2 is the most common combination on reagent reported in literature (**Ferreira et al., 2009**).

The role of the additive H_2O_2 is to be a reducing agent converting the insoluble Co(III) into Co(II), soluble in water.

Other parameters that influence the leaching stage efficiency are concentration of leaching acid, temperature, time, solid to liquid ratio **(He et al., 2017**). After leaching a series of precipitation reaction can select the solution pH suitable for the recovery of valuable metals. As a function of acid used, the metals are usually recovered as sulphate, oxalate, hydroxide or carbonate (**Gao et al., 2017; Yongxia et al., 2017**). Compared to the pyrometallurgical process, the hydrometallurgical processes require less energy because of the lower temperature involved. Moreover, this process allows lithium recovery in carbonate form and have a good efficiency on different LIBs chemistries. At the end, the leached metals could be used for new cathode production (**Gaines, 2018**). Figure 17 shows a generic hydrometallurgical process layout for NMC111 chemistry processing. The core unit operation is the leaching phase in which a solution of H_2SO_4 and H_2O_2 selectively leachs the metal of interest.

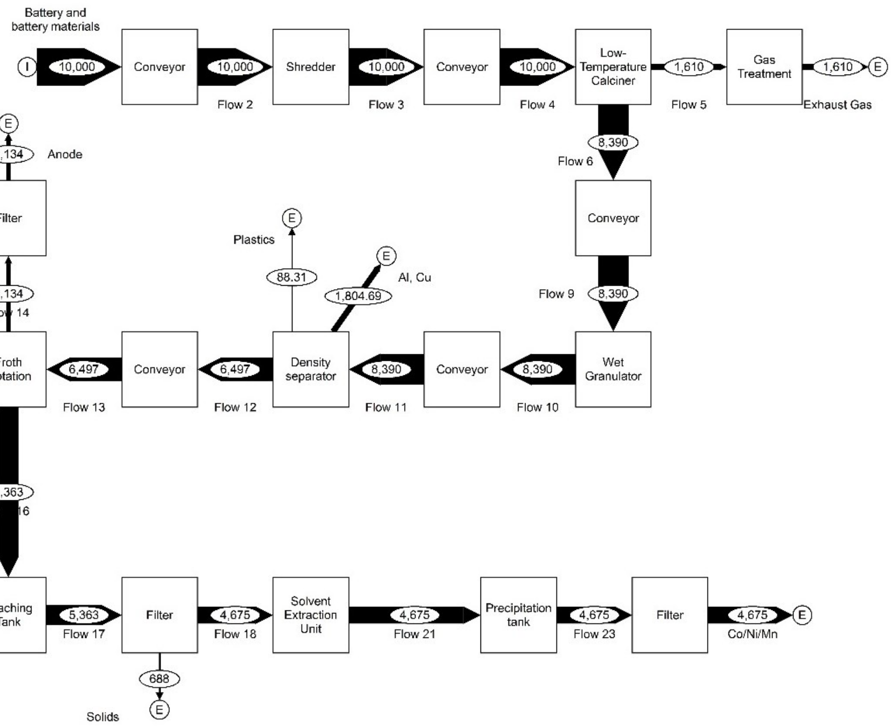


Figure 17: Generic hydrometallurgical process: flowsheet.

III.2 Economic and environmental models

The section is organized as follows: the first part reports all data and information needed to carry out the economic evaluation of the study. The second part reports all data and information used to perform the environmental analysis.

III.2.1 Economic model

A successfully industrialization of a process must be economically and environmentally suitable (**Fontana et al., 2019**). Accordingly, our aim was to assess the economic feasibility of hydrometallurgical and pyrometallurgical processes. To do this, the Liu et al., 2020 economic model, originally set up for battery waste from mobile phones, has been adapted in this work to evaluate the recovery cost of LIBs and adapted to the Italian financial rules and scenario. The model allows the calculation of the Return On Investment (ROI) according to equation (1):

ROI = Revenues - Expenditures – Taxes / (Expenditures + Taxes) (1)

Where:

- Revenues are the total yearly benefit obtained from the sale of recycled materials;
- Expenditures are the yearly total costs;
- Taxes are the mandatory taxes and fees that should be paid, per year.

The Revenues was calculated applying equation (2):

$$Revenues = \sum_{k=1}^{n} p_k * W_k \qquad (2)$$

Where:

- k is the index representing the k-th material;
- n is the total number of materials;
- p_k represents the specific sales value of the k-th material (the adopted values are reported in Table XV in the Appendix A);
- W_k represents the mass flow of the k-th material.

The Expenditures, all evaluated on a year base, were calculated with equation (3):

$$Expenditures = C_p + C_f + C_d + C_M + C_D + C_L \qquad (3)$$

Where:

- C_p is the total cost of material procurement, including raw materials and auxiliary materials (the adopted values are reported in Table S1 in the Supplementary material);
- C_f is the cost of fuel and power (the adopted values are reported in Table S2 in the Supplementary material);
- C_d is the total cost of depreciation and amortization, including construction and equipment;
- C_M is the total cost of maintenance;
- C_D is the total cost of environmental disposal;
- C_L is the total labour cost, calculated according to Italian CCNL (national collective labour agreement).

The total cost of material procurement (C_p), including raw materials and auxiliary materials, is calculated with equation (4)

$$C_p = \sum_{j=1}^{n} p_J * Q_j \tag{4}$$

Where:

- j is the index representing the j-th material;
- n is the total number of materials;
- p_j is the cost of procurement for the j-th material (the adopted values are reported in Table XV in the Appendix A);
- Q_j is the mass flow of the j-th material.

The total cost of fuel and power (C_f) is calculated with equation (5)

$$C_f = \sum_{m=1}^{n} p_m * Q_m \tag{5}$$

Where:

- m is the index representing the m-th combustible material or power;
- n is the total number of materials;
- p_m represents the cost for fuel and power procurement (the adopted values are reported in Table XVI in the Appendix A);
- Q_m represents the specific quantity combustible or power to be purchased.

The total cost of depreciation and amortization, including construction and equipment (C_d) calculated with equation (6)

$$C_d = C_R + \sum_{g=1}^{z} \frac{Q_g * P_g}{N_g} \tag{6}$$

Where:

- g is the index representing the g-th equipment;
- z is the total number of equipment;

- C_R is the cost of buildings per year;
- Q_g is the number of the g-th equipment;
- P_g is the cost of the g-th equipment (the rating equation parameters for equiments are reported in Table XVII in the Appendix A);
- N_g is the number of years considered for the amortization (lifespan of equipment) and it is 10 years in the base case.

The total cost of maintenance (C_M) calculated as the 5% of the C_d, according to Dai et al. (2019).

The total cost of environmental disposal (C_D), considering the hypothesis that the environmental disposal cost varies linearly with the scale of the plant, are calculated with equation (7) using the Liu et al., 2020 plant as reference.

$$C_D = C_{D0} \frac{Q_D}{Q_{D0}} \tag{7}$$

Where:
- C_{D0} is the cost of environmental disposal in the reference plant;
- Q_D is the capacity of the considered plant;
- Q_{D0} is the capacity of the reference plant.

The total labour fees (C_L) are calculated on Italian CCNL (national collective labour agreement) basis (Italian Ministry of Labor and Social Policies) . The labor fees used to perform calculation are reported in Table XVIII in the Appendix A.

The Taxes were calculated as sum of IRES and IRAP which are Italian taxes on production. IRES is calculated as 27.5% of the gross profit while the IRAP is calculated as the 4% of the difference between revenues and expenditures.

Summarizing, p represents the unit price of the substance; W represents the weight of the substance; Q represents the mass flow of the material; Ng represents the lifespan of the equipment.

The operability range of the model is between 0 and 100,000 t/y (corresponding to 200,000 batteries standard packs/y). The analysed chemistries are nickel-manganese-cobalt chemistries NMC(111) and NMC(811) and lithium-iron-phosphate (LFP). The difference between NMC(111) and NMC(811) is the cobalt quantity which is lower in the NMC(811). The NMC are chosen because are the most used and the LFP are chosen because they show the highest potential of substituting the NMC (**Tolomeo et al., 2020**).

III.2.2 Life Cycle Assessment

Life cycle assessment is a methodology that can be used to assess and quantify potential environmental impacts related to the life cycle of a product, process, or service. The approach can be from cradle to grave (full life cycle) or from cradle to gate, from gate to gate, etc. (i.e., considering only a portion of the life cycle). Using the cradle to gate approach considers all energy, material, and manufacturing and assembly transformations from resource extraction to the gate of the manufacturing facility. A cradle to grave approach considers the cradle to gate to which it adds use and disposal phase **(Tolomeo et al., 2020**).

The main goal of the LCA was to compare the potential environmental impacts of the hydrometallurgical and pyrometallurgical processes. The analysis follows the methodology defined under the ISO 14040 and 14044 standards (International Standard Organization, 2006), which define four main phases: (1) goal and scope definition; (2) life cycle inventory (LCI), (3) life cycle impact assessment (LCIA); and (4) interpretation. The approach used is cradle-to-grave considering the batteries production phase, the use phase, and the end-of-life recycling processes. To model the use phase of lithium-ion batteries, a life cycle of 150,000 km is considered. The energy mix considered is the Italian one which account for 55% of non-renewable energy sources.

In this study, the chosen functional unit is 20,000 battery pack/y (10,000 t/y). The data relating to the processes and necessary for the LCI phase were obtained from literature as well as from Batpac (*BatPaC*) and Everbatt (*EverBatt | Argonne National Laboratory*) databases, while the Ecoinvent 3.6 database was the main source of the background data and processes. The study was performed using the SimaPro 9.1.0.8 software tool.

Table X shows the bill of materials for the chosen LIBs chemistries (NMC (111), NMC (811) and LFP) derived from the Everbatt model and used in the LCI phase.

Table X: Bill of materials (BOM) for the chosen LIBs.

MATERIALS	**NMC (111)**	**NMC (622)**	**NMC (811)**	**LFP**
CATHODE	34.7%	32.4%	31.1%	32.7%
ANODE	19.4%	21%	20.6%	16.8%

CARBON BLACK	2.3%	2.2%	1.7%	2.2%
BINDER	3.0%	2.9%	3.6%	2.7%
COPPER	15.7%	16.1%	15.7%	13.9%
ALUMINIUM	8.2%	8.4%	8.2%	7.5%
ELECTROLYTE	14.6%	15.2%	17%	22.2%
PLASTICS	2.1%	2.2%	2.1%	1.9%

Table XI shows the material input requirements for the recycling processes used in the LCI phase derived from Everbat.

Table XI: Recycling processes, material requirements.

MATERIAL INPUTS/ENERGY CONSUMPTION	PYROMETALLURGY	HYDROMETALLURGY
LIMESTONE (KG/KGBATTERY)	0.30	-
SAND (KG/KGBATTERY)	0.15	-
HYDROCHLORIC	0.21	0.012

ACID (KG/KGBATTERY)		
HYDROGEN PEROXIDE (KG/KGBATTERY)	0.06	0.366
SODIUM HYDROXIDE (KG/KGBATTERY)	-	0.561
AMMONIUM HYDROXIDE (KG/KGBATTERY)	-	0.031
SULFURIC ACID (KG/KGBATTERY)	-	1.08
SODA ASH (KG/KGBATTERY	-	0.02

)		
WATER (KG/KGBATTERY)	-	3.79
DIESEL (MJ/KGBATTERY)	0.6	0.6
ELECTRICITY (MJ/KGBATTERY)	4.68	0.125
NATURAL GAS (MJ/KGBATTERY)	-	2.5

Table XII shows the material recovery efficiencies for the different recycling processes.

Table XII: Material recovery efficiencies for pyrometallurgical and hydrometallurgical processes.

MATERIAL	**PYROMETALLURGY**	**HYDROMETALLURGY**
COPPER	90%	90%
LITHIUM	-	90%

PLASTICS	-	50%
ALUMINIUM	-	90%
CO2+ IN OUTPUT	98%	98%
NI2+ IN OUTPUT	98%	98%
MN2+ IN OUTPUT	-	98%
ELECTROLYTE SOLVENTS	-	50%

The potential environmental impacts of the two recycling processes were estimated with the ReCiPe 2016 method considering a hierarchist perspective (H) using both the midpoint and endpoint levels. The ReCiPe 2016 method is a new version of ReCiPe 2008, and it was created by RIVM, Radboud University, Norwegian University of Science and Technology and PRé Consultants. ReCiPe 2016 combines a midpoint problem-oriented approach with an endpoint damage-oriented approach, comprising two sets of impact categories with associated sets of characterization factors. At the midpoint level, eighteen impact categories were considered, whereas, at the endpoint level, twenty-two categories were grouped into three macro-categories: human health (eight categories), ecosystems (twelve categories), and resources (two categories) (Huijbregts et al., 2017).

The damage unit for human health is DALYs (disability adjusted life years), which represents the years that are lost or that a person is disabled due to a disease or accident. The damage unit for ecosystem quality is local relative species loss in terrestrial, freshwater, and marine ecosystems, respectively, integrated over space and time (potentially disappeared fraction of species·m^2/year or potentially disappeared fraction of species·m^3/year). To aggregate the impacts of terrestrial, freshwater, and marine ecosystems into one single unit (species/year), the ReCiPe 2016 method includes species densities for these three types of ecosystems. Finally, the damage unit for resource scarcity is dollars (USD2013), which represents the extra costs involved for future mineral and fossil resource extraction **(Huijbregts et al., 2017**).

