

## **Recycling Lithium Batteries, a viable industrial process**

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I declare that this document is an original work of my own authorship and that it fulfils all the requirements of the Code of Conduct and Good Practices of the *Universidade de Lisboa*.

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"What we do in life, echoes in eternity"

- Aelius Maximus Decimus Meridius

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# Abstract

Recycling lithium-ion batteries or finding a second life for any spent battery is a growing industry in renewables, due to its environmental and economic benefits. Also, with the exponential growth in demand and supply of electric vehicles, renewable energy generation and its storage are factors in the growing waste management of lithium batteries. The report presented aims to introduce the various battery recycling methods, with their benefits and economic advantages and disadvantages, as well as for second-life battery applications. In addition, different reports, and case studies were studied to know the number of materials that can be recovered, and the efficiency of the method applied through the recycling process. Finally, three projects are analysed, there are, residential storage, commercial storage, and solar farm storage, the goal of the analysis is to calculate and compare the Net Present Value for the residential process within those parameters. The analysis demonstrated that the second-life battery project would be the one with the lowest Equivalent Annual Cost, making it the most viable industrial process out of the three. In addition, it presents the higher Net Present Value for the residential storage to the remainder of the 30-year project, the recycled battery shows the most Net Present Value.

## Keywords

lithium-ion batteries, second life, battery recycling, battery reuse, battery refurbishing.

# Resumo

Reciclar baterias de ião lítio ou encontrar uma segunda vida para qualquer bateria gasta é uma indústria em crescimento na área das energias renováveis, devido aos benefícios ambientais e económicos que oferece. Além disso, com o crescimento exponencial da procura e oferta de veículos elétricos, a geração de energia renovável e o seu armazenamento são fatores no crescente mercado da gestão de resíduos de baterias de lítio. O presente relatório tem como objetivo apresentar os vários métodos de reciclagem de baterias, com os seus benefícios, vantagens e desvantagens económicas, bem como aplicações de baterias de segunda vida. Além disso, diferentes relatórios e estudos de caso foram estudados para conhecer os materiais que podem ser recuperados e a eficiência do método aplicado através do processo de reciclagem. Por fim, são analisados três projetos, nomeadamente, armazenamento residencial, armazenamento comercial, e armazenamento solar. O objetivo da análise é calcular e comparar o Valor Presente Líquido (VPL) para o projeto