III.3 Results and discussion

III.3.1 Economic model

After the creation of the model, we evaluated the variation of ROI, revenues, and expenditures as a function of battery chemistry and the quantity of batteries processed per year.

III.3.1.1 Revenues

The revenues were calculated, according to equation (2) for every considered chemistry and for all the plant treatment capacities considered. Figure 18 shows the modelling results.

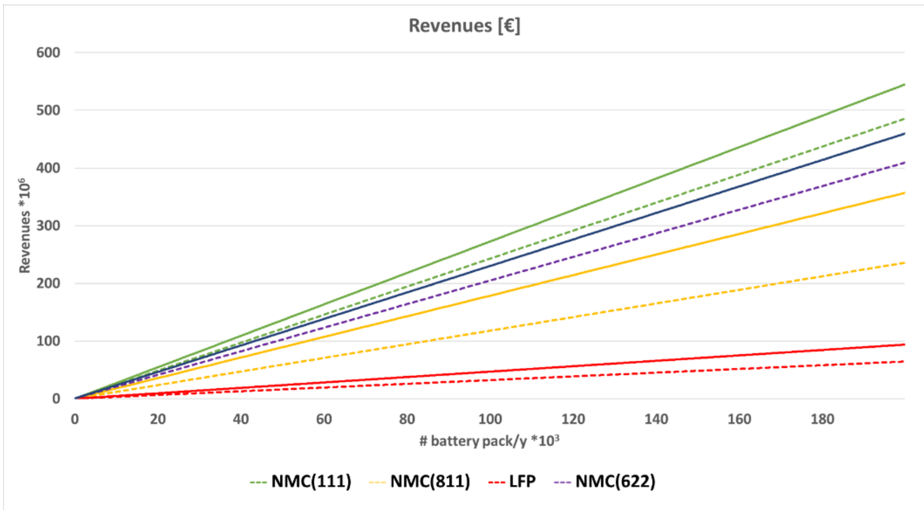


Figure 18: Revenues calculated as function of number of battery pack processed by the recycling plant (1 battery pack = 500 kg of batteries). Parameters are the battery chemistry and recycling process

A few papers in literature explicitly estimate economic aspects related to LIBs recycling processes. In particular, a sensitivity analysis done by Wang et al (X. Wang et al., 2014) shows that the profitability of a recycling process depends primarily on the chemistry of the LIBs for each recovery technology. As can be seen in Figure 18, the chemistry with the higher quantity of cobalt shows the highest revenues in both the considered processes: pyrometallurgy and hydrometallurgy. This is related to the fact that cobalt is the material with the highest reselling price and grants the highest income.

III.3.1.2 Expenditures

The expenditures were calculated according to equation (3) for every considered chemistry and for all the plant treatment capacities considered. Figure 19 shows the modelling results.

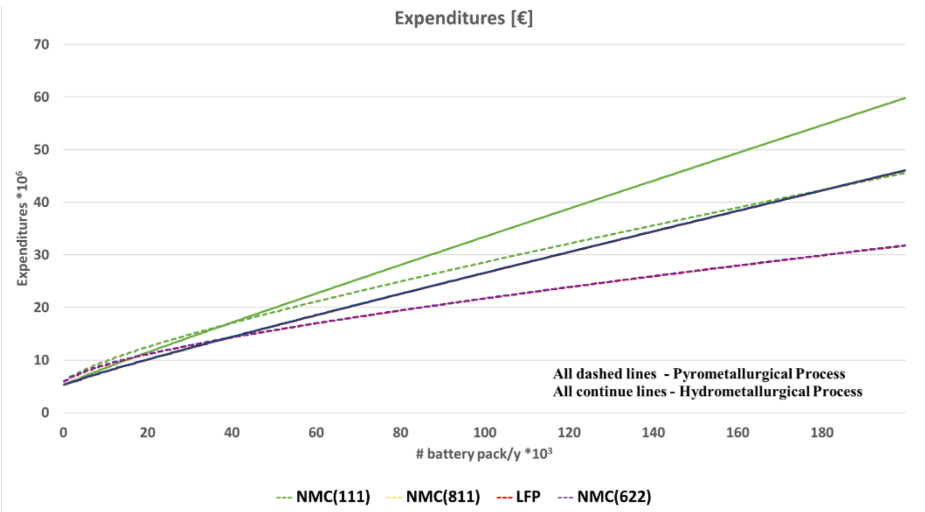


Figure 19: Expenditures calculated as function of number of battery pack processed by the recycling plant (1 battery pack = 500 kg of batteries). Parameters are the battery chemistry and recycling process.

Figure 19 shows that the curves of pyrometallurgy and hydrometallurgy revenues crosses among each other, for example for the NMC(111) chemistry this happens at 40,000 battery pack. This aspect could be related to the fact that as the plant increases its treatment capacity, the expenditures increase as consequence. In fact, the hydrometallurgical process needs more chemicals and in higher quantities than the pyrometallurgical one that uses high temperature to perform recycling. Moreover, the expenditures are function of the cost of investment that are nonlinear with the plant recycling capacity because of economies of scale. The hydrometallurgical process, in fact, has more unit operations than the pyrometallurgical one. The combination of these two contributions determines the overlapping, in one point of the two curves. This phenomenon happens in all the considered chemistries.

III.3.1.3 Return On Investment (ROI)

The ROI is calculated according to equation (1) for every considered chemistry and for all the plant treatment capacities considered. Figure 20 shows the modelling results.

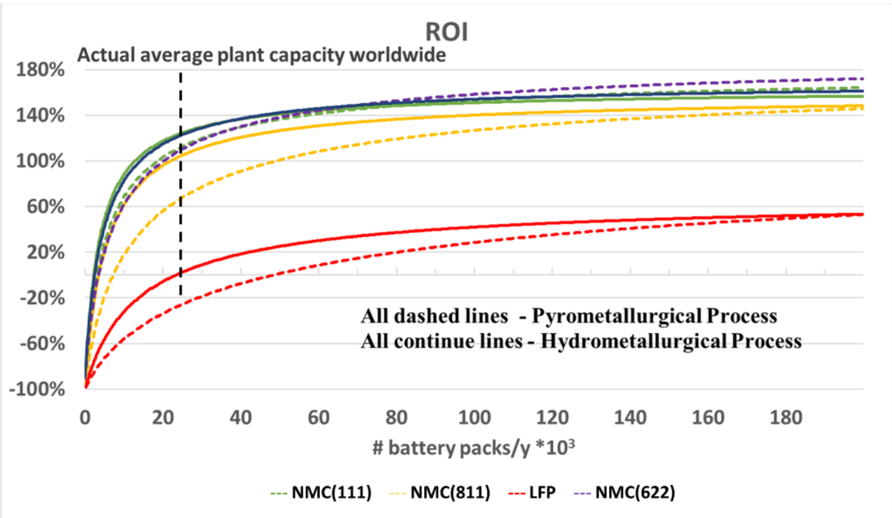


Figure 20: ROI calculated as function of number of battery pack processed by the recycling plant (1 battery pack = 500 kg of batteries). Parameters are the battery chemistry and recycling process.

Apart from LFP, the NMC chemistries have a positive ROI value early within the validity range of the model. The ROI calculated for NMC chemistries, in fact, became positive in the range of 3000-7500 battery packs/y while the ROI calculated for LFP batteries became positive in the range of 20000-48000 battery packs/y. As it can be seen from Figure 5, the ROI curves show that the pyrometallurgy and hydrometallurgy plots have a crossing point, as for expenditures curves. In the modelled range it happens only for NMC(111) and NMC(622) chemistries but, looking at the other chemistry curve trends, it seems likely to happen. Well-established battery recycling plants such as Umicore, one of the largest recyclers in Europe, revealed that "the profits from selling recovered metals are not major drivers for [their] recycling operation" and that they charge service fees to battery manufacturers or collectors (Gies, 2015). The ROI curves graph seems to confirm the fact that if these plants treat few thousand tons of battery per years, the process is not more profitable than other investment alternatives. Moreover, the hydrometallurgical process seems to be the one to prefer if the number of battery packs to be treated is less than 80,000/y (40,000 t/y). Now, in the world, there is no recycling plant operating at that scale of capacity. In fact, according to data from Sommerville et al., 2021, the medium recycling plant capacity is around 13,000 t/y.
If considered, the lithium-cobalt-oxide chemistry (LCO) would have shown the best economic results. NMC and LFP chemistries are, indeed, the most used ones (**Tolomeo et al., 2020**). Among those two, the latter is based on phosphate chemistry which contains no valuable metal and the economic performances are the lowest indeed (**Richa et al., 2017**).

III.3.2 LCA

A Life Cycle Assessment has been performed to evaluate the potential environmental impacts generated in the phases of battery production, use and recycling for both the hydrometallurgical and pyrometallurgical processes. Obviously, the absolute values for the use phase do not change between the two recycling processes, but they have been considered to perform a contribution analysis for the different battery chemistries considered: NMC (111), NMC (622), NMC (811), and LFP.

Table XIII and Table XIV contains the absolute values of the LCA results in terms of the ReCiPe 2016 Endpoint (H) and ReCiPe 2016 Midpoint (H) methods, respectively. The five midpoint impact categories considered in Table XIV are those accounting on the macro-areas of protection at the endpoint level the most. Climate Change and Human carcinogenic toxicity are the two midpoint categories (out of eight at the endpoint characterization level) with the highest incidence on the Human health macro area of protection. The midpoint characterisation factor used by ReCiPe 2016 for Climate change is the global warming potential (GWP), which quantifies the integrated infrared radiative forcing increase of a greenhouse gas (GHG), expressed in kg CO^2-eq. The fate and effects of chemical emissions expressed in kg 1,4-dichlorobenzene-equivalents (1,4DCB-eq) is used as characterisation factor at the midpoint level for Human carcinogenic toxicity (**Huijbregts et al., 2017**)
Land use and Water consumption are the two midpoint categories (out of twelve) with the highest incidence on the Ecosystems macro area of protection. In terms of Land use, the midpoint characterisation factor (in m^2/yr annual crop equivalents) refers to the relative species loss caused by a specific land use type. While, for Water consumption the characterisation factor at midpoint level is m3 of water consumed per m^3 of water extracted (**Huijbregts et al., 2017**).
Fossil resource scarcity is the midpoint category (out of two) with the highest incidence on the Resources macro area of protection. The midpoint indicator for fossil resource scarcity, expressed in terms of kg oil eq., in ReCiPe 2016 is defined as the ratio between the higher heating value of a fossil resource and the energy content of crude oil (**Huijbregts et al., 2017**).

Table XIII: Absolute values of the three ReCiPe 2016 endpoint (H) categories for the battery production (a), use phase (b), hydrometallurgical recycling (c), pyrometallurgical recycling (d), total life cycle including the hydrometallurgical recycling (a+b+c), total life cycle including the pyrometallurgical recycling (a+b+d).

PHASE	**HUMAN HEALTH (DALY)**			
	NMC (111)	NMC (622)	NMC (811)	LFP

BATTERY PRODUCTION	1332,30	1687,32	1912,34	760,70
USE PHASE	403,21	403,21	403,21	403,21
HYDRO	-644,91	-898,10	-1043,89	-351,14
PYRO	-645,53	-916,50	-1066,65	-303,80
TOTAL (HYDRO)	1090,60	1192,42	1271,65	812,76
TOTAL (PYRO)	1089,97	1174,03	1248,89	860,10
PHASE	**Ecosystems (species/yr)**			
	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	4,33	4,88	5,18	2,78
USE PHASE	1,51	1,51	1,51	1,51
HYDRO	-1,79	-2,21	-2,41	-1,02
PYRO	-1,76	-2,22	-2,42	-0,86
TOTAL (HYDRO)	4,05	4,18	4,28	3,27
TOTAL (PYRO)	4,08	4,17	4,27	3,43
PHASE	**Resources (USD2013)**			
	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	11735143	12551914	14036849	9164757
USE PHASE	17336975	17336975	17336975	17336975
HYDRO	-662563	-1411898	-1790297	-1783766
PYRO	-1737924	-2570496	-2991742	-365764
TOTAL (HYDRO)	28409555	28476992	29583527	24717966
TOTAL (PYRO)	27334194	27318393	28382082	26135968

Table XIV: Absolute values of the five most impacting ReCiPe 2016 midpoint (H) categories for the battery production (a), use phase (b), hydrometallurgical recycling (c), pyrometallurgical recycling (d), total life *cycle including the hydrometallurgical recycling (a+b+c), total life cycle including the pyrometallurgical recycling (a+b+d).*

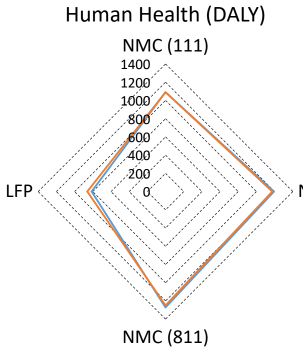
PHASE	CLIMATE CHANGE (KG CO_2 EQ.)			
	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	103554930	109428230	120531220	80441434
USE PHASE	639320358	639320358	639320358	639320358
HYDRO	-2374326	-8333792,5	-11277368	-12488341
PYRO	-83954	-6692430	-9970517	-11406623
TOTAL (HYDRO)	740500962	740414796	748574210	707273451
TOTAL (PYRO)	742791334	742056158	749881061	708355169
PHASE	Human carcinogenic toxicity (kg 1,4-DCB)			
	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	10414382	12085514	13448585	10810957
USE PHASE	52094846	52094846	52094846	52094846
HYDRO	-6561321	-7743190	-8260762	-7636089
PYRO	-6404900	-7670987	-8214341	-4020079
TOTAL (HYDRO)	55947907	56437169	57282668	55269713
TOTAL (PYRO)	56104327	56509373	57329090	58885724
PHASE	Land use (m_2a crop eq.)			

	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	178739400	193617360	200068500	89306780
USE PHASE	207787391	207787391	207787391	207787391
HYDRO	-131982030	-147433660	-151100290	-76116396
PYRO	-128514680	-145266960	-149201340	-69996245
TOTAL (HYDRO)	254544761	253971091	256755601	220977775
TOTAL (PYRO)	258012111	256137791	258654551	227097926
PHASE	Water consumption (m^3)			
	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	108749260	111016340	108926010	100073810
USE PHASE	89189301	89189301	89189301	89189301
HYDRO	-84502	-157820	-183852	-707541
PYRO	-336312	-417119	-446201	-126987
TOTAL (HYDRO)	197854059	200047821	197931459	188555570
TOTAL (PYRO)	197602249	199788522	197669110	189136124
PHASE	Fossil resource scarcity (kg oil eq.)			
	NMC (111)	NMC (622)	NMC (811)	LFP
BATTERY PRODUCTION	30975844	32299892	34938602	24297303
USE PHASE	186041283	186041283	186041283	186041283
HYDRO	-779855	-1215948	-1871537	-3098973

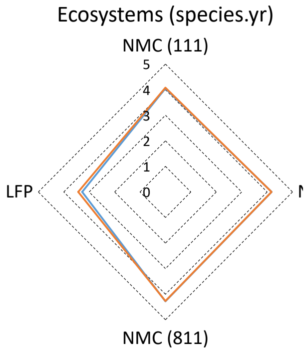
PYRO	-3083055	-4702648	-5472666	-50566
TOTAL (HYDRO)	216237272	217125227	219108348	207239613
TOTAL (PYRO)	213934072	213638527	215507219	210288020

Table XIII and Table XIV point out that the life cycle of the LFP battery with the hydrometallurgical recycling was the most environmentally sound for all the endpoint and midpoint categories taken into consideration. On the contrary, the situation was diversified in terms of the worst environmental alternative. NMC (811) + pyrometallurgy was the worst combination for all the damage categories as well as in terms of Climate change and Land use. The other worst environmental combinations were LFP + pyrometallurgy for Human carcinogenic toxicity, NMC (622) + hydrometallurgy for Land use, and NMC (811) + hydrometallurgy for Fossil resource scarcity. In any case, for both the endpoint and midpoint categories considered the percentage differences between the average total life cycle impacts of the two types of batteries were less than 2%.

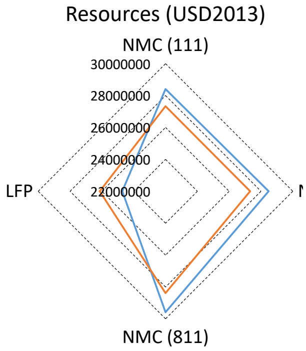
Figure 21 gives an overview of the situation for all the impact categories and battery chemistries considered. As it can be seen, the environmental performances of the life cycles of the batteries were very near for all the impact and damage categories except for Resources (Figure 21c), Human carcinogenic toxicity (Figure 21β), Land use (figure 21γ), and Fossil resource scarcity (Figure 21ε). In terms of Resources and, obviously, in terms of Fossil resource scarcity, the quadrilateral representing the life cycles with the hydrometallurgical recycling goes out the margins of the quadrilateral representing the life cycles with the pyrometallurgical recycling, except for the LFP battery.


Human Health (DALY)
NMC (111)
1400
1200
1000
800
600
400
200
0
LFP
NMC (811)

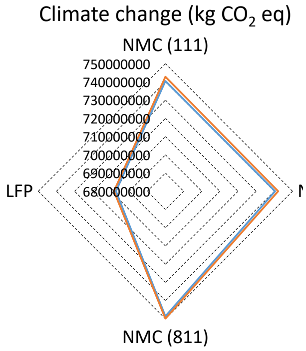
(a)


Ecosystems (species.yr)
NMC (111)
5
4
3
2
1
0
LFP
NMC (811)

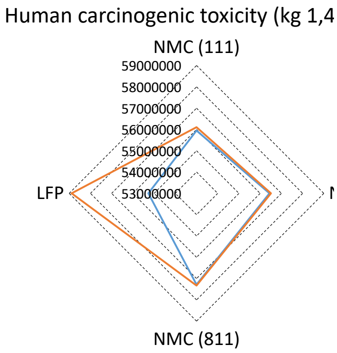
(b)


Resources (USD2013)
NMC (111)
30000000
28000000
26000000
24000000
22000000
LFP
NMC (811)

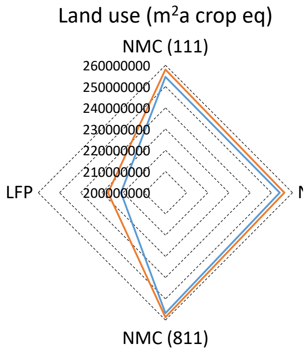
(c)


Climate change (kg CO2 eq)
NMC (111)
750000000
740000000
730000000
720000000
710000000
700000000
690000000
680000000
LFP
NMC (811)

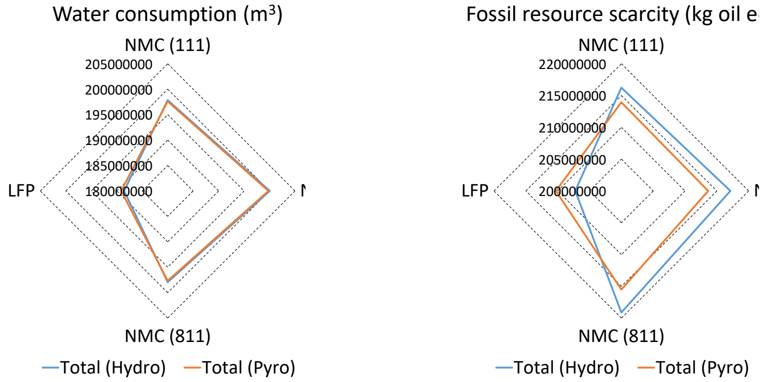
(α)


Human carcinogenic toxicity (kg 1,4
NMC (111)
59000000
58000000
57000000
56000000
55000000
54000000
53000000
LFP
NMC (811)

(β)


Land use (m2a crop eq)
NMC (111)
260000000
250000000
240000000
230000000
220000000
210000000
200000000
LFP
NMC (811)

(γ)



(δ) (ε)

Figure 21: Potential environmental impacts calculated with the ReCiPe 2016 (H) method for the four battery chemistries considered. Endpoint categories: a) Human Health; b) Ecosystems; c) Resources. Midpoint categories: α) Climate change; β) Human carcinogenic toxicity; γ) Land use; δ) Water consumption; ε) Fossil resource scarcity.

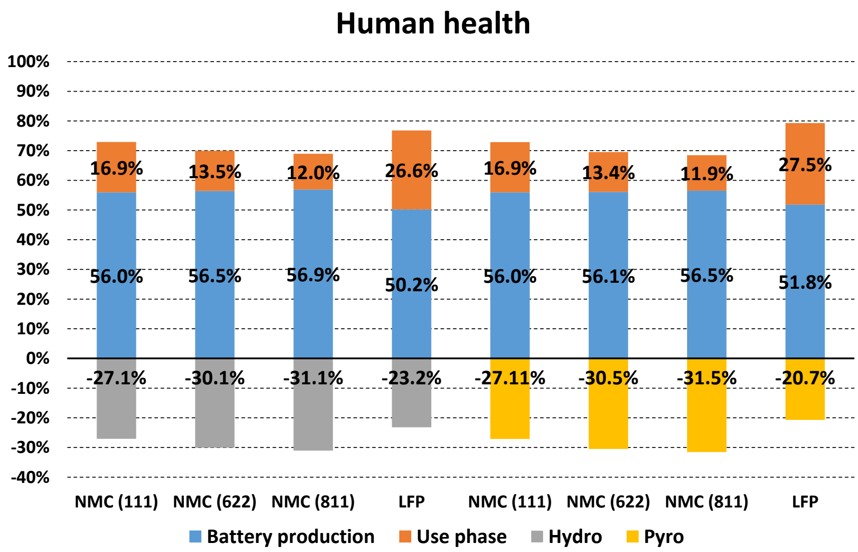
Figure 22 shows the percentage contribution analysis for the four battery chemistries considered. Figures 22a and Figure22b show that when looking at the Human health and Ecosystems damage categories, the production phase is that with the highest contribution. This is due to the metal extraction performed with non-environmentally friendly processes (**Yuan et al., 2021**). It is in these damage categories that the recycling processes show the highest contribution in terms of avoided damages. The production of secondary raw materials avoids the production of virgin raw materials (**Dunn et al., 2021**).

In the Resources damage category, in Figure 22c, the use phase is that with the highest contribution because of the energy generation and consumption for batteries charging over their life cycle. Recycling processes contribute for less than 10% of avoided damages because themselves consume resources to be performed.

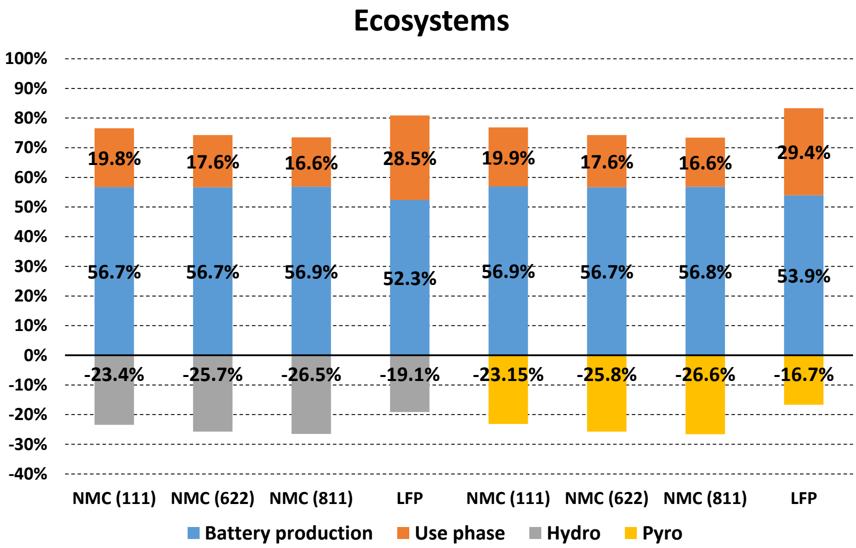
Climate change and Fossil resource scarcity are shown in Figure 22α and 22ε. As for the Resource damage category, the highest contribution is due to the use phase and is related to the electricity generation with the Italian energy mix. The hydrometallurgical and pyrometallurgical processes shows no contribution in avoiding global warming impacts.

Figure 22β shows that the use phase contributes the most in terms of Human carcinogenic toxicity. Both the recycling processes shown in

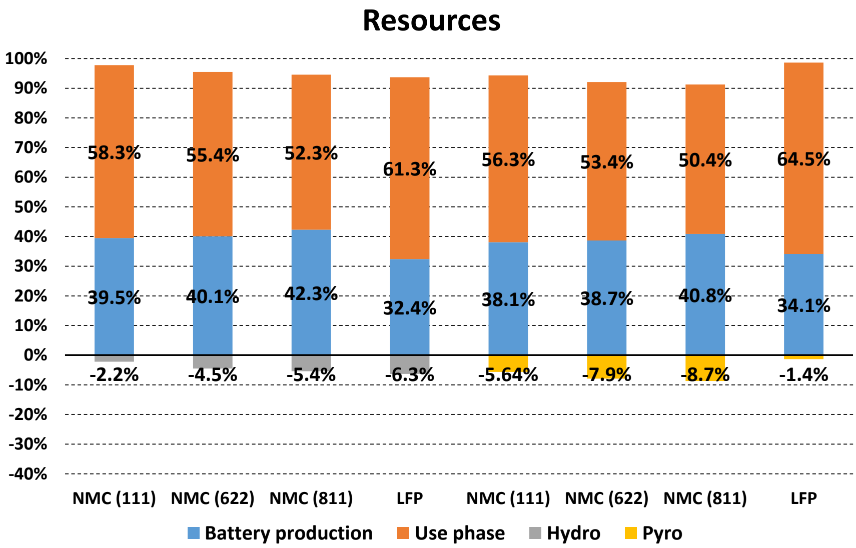
Figure 22γ have a high contribution in avoiding Land use impacts. Their contribution is related to the production of secondary raw materials. The Water consumption impact category, shown in Figure 22δ, has no benefits from the recycling processes of LIBs because both the recycling processes use water to perform the materials recovery. Moreover, there is no significant difference in the magnitude of avoided impacts shown from both processes. Finally, in terms of Fossil resource scarcity, as shown in Figure 22ε, there is a great predominance of the use phase due to the non-renewable share of the Italian energy mix used in LIBs charging phase.



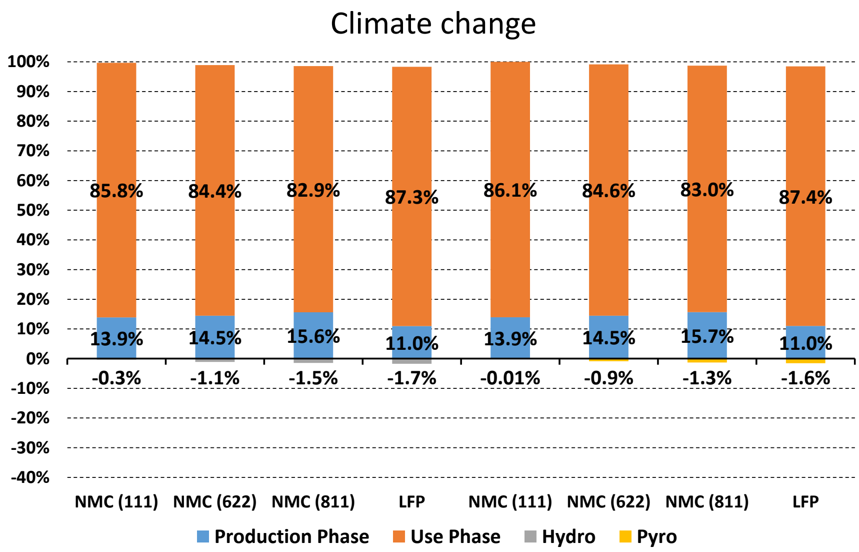
(a)


Ecosystems
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
-10%
-20%
-30%
-40%
19.8%
17.6%
16.6%
28.5%
19.9%
17.6%
16.6%
29.4%
56.7%
56.7%
56.9%
52.3%
56.9%
56.7%
56.8%
53.9%
-23.4%
-25.7%
-26.5%
-19.1%
-23.15%
-25.8%
-26.6%
-16.7%
NMC (111)
NMC (622)
NMC (811)
LFP
NMC (111)
NMC (622)
NMC (811)
LFP
Battery production
Use phase
Hydro
Pyro

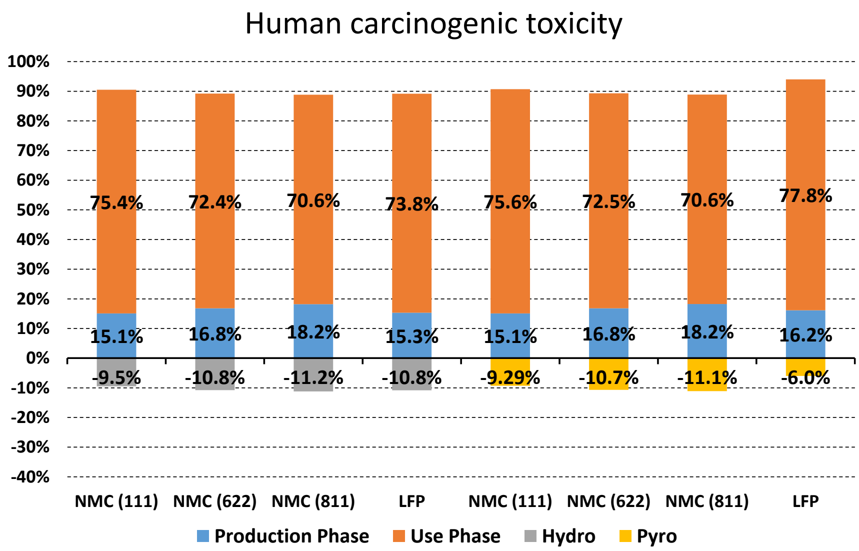
(b)


Resources
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
-10%
-20%
-30%
-40%
58.3%
55.4%
52.3%
61.3%
56.3%
53.4%
50.4%
64.5%
39.5%
40.1%
42.3%
32.4%
38.1%
38.7%
40.8%
34.1%
-2.2%
-4.5%
-5.4%
-6.3%
-5.64%
-7.9%
-8.7%
-1.4%
NMC (111)
NMC (622)
NMC (811)
LFP
NMC (111)
NMC (622)
NMC (811)
LFP
Battery production
Use phase
Hydro
Pyro

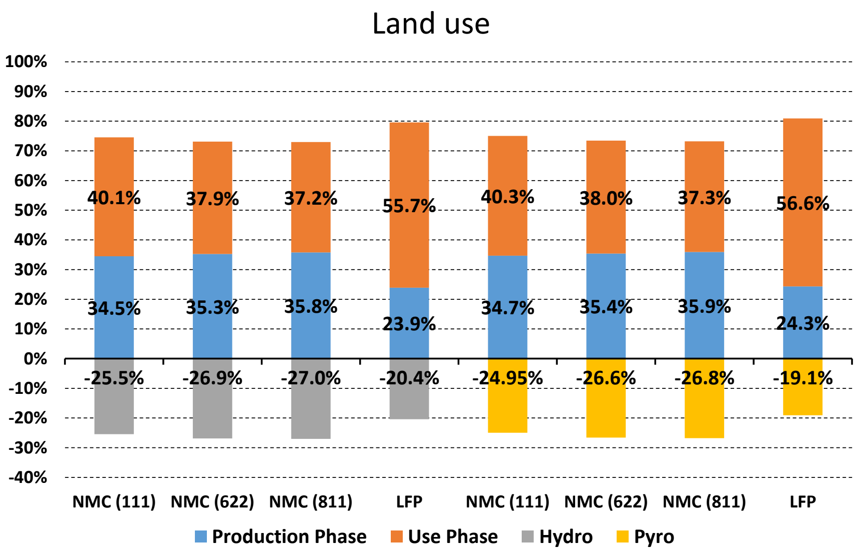
(c)


Climate change
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
-10%
-20%
-30%
-40%
85.8%
84.4%
82.9%
87.3%
86.1%
84.6%
83.0%
87.4%
13.9%
14.5%
15.6%
11.0%
13.9%
14.5%
15.7%
11.0%
-0.3%
-1.1%
-1.5%
-1.7%
-0.01%
-0.9%
-1.3%
-1.6%
NMC (111)
NMC (622)
NMC (811)
LFP
NMC (111)
NMC (622)
NMC (811)
LFP
Production Phase
Use Phase
Hydro
Pyro

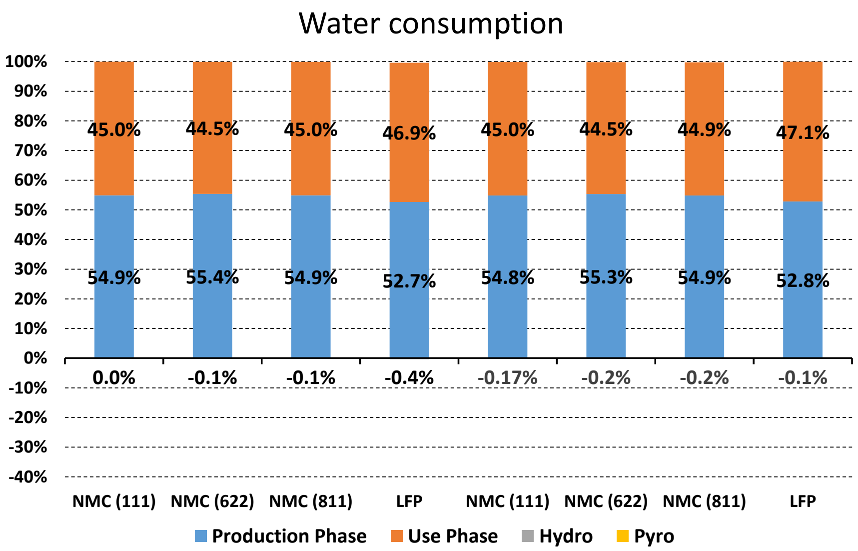
(α)


Human carcinogenic toxicity
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
-10%
-20%
-30%
-40%
75.4%
72.4%
70.6%
73.8%
75.6%
72.5%
70.6%
77.8%
15.1%
16.8%
18.2%
15.3%
15.1%
16.8%
18.2%
16.2%
-9.5%
-10.8%
-11.2%
-10.8%
-9.29%
-10.7%
-11.1%
-6.0%
NMC (111)
NMC (622)
NMC (811)
LFP
NMC (111)
NMC (622)
NMC (811)
LFP
Production Phase
Use Phase
Hydro
Pyro

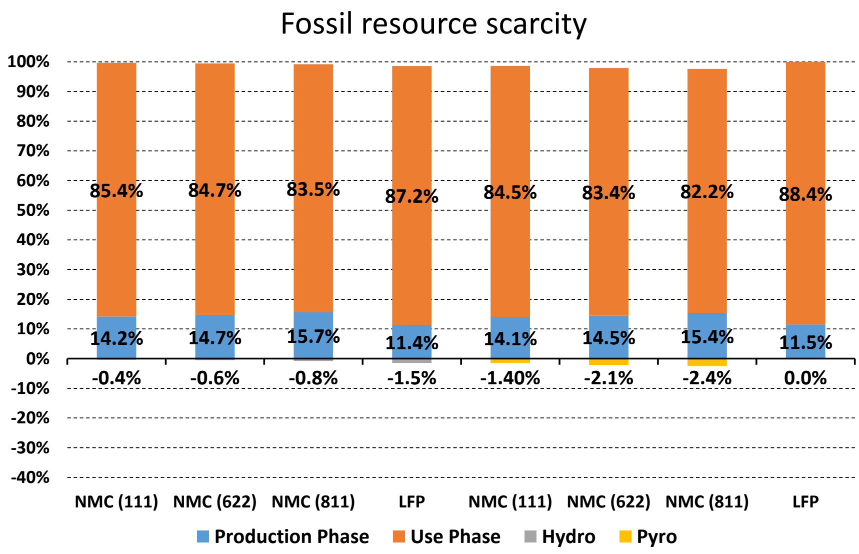
(β)


Land use
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
-10%
-20%
-30%
-40%
40.1%
37.9%
37.2%
55.7%
40.3%
38.0%
37.3%
56.6%
34.5%
35.3%
35.8%
23.9%
34.7%
35.4%
35.9%
24.3%
-25.5%
-26.9%
-27.0%
-20.4%
-24.95%
-26.6%
-26.8%
-19.1%
NMC (111)
NMC (622)
NMC (811)
LFP
NMC (111)
NMC (622)
NMC (811)
LFP
Production Phase
Use Phase
Hydro
Pyro

(γ)


Water consumption
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
-10%
-20%
-30%
-40%
45.0%
44.5%
45.0%
46.9%
45.0%
44.5%
44.9%
47.1%
54.9%
55.4%
54.9%
52.7%
54.8%
55.3%
54.9%
52.8%
0.0%
-0.1%
-0.1%
-0.4%
-0.17%
-0.2%
-0.2%
-0.1%
NMC (111)
NMC (622)
NMC (811)
LFP
NMC (111)
NMC (622)
NMC (811)
LFP
Production Phase
Use Phase
Hydro
Pyro

(δ)



(ε)

Figure 22: Contribution analysis for the four battery chemistries considered. Endpoint categories: a) Human Health; b) Ecosystems; c) Resources. Midpoint categories: α) Climate change; β) Human carcinogenic toxicity; γ) Land use; δ) Water consumption; ε) Fossil resource scarcity.

III.3.3 Conclusions

The hydrometallurgical process is often subject to criticism because of its complexity if compared with the pyrometallurgical process. On the other hand, the pyrometallurgical process is energy intensive and can't be used to recover all the valuable materials. However, the obtained results confirm that the economic profitability of the hydrometallurgical process is significantly superior to the pyrometallurgical process due to the higher purity and larger number of recoverable materials. Therefore, from an economic point of view the hydrometallurgical process is the preferable one. Instead, from an environmental point of view, for both the endpoint and midpoint categories considered the percentage differences between the average total life cycle impacts of the two types of batteries were less than 2%. In more details, the life cycle of the lithium-iron-phosphate (LFP) battery with the hydrometallurgical recycling was the most environmentally sound for all the endpoint and midpoint categories taken into consideration. This result is related to the utilization of cheap, largely available and environmentally sound materials. Overall, we can conclude that the hydrometallurgical process was the best option when looking at both economic and environmental aspects and the LFP batteries will be a candidate solution to reduce the environmental burden related to the electrification of transportation sector.

Conclusions

The recovery processes of valuable materials from LIBs are of great interest in the perspective of circular economy and in view of the reuse of rare metals they contain.
At present, the recovery processes ready for industrial development can be divided into two families: pyrometallurgy and hydrometallurgy. The first technique is based on the preliminary high-temperature thermal destruction of every organic compound present in the batteries, followed by the selective recovery of what is contained in the unburned residue; the second technique is based on selective extractions of precious metal compounds, using inorganic and organic solvents.
A first aim of this thesis was to evaluate, on the basis of the existing literature, whether one technique was preferable to the other, evaluating them first in terms of environmental impact and then in terms of cost and profitability.
We therefore proceeded to carry out an evaluation of the environmental impact for both processes, using the SimaPro software and the relative databases (Ecoinvent). In detail, the analysed processes are therefore those experimentally more mature, which are already present in the Ecoinvent database. Apart from the thermal destruction phase, which is based on an incineration furnace, both the processes examined essentially involve extraction with inorganic acids for the selective recovery of precious metals. In the hydrometallurgical process, the extraction units are more numerous than in the pyrometallurgical process, allowing the recovery of a greater number of materials.
The comparative environmental assessment of these processes resulted, with reference to the main impact factors, in a substantial balance. The advantages of recovering a greater number of materials from the hydrometallurgical process are offset by the need for greater use of solvents, chemicals whose production nevertheless has an environmental impact.
From an environmental point of view, therefore, the two processes are, to date, quite equivalent.

From the point of view of economic investment, the results have been obtained from a home-made model that takes into account the chemistry of LIBs and evaluates the investment costs of individual equipment and their operating costs (energy consumption, raw material costs and the

value of the products obtained). All results are evaluated as a function of plant capacity.
The analysis of the economic results shows that both processes have their own profitability, starting from a certain minimum capacity (break even production). The Return Of Investment (ROI) depends on the chemistry of the spent batteries treated.
The hydrometallurgical plant has lower investment costs than the pyrometallurgical ones. Moreover, the ROI - with the same chemistry of spent batteries - is higher and the investment is profitable starting from plants with lower capacity (if the capacity is expressed in terms of quantity of treated batteries).
Finally, it should not be overlooked that the hydrometallurgical process, also allowing the recovery of precious lithium, which is impossible with the alternative process, fully meets the requirements of the draft European regulation for battery recovery, which provides for an obligation to recover also this material in the coming years.

The impact of battery recycling compared to the use of virgin mining materials has also been assessed. The result shows a reduction in impact, which obviously depends on the impact categories analyzed. Taking Acidification, Eutrophication and Ecotoxicity categories as examples, there is a reduction in impact more than 50%. Although the percentage is significant, it is believed that in the near future this percentage may increase, thanks to the development of new and more selective processes for extracting precious metals from spent batteries.

The use in automotive applications is the main destination of lithium-ion batteries. In this PhD thesis a technical comparison has been carried out, therefore, between thermal and electric vehicles belonging to all consumer segments, in order to understand if the latter represent an environmentally sounder alternative and a methodological comparison between LCA software tools has been carried out between a generalist professional tool (SimaPRO) and a specialized and open-source tool (GREET).

The results of the methodological comparison show that the two software tools provide results with the same trends and of the same orders of magnitude. Comparing those vehicles using climate change impact category, electric vehicles are the undisputed winners. The

sensitivity analysis conducted on the energy mix, increasing the content of renewables, also shows how this range can widen, disfavoring thermal vehicles.
Moreover, by analysing emission trends as a function of kilometers driven, it is possible to identify a crossover point between thermal and electric vehicles, below which (number of kilometers driven lower than that of this characteristic point) thermal vehicles are to be preferred to electric vehicles. This crossover point varies according to the chosen segment of the vehicle but, in general, it is in the range between 35000 and 76000 kilometers.
It is precisely the upper end of this range that, when compared to the value of 77000 kilometers declared by Volvo in its official comparative LCA document, provides value and robustness to the modelling carried out.

In any case, relying only on climate change does not provide a complete picture. In fact, looking at all impact categories of the ReCiPe method, thermal vehicles come out as winners in 11 out of 18 categories. Finally, considering the damage categories, the comparison between the two classes of vehicles is perfectly balanced.

References

Aguirre, K., Eisenhardt, L., Lim, C., Nelson, B., Norring, A., Slowik, P., Tu, N., & Rajagopal, D. D. (n.d.). *Lifecycle Analysis Comparison of a Battery Electric Vehicle and a Conventional Gasoline Vehicle*. 33.

Ahmadi, L., Yip, A., Fowler, M., Young, S. B., & Fraser, R. A. (2014). Environmental feasibility of re-use of electric vehicle batteries. *Sustainable Energy Technologies and Assessments*, *6*, 64–74. https://doi.org/10.1016/j.seta.2014.01.006

Ahmadi, L., Young, S., Fowler, M., Fraser, R., & Ahmadi achachlouei, M. (2015). A cascaded life cycle: Reuse of electric vehicle lithium-ion battery packs in energy storage systems. *The International Journal of Life Cycle Assessment*. https://doi.org/10.1007/s11367-015-0959-7

Alessia, A., Alessandro, B., Maria, V.-G., Carlos, V.-A., & Francesca, B. (2021). Challenges for sustainable lithium supply: A critical review. *Journal of Cleaner Production*, 126954. https://doi.org/10.1016/j.jclepro.2021.126954

Ambrose, H., & Kendall, A. (2016). Effects of battery chemistry and performance on the life cycle greenhouse gas intensity of electric mobility. *Transportation Research Part D: Transport and Environment*, *47*, 182–194. https://doi.org/10.1016/j.trd.2016.05.009

Anderson, R., Keshwani, D., Guru, A., Yang, H., Irmak, S., & Subbiah, J. (2018). An integrated modeling framework for crop and biofuel systems using the DSSAT and GREET models. *Environmental Modelling & Software*, *108*, 40–50. https://doi.org/10.1016/j.envsoft.2018.07.004

Andersson, Ö., & Börjesson, P. (2021). The greenhouse gas emissions of an electrified vehicle combined with renewable fuels: Life cycle assessment and policy implications. *Applied Energy*, *289*, 116621. https://doi.org/10.1016/j.apenergy.2021.116621

Anonymous. (2016, July 5). *Vehicle categories* [Text]. Internal Market, Industry, Entrepreneurship and SMEs - European Commission. https://ec.europa.eu/growth/sectors/automotive/vehicle-categories_en

Baldè, C. P., Forti, V., Gray, V., Kuehr, R., & Stegmann, P. (2017). *The Global E-waste Monitor – 2017,*. United Nations University (UNU), International Telecommunication Union (ITU) & International Solid Waste Association (ISWA), Bonn/Geneva/Vienna.

Banza Lubaba Nkulu, C., Casas, L., Haufroid, V., De Putter, T., Saenen, N. D., Kayembe-Kitenge, T., Musa Obadia, P., Kyanika Wa Mukoma, D., Lunda Ilunga, J.-M., Nawrot, T. S., Luboya Numbi, O., Smolders, E., & Nemery, B. (2018). Sustainability of artisanal mining of cobalt in DR Congo. *Nature Sustainability*, *1*(9), 495–504. https://doi.org/10.1038/s41893-018-0139-4

Baptista, P. C., Silva, C. M., Peças Lopes, J. A., Soares, F. J., & Almeida, P. R. (2013). Evaluation of the benefits of the introduction of electricity powered vehicles in an island. *Energy Conversion and Management*, *76*, 541–553. https://doi.org/10.1016/j.enconman.2013.07.075

BatPaC: Battery Manufacturing Cost Estimation | Argonne National Laboratory. (n.d.). Retrieved June 7, 2021, from https://www.anl.gov/partnerships/batpac-battery-manufacturing-cost-estimation

Bjørn, A., Owsianiak, M., Molin, C., & Hauschild, M. Z. (2018). LCA History. In M. Z. Hauschild, R. K. Rosenbaum, & S. I. Olsen (Eds.), *Life Cycle Assessment: Theory and Practice* (pp. 17–30). Springer International Publishing. https://doi.org/10.1007/978-3-319-56475-3_3

Bobba, S., Mathieux, F., Ardente, F., Blengini, G. A., Cusenza, M. A., Podias, A., & Pfrang, A. (2018). Life Cycle Assessment of repurposed electric vehicle batteries: An adapted method based on modelling energy flows. *Journal of Energy Storage*, *19*, 213–225. https://doi.org/10.1016/j.est.2018.07.008

Boyden, A., Soo, V. K., & Doolan, M. (2016). The Environmental Impacts of Recycling Portable Lithium-Ion Batteries. *Procedia CIRP*, *48*, 188–193. https://doi.org/10.1016/j.procir.2016.03.100

Brady, J., & O'Mahony, M. (2011). Travel to work in Dublin. The potential impacts of electric vehicles on climate change and urban air quality. *Transportation Research Part D: Transport and Environment*, *16*(2), 188–193. https://doi.org/10.1016/j.trd.2010.09.006

Burchart-Korol, D., Pustejovska, P., Blaut, A., Jursová, S., & Korol, J. (2018). Comparative life cycle assessment of current and future electricity generation systems in the Czech Republic and Poland. *The International Journal of Life Cycle Assessment*. https://doi.org/10.1007/s11367-018-1450-z

Camus, C., Farias, T., & Esteves, J. (2011). Potential impacts assessment of plug-in electric vehicles on the Portuguese energy market. *Energy Policy*, *39*(10), 5883–5897. https://doi.org/10.1016/j.enpol.2011.06.042

Cao, Z., Hashinokuchi, M., Doi, T., & Inaba, M. (2019). Improved Cycle Performance of LiNi0.8Co0.1Mn0.1O2 Positive Electrode Material in Highly Concentrated LiBF4/DMC. *Journal of The Electrochemical Society*, *166*(2), A82. https://doi.org/10.1149/2.0291902jes

Cerdas, F., Andrew, S., Thiede, S., & Herrmann, C. (2018). Environmental Aspects of the Recycling of Lithium-Ion Traction Batteries. In A. Kwade & J. Diekmann (Eds.), *Recycling of Lithium-Ion Batteries: The LithoRec Way* (pp. 267–288). Springer International Publishing. https://doi.org/10.1007/978-3-319-70572-9_16

Chandran, V., Patil, C. K., Karthick, A., Ganeshaperumal, D., Rahim, R., & Ghosh, A. (2021). State of Charge Estimation of Lithium-Ion Battery for Electric Vehicles Using Machine Learning Algorithms. *World Electric Vehicle Journal*, *12*(1), 38. https://doi.org/10.3390/wevj12010038

Cheng, S. H.-S., He, K.-Q., Liu, Y., Zha, J.-W., Kamruzzaman, M., Ma, R. L.-W., Dang, Z.-M., Li, R. K. Y., & Chung, C. Y. (2017). Electrochemical performance of all-solid-state lithium batteries using inorganic lithium garnets particulate reinforced PEO/LiClO4 electrolyte. *Electrochimica Acta*, *253*, 430–438. https://doi.org/10.1016/j.electacta.2017.08.162

Choi, W., & Song, H. H. (2018). Well-to-wheel greenhouse gas emissions of battery electric vehicles in countries dependent on the import of fuels through maritime transportation: A South Korean case study. *Applied Energy*, *230*, 135–147. https://doi.org/10.1016/j.apenergy.2018.08.092

Choma, E. F., & Ugaya, C. M. L. (2017). Environmental impact assessment of increasing electric vehicles in the Brazilian fleet. *Journal of Cleaner Production*, *152*, 497–507. https://doi.org/10.1016/j.jclepro.2015.07.091

Ciez, R. E., & Whitacre, J. F. (2019). Examining different recycling processes for lithium-ion batteries. *Nature Sustainability*, *2*(2), 148–156. https://doi.org/10.1038/s41893-019-0222-5

Classification and Definition of Vehicles | UNECE. (n.d.). Retrieved August 28, 2021, from https://unece.org/classification-and-definition-vehicles

Couper, J. R., Penney, W. R., Fair, J. R., & Walas, S. M. (2005). *Chemical process equipment: Selection and design*. Gulf professional publishing.

Curry, C. (2017). Lithium-ion battery costs and market. *Bloomberg New Energy Finance*, *5*.

Dai, Q., Kelly, J. C., Gaines, L., & Wang, M. (2019). Life Cycle Analysis of Lithium-Ion Batteries for Automotive Applications. *Batteries*, *5*(2), 48. https://doi.org/10.3390/batteries5020048

Deng, Z., Hu, X., Lin, X., Xu, L., Che, Y., & Hu, L. (2021). General Discharge Voltage Information Enabled Health Evaluation for Lithium-Ion Batteries. *IEEE/ASME Transactions on Mechatronics*, *26*(3), 1295–1306. https://doi.org/10.1109/TMECH.2020.3040010

Diekmann, J., Hanisch, C., Froböse, L., Schälicke, G., Loellhoeffel, T., Fölster, A.-S., & Kwade, A. (2016). Ecological Recycling of Lithium-Ion Batteries from Electric Vehicles with Focus on Mechanical

Processes. *Journal of The Electrochemical Society*, *164*(1), A6184. https://doi.org/10.1149/2.0271701jes

Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives (Text with EEA relevance), 32008L0098, EP, CONSIL, OJ L 312 (2008). http://data.europa.eu/eli/dir/2008/98/oj/eng

Dolganova, I., Rödl, A., Bach, V., Kaltschmitt, M., & Finkbeiner, M. (2020). A Review of Life Cycle Assessment Studies of Electric Vehicles with a Focus on Resource Use. *Resources*, *9*(3), 32. https://doi.org/10.3390/resources9030032

Dorella, G., & Mansur, M. B. (2007). A study of the separation of cobalt from spent Li-ion battery residues. *Journal of Power Sources*, *170*(1), 210–215. https://doi.org/10.1016/j.jpowsour.2007.04.025

Dunn, J. B., Gaines, L., Kelly, J. C., James, C., & Gallagher, K. G. (2014). The significance of Li-ion batteries in electric vehicle life-cycle energy and emissions and recycling's role in its reduction. *Energy & Environmental Science*, *8*(1), 158–168. https://doi.org/10.1039/C4EE03029J

Dunn, J. B., Gaines, L., Sullivan, J., & Wang, M. Q. (2012). Impact of recycling on cradle-to-gate energy consumption and greenhouse gas emissions of automotive lithium-ion batteries. *Environmental Science*

& *Technology*, *46*(22), 12704–12710. https://doi.org/10.1021/es302420z

Dunn, J., Slattery, M., Kendall, A., Ambrose, H., & Shen, S. (2021). Circularity of Lithium-Ion Battery Materials in Electric Vehicles. *Environmental Science & Technology*, *55*(8), 5189–5198. https://doi.org/10.1021/acs.est.0c07030

Dutta, D., Panda, R., Kumari, A., Goel, S., & Jha, M. K. (2018). Sustainable recycling process for metals recovery from used printed circuit boards (PCBs). *Sustainable Materials and Technologies*, *17*, e00066. https://doi.org/10.1016/j.susmat.2018.e00066

Egede, P., Dettmer, T., Herrmann, C., & Kara, S. (2015). Life Cycle Assessment of Electric Vehicles – A Framework to Consider Influencing Factors. *Procedia CIRP*, *29*, 233–238. https://doi.org/10.1016/j.procir.2015.02.185

Ellingsen, L. A.-W., Majeau-Bettez, G., Singh, B., Srivastava, A. K., Valøen, L. O., & Strømman, A. H. (2014). Life Cycle Assessment of a Lithium-Ion Battery Vehicle Pack. *Journal of Industrial Ecology*, *18*(1), 113–124. https://doi.org/10.1111/jiec.12072

Ellingsen, L. A.-W., Singh, B., & Strømman, A. H. (2016). The size and range effect: Lifecycle greenhouse gas emissions of electric vehicles. *Environmental Research Letters*, *11*(5), 054010. https://doi.org/10.1088/1748-9326/11/5/054010

Elzein, H., Dandres, T., Levasseur, A., & Samson, R. (2019). How can an optimized life cycle assessment method help evaluate the use phase of energy storage systems? *Journal of Cleaner Production*, *209*, 1624–1636. https://doi.org/10.1016/j.jclepro.2018.11.076

EverBatt | Argonne National Laboratory. (n.d.). Retrieved June 7, 2021, from https://www.anl.gov/egs/everbatt

Faria, R., Marques, P., Garcia, R., Moura, P., Freire, F., Delgado, J., & de Almeida, A. (2014). Primary and secondary use of electric mobility batteries from a life cycle perspective. *Journal of Power Sources*, *262*, 169–177. https://doi.org/10.1016/j.jpowsour.2014.03.092

Faria, R., Marques, P., Moura, P., Freire, F., Delgado, J., & de Almeida, A. T. (2013). Impact of the electricity mix and use profile in the life-cycle assessment of electric vehicles. *Renewable and Sustainable Energy Reviews*, *24*, 271–287. https://doi.org/10.1016/j.rser.2013.03.063

Faria, R., Moura, P., Delgado, J., & de Almeida, A. T. (2012). A sustainability assessment of electric vehicles as a personal mobility system. *Energy Conversion and Management*, *61*, 19–30. https://doi.org/10.1016/j.enconman.2012.02.023

Ferreira, D. A., Prados, L. M. Z., Majuste, D., & Mansur, M. B. (2009). Hydrometallurgical separation of aluminium, cobalt, copper and lithium from spent Li-ion batteries. *Journal of Power Sources*, *187*(1), 238–246. https://doi.org/10.1016/j.jpowsour.2008.10.077

Fontana, D., Pietrantonio, M., Pucciarmati, S., Rao, C., & Forte, F. (2019). A comprehensive characterization of End-of-Life mobile phones for secondary material resources identification. *Waste Management*, *99*, 22–30. https://doi.org/10.1016/j.wasman.2019.08.011

Frischknech, R. (2011, April 6). *Life Cycle Assessment of Driving Electric Cars and Scope Dependent LCA models. Life cycle assessment of electric cars—PDF Free Download*. https://docplayer.net/130868190-Outline-life-cycle-assessment-of-driving-electric-cars-and-scope-dependent-lca-models-life-cycle-assessment-of-electric-cars.html

Gaines, L. (2018). Lithium-ion battery recycling processes: Research towards a sustainable course. *Sustainable Materials and Technologies*, *17*, e00068. https://doi.org/10.1016/j.susmat.2018.e00068

Gaines, L., & Cuenca, R. (2000). *Costs of Lithium-Ion Batteries for Vehicles*. https://doi.org/10.2172/761281

Gaines, L., Sullivan, J., & Burnham, A. (2011). Life-Cycle Analysis for Lithium-Ion Battery Production and Recycling. *Transportation Research Board 90th Annual Meeting, Washington, DC*.

Gao, W., Zhang, X., Hong, Z., Cao, H., Zhang, Y., & Sun, Z. H. I. (2017). Lithium Carbonate Recovery from Cathode Scrap of Spent Lithium-Ion Battery: A Closed-Loop Process. *Environmental Science & Technology*, *51*. https://doi.org/10.1021/acs.est.6b03320

Gerssen-Gondelach, S. (2012). Performance of batteries for electric vehicles on short and longer term. *Journal of Power Sources*, *212*, 111–129. https://doi.org/10.1016/j.jpowsour.2012.03.085

Gies, E. (2015). Recycling: Lazarus batteries. *Nature*, *526*(7575), S100–S101. https://doi.org/10.1038/526S100a

Girardi, P., Gargiulo, A., & Brambilla, P. C. (2015). A comparative LCA of an electric vehicle and an internal combustion engine vehicle using the appropriate power mix: The Italian case study. *The International Journal of Life Cycle Assessment*, *20*(8), 1127–1142. https://doi.org/10.1007/s11367-015-0903-x

Golmohammadzadeh, R., Faraji, F., & Rashchi, F. (2018). Recovery of lithium and cobalt from spent lithium ion batteries (LIBs) using organic acids as leaching reagents: A review. *Resources, Conservation and Recycling*, *136*, 418–435. https://doi.org/10.1016/j.resconrec.2018.04.024

Goodenough, J. B., & Park, K.-S. (2013). The Li-ion rechargeable battery: A perspective. *Journal of the American Chemical Society*, *135*(4), 1167–1176. https://doi.org/10.1021/ja3091438

Greenhouse gas emissions from transport in Europe—European Environment Agency. (n.d.). [Indicator Assessment]. Retrieved May 17, 2021, from https://www.eea.europa.eu/data-and-maps/indicators/transport-emissions-of-greenhouse-gases-7/assessment

Habib, S., Kamran, M., & Rashid, U. (2015). Impact analysis of vehicle-to-grid technology and charging strategies of electric vehicles on distribution networks – A review. *Journal of Power Sources*, *277*, 205–214. https://doi.org/10.1016/j.jpowsour.2014.12.020

Hammond, G. P., & Hazeldine, T. (2015). Indicative energy technology assessment of advanced rechargeable batteries. *Applied Energy*, *138*, 559–571. https://doi.org/10.1016/j.apenergy.2014.10.037

Hamut, H., Dincer, I., & Naterer, G. (2014). An Exergoeconomic Analysis of Hybrid Electric Vehicle Thermal Management Systems. *Journal of Cleaner Production*, *67*, 187–196. https://doi.org/10.1115/1.4025419

Hausbrand, R., Cherkashinin, G., Ehrenberg, H., Gröting, M., Albe, K., Hess, C., & Jaegermann, W. (2015). Fundamental degradation mechanisms of layered oxide Li-ion battery cathode materials: Methodology, insights and novel approaches. *Materials Science and Engineering: B*, *192*, 3–25. https://doi.org/10.1016/j.mseb.2014.11.014

Hauschild, M. Z. (2018). Introduction to LCA Methodology. In M. Z. Hauschild, R. K. Rosenbaum, & S. I. Olsen (Eds.), *Life Cycle Assessment: Theory and Practice* (pp. 59–66). Springer International Publishing. https://doi.org/10.1007/978-3-319-56475-3_6

Hawkins, T., Singh, B., Majeau-Bettez, G., & Strømman, A. (2013). Comparative Environmental Life Cycle Assessment of Conventional and Electric Vehicles. *Journal of Industrial Ecology*, *17*. https://doi.org/10.1111/j.1530-9290.2012.00532.x

He, L.-P., Sun, S.-Y., Song, X.-F., & Yu, J.-G. (2017). Leaching process for recovering valuable metals from the LiNi1/3Co1/3Mn1/3O2 cathode of lithium-ion batteries. *Waste Management*, *64*, 171–181. https://doi.org/10.1016/j.wasman.2017.02.011

Held, M. (n.d.). *Fraunhofer System Research for E-Mobility (FSEM) Current LCA results and need for further research—PDF Free Download*. Retrieved December 10, 2019, from https://docplayer.net/14033601-Fraunhofer-system-research-for-e-mobility-fsem-current-lca-results-and-need-for-further-research.html

Helmers, E., Dietz, J., & Weiss, M. (2020). Sensitivity Analysis in the Life-Cycle Assessment of Electric vs. Combustion Engine Cars under Approximate Real-World Conditions. *Sustainability*, *12*(3), 1241. https://doi.org/10.3390/su12031241

Hernandez, M., Messagie, M., De Gennaro, M., & Van Mierlo, J. (2017). Resource depletion in an electric vehicle powertrain using different LCA impact methods. *Resources, Conservation and Recycling*, *120*. https://doi.org/10.1016/j.resconrec.2016.11.005

Hischier, R., Classen, M., Lehmann, M., & Scharnhorst, W. (2007). *Life cycle inventories of electric and electronic equipment: Production, use and disposal*.

Hofmann, J., Guan, D., Chalvatzis, K., & Huo, H. (2016). Assessment of electrical vehicles as a successful driver for reducing CO2 emissions in China. *Applied Energy*, *184*, 995–1003. https://doi.org/10.1016/j.apenergy.2016.06.042

Hu, J., Zhang, J., Li, H., Chen, Y., & Wang, C. (2017). A promising approach for the recovery of high value-added metals from spent lithium-ion batteries. *Journal of Power Sources*, *351*, 192–199. https://doi.org/10.1016/j.jpowsour.2017.03.093

Huijbregts, M., Steinmann, Z., Elshout, P., Stam, G., Verones, F., Vieira, M., Hollander, A., & Zijp, M. (2017). ReCiPe 2016 v1. 1-A harmonized life cycle impact assessment method at midpoint and endpoint level: Report I. Characterization (No. RIVM Report 2016-0104a). *National Institute for Public Health and the Environment, Bilthoven, The Netherlands*.

Iannicelli-Zubiani, E. M., Giani, M. I., Recanati, F., Dotelli, G., Puricelli, S., & Cristiani, C. (2017). Environmental impacts of a hydrometallurgical process for electronic waste treatment: A life cycle assessment case study. *Journal of Cleaner Production*, *140*, 1204–1216. https://doi.org/10.1016/j.jclepro.2016.10.040

International Standard Organization. (2006a). *ISO 14040: Environmental Management—Life Cycle Assessment—Principles and Framework*. https://books.google.it/books?id=P-0aMwAACAAJ

International Standard Organization. (2006b). *ISO 14044: Environmental management—Life cycle assessment—Requirements and guidelines*. https://books.google.it/books?id=P-0aMwAACAAJ

Ioakimidis, C. S., Murillo-Marrodán, A., Bagheri, A., Thomas, D., & Genikomsakis, K. N. (2019). Life Cycle Assessment of a Lithium Iron Phosphate (LFP) Electric Vehicle Battery in Second Life Application Scenarios. *Sustainability*, *11*(9), 1–14.

Ishihara, K., Kihira, N., Terada, N., & Iwahori, T. (2002). Environmental burdens of large lithium-ion batteries developed in a Japanese national project. *Central Research Institute of Electric Power Industry, Japan*.

Italian Ministry of Labor and Social Policies. (n.d.). *Chimica—Chimica e Affini—Industria—Chimica—Industria—IlCCNL.it*. Retrieved July 5, 2021, from https://ilccnl.it/ccnl_Chimica_-_Industria.html

Jha, M. K., Kumari, A., Jha, A. K., Kumar, V., Hait, J., & Pandey, B. D. (2013). Recovery of lithium and cobalt from waste lithium ion batteries of mobile phone. *Waste Management*, *33*(9), 1890–1897. https://doi.org/10.1016/j.wasman.2013.05.008

Jursova, S., Burchart-Korol, D., & Pustejovska, P. (2019). Carbon Footprint and Water Footprint of Electric Vehicles and Batteries Charging in View of Various Sources of Power Supply in the Czech Republic.

Environments, *6*(3), 38. https://doi.org/10.3390/environments6030038

Kallitsis, E., Korre, A., Kelsall, G., Kupfersberger, M., & Nie, Z. (2020). Environmental life cycle assessment of the production in China of lithium-ion batteries with nickel-cobalt-manganese cathodes utilising novel electrode chemistries. *Journal of Cleaner Production*, *254*, 120067. https://doi.org/10.1016/j.jclepro.2020.120067

Kawamoto, R., Mochizuki, H., Moriguchi, Y., Nakano, T., Motohashi, M., Sakai, Y., & Inaba, A. (2019). Estimation of CO2 Emissions of Internal Combustion Engine Vehicle and Battery Electric Vehicle Using LCA. *Sustainability*, *11*, 2690. https://doi.org/10.3390/su11092690

Keersemaker, M. (2020). Critical Raw Materials. *Suriname Revisited: Economic Potential of Its Mineral Resources*, 69–82. https://doi.org/10.1007/978-3-030-40268-6_9

Koiwanit, J., & Hamontree, C. (2018). A Comparative of Life Cycle Assessment of a Conventional Van and a Battery Electric Van for an Online Shopping System in Thailand. *IOP Conference Series: Earth and Environmental Science*, *150*, 012030. https://doi.org/10.1088/1755-1315/150/1/012030

Kushnir, D., & Sandén, B. (2011). Multi-level energy analysis of emerging technologies: A case study in new materials for lithium ion batteries.

Journal of Cleaner Production - J CLEAN PROD, *19*, 1405–1416. https://doi.org/10.1016/j.jclepro.2011.05.006

Lastoskie, C. M., & Dai, Q. (2015). Comparative life cycle assessment of laminated and vacuum vapor-deposited thin film solid-state batteries. *Journal of Cleaner Production*, *C*(91), 158–169. https://doi.org/10.1016/j.jclepro.2014.12.003

Li, B., Gao, X., Li, J., & Yuan, C. (2014). Life Cycle Environmental Impact of High-Capacity Lithium Ion Battery with Silicon Nanowires Anode for Electric Vehicles. *Environmental Science & Technology*, *48*(5), 3047–3055. https://doi.org/10.1021/es4037786

Liang, Y., Su, J., Xi, B., Yu, Y., Ji, D., Sun, Y., Cui, C., & Zhu, J. (2016). Life cycle assessment of lithium-ion batteries for greenhouse gas emissions. *Resources, Conservation and Recycling*, *117*. https://doi.org/10.1016/j.resconrec.2016.08.028

Lin, C., Tang, A., Mu, H., Wang, W., & Wang, C. (2015). Aging Mechanisms of Electrode Materials in Lithium-Ion Batteries for Electric Vehicles. *Journal of Chemistry*, *2015*. https://doi.org/10.1155/2015/104673

Liu, J., Xu, H., Zhang, L., & Liu, C. T. (2020). Economic and environmental feasibility of hydrometallurgical process for recycling waste mobile phones. *Waste Management*, *111*, 41–50. https://doi.org/10.1016/j.wasman.2020.05.017

Lowe, E. A., & Evans, L. K. (1995). Industrial ecology and industrial ecosystems. *Journal of Cleaner Production*, *3*(1), 47–53. https://doi.org/10.1016/0959-6526(95)00045-G

Luo, J., & Zeng, Y. (2017). Research on Development of the Lithium-Ion Battery. *Chemical Engineering Transactions*, *62*, 97–102. https://doi.org/10.3303/CET1762017

Lutsey, N., & Hall, D. (2018). *Effects of battery manufacturing on electric vehicle life-cycle greenhouse gas emissions*.

Ma, Y., Houghton, T., Cruden, A., & Infield, D. (2012). Modeling the Benefits of Vehicle-to-Grid Technology to a Power System. *IEEE Transactions on Power Systems*, *27*(2), 1012–1020. https://doi.org/10.1109/TPWRS.2011.2178043

Majeau-Bettez, G., Hawkins, T., & Strømman, A. (2011). Life Cycle Environmental Assessment of Lithium-Ion and Nickel Metal Hydride Batteries for Plug-In Hybrid and Battery Electric Vehicles. *Environmental Science & Technology*, *45*, 5454. https://doi.org/10.1021/es2015082

Makuza, B., Tian, Q., Guo, X., Chattopadhyay, K., & Yu, D. (2021). Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. *Journal of Power Sources*, *491*, 229622. https://doi.org/10.1016/j.jpowsour.2021.229622

Marques, P., Garcia, R., Kulay, L., & Freire, F. (2019). Comparative life cycle assessment of lithium-ion batteries for electric vehicles addressing capacity fade. *Journal of Cleaner Production*, *229*. https://doi.org/10.1016/j.jclepro.2019.05.026

Mayyas, A., Steward, D., & Mann, M. (2019). The case for recycling: Overview and challenges in the material supply chain for automotive li-ion batteries. *Sustainable Materials and Technologies*, *19*, e00087. https://doi.org/10.1016/j.susmat.2018.e00087

Mcmanus, M. (2012). Environmental consequences of the use of batteries in low carbon systems: The impact of battery production. *Applied Energy*, *93*. https://doi.org/10.1016/j.apenergy.2011.12.062

Menikpura, S. N. M., Santo, A., & Hotta, Y. (2014). Assessing the climate co-benefits from Waste Electrical and Electronic Equipment (WEEE) recycling in Japan. *Journal of Cleaner Production*, *74*, 183–190. https://doi.org/10.1016/j.jclepro.2014.03.040

Notter, D., Gauch, M., Widmer, R., Wäger, P., Stamp, A., Zah, R., & Althaus, H.-J. (2010). Contribution of Li-Ion Batteries to the Environmental Impact of Electric Vehicles. *Environmental Science & Technology*, *44*. https://doi.org/10.1021/es1029156

Onat, N. C., Kucukvar, M., & Tatari, O. (2018). Well-to-wheel water footprints of conventional versus electric vehicles in the United States: A state-based comparative analysis. *Journal of Cleaner*

Production, *204*, 788–802. https://doi.org/10.1016/j.jclepro.2018.09.010

Onwunta, O. E. K. (2021). *Plug In Hybrid Electric Vehicles and Electric Loading in the African Transportation Solution* (SSRN Scholarly Paper ID 3901330). Social Science Research Network. https://doi.org/10.2139/ssrn.3901330

Pero, F. D., Delogu, M., & Pierini, M. (2018). Life Cycle Assessment in the automotive sector: A comparative case study of Internal Combustion Engine (ICE) and electric car. *Procedia Structural Integrity*, *12*, 521–537. https://doi.org/10.1016/j.prostr.2018.11.066

Peters, M. S., Timmerhaus, K. D., & West, R. E. (2003). *Plant design and economics for chemical engineers* (Vol. 4). McGraw-Hill New York.

Petrauskienė, K., Skvarnavičiūtė, M., & Dvarionienė, J. (2020). Comparative environmental life cycle assessment of electric and conventional vehicles in Lithuania. *Journal of Cleaner Production*, *246*, 119042. https://doi.org/10.1016/j.jclepro.2019.119042

Pinna, E. G., Ruiz, M. C., Ojeda, M. W., & Rodriguez, M. H. (2017). Cathodes of spent Li-ion batteries: Dissolution with phosphoric acid and recovery of lithium and cobalt from leach liquors. *Hydrometallurgy*, *167*, 66–71. https://doi.org/10.1016/j.hydromet.2016.10.024

Qiao, Q., Zhao, F., Liu, Z., Jiang, S., & Hao, H. (2017). Cradle-to-gate greenhouse gas emissions of battery electric and internal combustion engine vehicles in China. *Applied Energy*, *204*, 1399–1411. https://doi.org/10.1016/j.apenergy.2017.05.041

Rahman, A. (2017). Lithium battery recycling management and policy. *Int. J. Energy Technology and Policy*, *13*, 278–291. https://doi.org/10.1504/IJETP.2017.084497

Raugei, M., & Winfield, P. (2018). Prospective LCA of the production and EoL recycling of a novel type of Li-ion battery for electric vehicles. *Journal of Cleaner Production*, *213*. https://doi.org/10.1016/j.jclepro.2018.12.237

ReCiPe | PRé Sustainability. (n.d.). Retrieved March 1, 2020, from https://www.pre-sustainability.com/recipe

Rezaee, S., Farjah, E., & Khorramdel, B. (2013). Probabilistic Analysis of Plug-In Electric Vehicles Impact on Electrical Grid Through Homes and Parking Lots. *IEEE Transactions on Sustainable Energy*, *4*(4), 1024–1033. https://doi.org/10.1109/TSTE.2013.2264498

Richa, K., Babbitt, C., & Gaustad, G. (2017). Eco-Efficiency Analysis of a Lithium-Ion Battery Waste Hierarchy Inspired by Circular Economy: LIB Waste Hierarchy Inspired by Circular Economy. *Journal of Industrial Ecology*, *21*. https://doi.org/10.1111/jiec.12607

Rupp, M., Handschuh, N., Rieke, C., & Kuperjans, I. (2019). Contribution of country-specific electricity mix and charging time to environmental impact of battery electric vehicles: A case study of electric buses in

Germany. *Applied Energy*, *237*, 618–634. https://doi.org/10.1016/j.apenergy.2019.01.059

Rydh, C. J., & Sandén, B. A. (2005). Energy analysis of batteries in photovoltaic systems. Part I: Performance and energy requirements. *Energy Conversion and Management*, *46*(11), 1957–1979. https://doi.org/10.1016/j.enconman.2004.10.003

Sagaria, S., Moreira, A., Margarido, F., & Baptista, P. (2021). From Microcars to Heavy-Duty Vehicles: Vehicle Performance Comparison of Battery and Fuel Cell Electric Vehicles. *Vehicles*, *3*(4), 691–720. https://doi.org/10.3390/vehicles3040041

Samaras, C., & Meisterling, K. (2008). Life Cycle Assessment of Greenhouse Gas Emissions from Plug-in Hybrid Vehicles: Implications for Policy. *Environmental Science & Technology*, *42*, 3170–3176. https://doi.org/10.1021/es702178s

Sambamurthy, S., Raghuvanshi, S., & Sangwan, K. S. (2021). Environmental impact of recycling spent lithium-ion batteries. *Procedia CIRP*, *98*, 631–636. https://doi.org/10.1016/j.procir.2021.01.166

Sarath, P., Bonda, S., Mohanty, S., & Nayak, S. K. (2015). Mobile phone waste management and recycling: Views and trends. *Waste Management*, *46*, 536–545. https://doi.org/10.1016/j.wasman.2015.09.013

Schmuch, R., Wagner, R., Hörpel, G., Placke, T., & Winter, M. (2018). Performance and cost of materials for lithium-based rechargeable automotive batteries. *Nature Energy*, *3*(4), 267–278. https://doi.org/10.1038/s41560-018-0107-2

Simon, B., & Weil, M. (2013). Analysis of materials and energy flows of different lithium ion traction batteries. *Revue de Métallurgie*, *1*, 65–76. https://doi.org/10.1051/metal/2013056

Sommerville, R., Zhu, P., Rajaeifar, M. A., Heidrich, O., Goodship, V., & Kendrick, E. (2021). A qualitative assessment of lithium ion battery recycling processes. *Resources, Conservation and Recycling*, *165*, 105219. https://doi.org/10.1016/j.resconrec.2020.105219

Sonoc, A., Jeswiet, J., & Soo, V. K. (2015). Opportunities to Improve Recycling of Automotive Lithium Ion Batteries. *Procedia CIRP*, *29*, 752–757. https://doi.org/10.1016/j.procir.2015.02.039

Souza, L., Lora, E., Palacio, J., Rocha, M., Renó, M., & Venturini, O. (2018). Comparative environmental life cycle assessment of conventional vehicles with different fuel options, plug-in hybrid and electric vehicles for a sustainable transportation system in Brazil. *Journal of Cleaner Production*, *203*. https://doi.org/10.1016/j.jclepro.2018.08.236

Sullivan, J., Burnham, A., & Wang, M. (2010). Energy-consumption and carbon-emission analysis of vehicle and component manufacturing. *Journal of Industrial Ecology*, *17*. https://doi.org/10.2172/993394

Sun, L., & Qiu, K. (2011). Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries. *Journal of Hazardous Materials*, *194*, 378–384. https://doi.org/10.1016/j.jhazmat.2011.07.114

Sun, Q., Yang, L., Huang, S., Xu, Z., Li, Y., & Wang, W. (2019a). Synergistic solvent extraction of nickel by 2-hydroxy-5-nonylacetophenone oxime mixed with neodecanoic acid and bis(2-ethylhexyl) phosphoric acid: Stoichiometry and structure investigation. *Minerals Engineering*, *132*, 284–292. https://doi.org/10.1016/j.mineng.2018.11.049

Sun, Q., Yang, L., Huang, S., Xu, Z., Li, Y., & Wang, W. (2019b). Synergistic solvent extraction of nickel by 2-hydroxy-5-nonylacetophenone oxime mixed with neodecanoic acid and bis(2-ethylhexyl) phosphoric acid: Stoichiometry and structure investigation. *Minerals Engineering*, *132*, 284–292. https://doi.org/10.1016/j.mineng.2018.11.049

Swiss Centre for Life Cycle Inventories. Ecoinvent 2017—Ecoinvent Database v3.6, 2019. (n.d.). Retrieved March 26, 2020, from https://www.ecoinvent.org/

Tadesse, B., Makuei, F., Albijanic, B., & Dyer, L. (2019). The beneficiation of lithium minerals from hard rock ores: A review. *Minerals Engineering*, *131*, 170–184. https://doi.org/10.1016/j.mineng.2018.11.023

Tagliaferri, C., Evangelisti, S., Acconcia, F., Domenech, T., Ekins, P., Barletta, D., & Lettieri, P. (2016). Life cycle assessment of future electric and hybrid vehicles: A cradle-to-grave systems engineering approach. *Chemical Engineering Research and Design*, *112*, 298–309. https://doi.org/10.1016/j.cherd.2016.07.003

Tolomeo, R., De Feo, G., Adami, R., & Sesti Osséo, L. (2020). Application of Life Cycle Assessment to Lithium Ion Batteries in the Automotive Sector. *Sustainability*, *12*(11), 4628. https://doi.org/10.3390/su12114628

Tornheim, A., Sharifi-Asl, S., Garcia, J. C., Bareño, J., Iddir, H., Shahbazian-Yassar, R., & Zhang, Z. (2019). Effect of electrolyte composition on rock salt surface degradation in NMC cathodes during high-voltage potentiostatic holds. *Nano Energy*, *55*, 216–225. https://doi.org/10.1016/j.nanoen.2018.10.065

Tran, M.-K., Bhatti, A., Vrolyk, R., Wong, D., Panchal, S., Fowler, M., & Fraser, R. (2021). A Review of Range Extenders in Battery Electric Vehicles: Current Progress and Future Perspectives. *World Electric Vehicle Journal*, *12*(2), 54. https://doi.org/10.3390/wevj12020054

Troy, S., Schreiber, A., Reppert, T., Gehrke, H.-G., Finsterbusch, M., Uhlenbruck, S., & Stenzel, P. (2016). Life Cycle Assessment and resource analysis of all-solid-state batteries. *Applied Energy*, *169*, 757–767. https://doi.org/10.1016/j.apenergy.2016.02.064

Un-Noor, F., Padmanaban, S., Mihet-Popa, L., Mollah, M. N., & Hossain, E. (2017). A Comprehensive Study of Key Electric Vehicle (EV) Components, Technologies, Challenges, Impacts, and Future Direction of Development. *Energies*, *10*(8), 1217. https://doi.org/10.3390/en10081217

Unterreiner, L., Jülch, V., & Reith, S. (2016). Recycling of Battery Technologies – Ecological Impact Analysis Using Life Cycle Assessment (LCA). *Energy Procedia*, *99*, 229–234. https://doi.org/10.1016/j.egypro.2016.10.113

Van den Bossche, P., Vergels, F., Van Mierlo, J., Matheys, J., & Van Autenboer, W. (2006). SUBAT: An assessment of sustainable battery technology. *Journal of Power Sources*, *162*, 913–919. https://doi.org/10.1016/j.jpowsour.2005.07.039

Van Mierlo, J., Boureima, F., Messagie, M., Sergeant, N., Govaerts, L., Denys, T., Michiels, H., Vernaillen, S., Schrooten, L., & Beckx, C. (2011). CLEAN VEHICLE RESEARCH: LCA AND POLICY MEASURES «CLEVER». *Final Report of the Project Funded by the Belgian Science Policy*.

Vandepaer, L., Cloutier, J., & Amor, B. (2017). Environmental impacts of Lithium Metal Polymer and Lithium-ion stationary batteries. *Renewable and Sustainable Energy Reviews*, *78*, 46–60. https://doi.org/10.1016/j.rser.2017.04.057

Volvo Recharge – our range of pure electric and plug-in hybrid cars. (n.d.). Volvo Cars. Retrieved January 2, 2022, from https://www.volvocars.com/intl/v/cars/recharge

Wang, F., Deng, Y., & Yuan, C. (2020). Life cycle assessment of lithium oxygen battery for electric vehicles. *Journal of Cleaner Production*, *264*, 121339. https://doi.org/10.1016/j.jclepro.2020.121339

Wang, L., Wu, H., Hu, Y., Yu, Y., & Huang, K. (2019). Environmental Sustainability Assessment of Typical Cathode Materials of Lithium-Ion Battery Based on Three LCA Approaches. *Processes*, *7*, 83. https://doi.org/10.3390/pr7020083

Wang, X., Gaustad, G., Babbitt, C. W., & Richa, K. (2014). Economies of scale for future lithium-ion battery recycling infrastructure. *Resources, Conservation and Recycling*, *83*, 53–62. https://doi.org/10.1016/j.resconrec.2013.11.009

Weldon, P., Morrissey, P., & O’Mahony, M. (2016). Environmental impacts of varying electric vehicle user behaviours and comparisons to internal combustion engine vehicle usage – An Irish case study.

Journal of Power Sources, *319*, 27–38. https://doi.org/10.1016/j.jpowsour.2016.04.051

Wilken, D., Oswald, M., Draheim, P., Pade, C., Brand, U., & Vogt, T. (2020). Multidimensional assessment of passenger cars: Comparison of electric vehicles with internal combustion engine vehicles. *Procedia CIRP*, *90*, 291–296. https://doi.org/10.1016/j.procir.2020.01.101

Winslow, K. M., Laux, S. J., & Townsend, T. G. (2018). A review on the growing concern and potential management strategies of waste lithium-ion batteries. *Resources, Conservation and Recycling*, *129*, 263–277. https://doi.org/10.1016/j.resconrec.2017.11.001

Woo, J., Choi, H., & Ahn, J. (2017). Well-to-wheel analysis of greenhouse gas emissions for electric vehicles based on electricity generation mix: A global perspective. *Transportation Research Part D: Transport and Environment*, *51*, 340–350. https://doi.org/10.1016/j.trd.2017.01.005

Yin, R., Hu, S., & Yang, Y. (2019). Life cycle inventories of the commonly used materials for lithium-ion batteries in China. *Journal of Cleaner Production*, *227*, 960–971. https://doi.org/10.1016/j.jclepro.2019.04.186

Yongxia, Y., Hong, Z., Zhao, C., Cao, H., Ning, P., Zhang, Y., Jin, W., & Sun, Z. H. I. (2017). A Closed-Loop Process for Selective Metal Recovery from Spent Lithium Iron Phosphate Batteries through

Mechanochemical Activation. *ACS Sustainable Chemistry & Engineering*, *5*. https://doi.org/10.1021/acssuschemeng.7b01914

Yuan, C., Cao, H., Shen, K., Deng, Y., Zeng, D., Dong, Y., & Hauschild, M. (2021). Water-based manufacturing of lithium ion battery for life cycle impact mitigation. *CIRP Annals*. https://doi.org/10.1016/j.cirp.2021.04.038

Zackrisson, M. (2016). *Life cycle assessment of long life lithiumelectrode for electric vehicle batteries: 5Ah cell*.

Zackrisson, M., Avellán, L., & Orlenius, J. (2010). Life cycle assessment of lithium-ion batteries for plug-in hybrid electric vehicles – Critical issues. *Journal of Cleaner Production*, *18*, 1519–1529. https://doi.org/10.1016/j.jclepro.2010.06.004

Zhao, G. (2017). *Reuse and recycling of lithium-Ion power batteries*. Wiley Online Library.

Zhao, S., & You, F. (2019). Comparative Life-Cycle Assessment of Li-Ion Batteries through Process-Based and Integrated Hybrid Approaches. *ACS Sustainable Chemistry & Engineering*, *7*. https://doi.org/10.1021/acssuschemeng.8b05902

Zheng, M., Tang, H., Li, L., Hu, Q., Zhang, L., Xue, H., & Pang, H. (2018). Hierarchically Nanostructured Transition Metal Oxides for Lithium-Ion Batteries. *Advanced Science*, *5*(3), 1700592. https://doi.org/10.1002/advs.201700592

Zhu, P., Gastol, D., Marshall, J., Sommerville, R., Goodship, V., & Kendrick, E. (2021). A review of current collectors for lithium-ion batteries. *Journal of Power Sources*, *485*, 229321. https://doi.org/10.1016/j.jpowsour.2020.229321

Appendix A

Table XV: Specific sales values and costs adopted in the economic model ("BatPaC")

Element	***Price (€/kg)***
Co	44.00
Ni	11.00
Mn	2.53
Hydrochloric Acid	0.36
Hydrogen Peroxide	0.58
Lime	0.12
Limestone	0.12
Sand	0.042
Sodium Hydroxide	0.40
Sulfuric Acid	0.06
Aluminum	1.45
Copper	5.43
Steel	0.28
Plastics	0.10

LCO	40.00
NMC(111)	21.00
NMC(532)	20.00
NMC(622)	20.60
NMC(811)	22.00
Lithium carbonate	14.73
Ni^{2+} in Ni salt/oxide	13.00
Co^{2+} in Co salt/oxide	43.00
Mn^{2+} in Mn salt	3.00
LMO	16.00
NCA	21.50
LFP	14.00
Electrolyte organics	0.15
Graphite	0.20

Table XVI: Cost for fuel and electricity adopted in economic model.

Element	Price (€/kWh)
Diesel	1
Natural gas	0.0071

Electricity	0.16

Table XVII: Equipment costs and power rating parameters adopted in the economic model (Couper et al., 2005; Peters et al., 2003)

Equipment	Cost coefficient				Power consumption			Notes
	a	**b**	**c**	**adj**	**m**	**n**	**p**	
Ball mill	61000	0.69	0	1.412	34	1.006	0	(0.25 inch x 200 mesh)
Brine Soak	31862	0	0	1.412	7.5	0	0	Screw conveyor, stainless steel, 0.3048m diameter
Briquetter	16048	0	0	1.435	75	0	0	Roll type extruder
Calciner	1313832	0.512	0	0.985	5861	1	0	Incinerator, cylindrical, low-hazard feed material
Cell perforator	17636	0	0	1.435	75	0	0	Roll crusher
Conveyor	102600	0	0	0.985	15	0	0	Belt, open, short, 1 m widht and 30.5 m lenght
Crusher	106512	0	0	1.435	75	0	0	Giratory crusher
Density Separator	2760	0.96	0	1.412	75	0	0	Cyclone separator, heavy duty
Dryer	591236	0.6	0	1.412	729	1	0	Steam tube dryer, class II, 304 stainless steel
Filter press	173000	0	0	0.985	15	0	0	Plate and frame, 18.58 m^2 area, stainless 304

Froth flotation cell	131410	0.5301	0	0.985	75	0	0	Jacketed and agitated, stainless 304, atmospheric to 1.72 bar
Gas treatment	3000000	0	0	1	1000	0	0	
Granulator	29902	0.6671	0	0.985	1.361	1	-0.5806	Agglomerator, disk with motor, stainless steel 304
Hopper	38700	0	0	0.985	15	0	0	With bottom, bolted, carbon steel, 141.5842m^3 bin volume
Hydrocyclone	50300	0	0	0.985	75	1	0	Cyclone, wet, ceramic lined, 0.762 m diameter
Leaching tank	473892	0.4481	0	0.985	15	1	0	Mixer/settler, stainless 304, atmospheric to 1.72 bar
Mixin (g tank	473892	0.4481	0	0.985	15	1	0	Mixer/settler, stainless 304, atmospheric to 1.72 bar
Oxidizer	494284	0.7601	0	0.985	5861	0	0	Incinerator, catalytic, low-hazard feed material, atmospheric pressure
Precipitation Tank	473892	0.4481	0	0.985	15	1	0	Mixer/settler, stainless 304, atmospheric to 1.72 bar
Pump	3009	0	0	1.435	3.192	1	0	Centrifugal, cast iron, 1035 kpa
Screener	1218	1	3752.8	0.985	15	1	0	DSM screen, stainless steel, with medium carbon steel wire
Skid steer	40000	0	0	1	0	0	0	Diesel-fueled
Smelter	6137979	0.48	0	0.985	0	0	0	Incinerator, rotary kiln, hazardous feed material, atmospheric pressure

The costs of the equipment have been evaluated with equation (S1):

$$\text{Cost (\$2017)} = (a*\text{capacity}^b+c)*\text{adj} \qquad \text{(S1)}$$

Where:

- Cost ($2017) is the cost of the equipment as a result of the calculations;
- capacity is the flow mass entering the equipment expressed in t/hr.
- a, b, c, adj are the coefficients reported in Table XVII and deduced from Couper et al., 2005 and Peters et al., 2003.

The power required by equipments have been evaluated with equation (S2):

$$\text{Power (kW)} = m*(\text{capacity}^n)+p \qquad \text{(S2)}$$

Where:

- Power (kW) is the power required for the equipment operation.
- capacity is the flow mass entering the equipment expressed in t/hr.
- m, n and p are the coefficients reported in Table XVII and deduced from Couper et al., 2005 and Peters et al., 2003

Table XVIII: Labor fees derived from italian national collective labour agreement (Italian Ministry of Labor and Social Policies).

Element	Labor fee (€/hr)
Specialised worker	18.97

Management figures	30.21

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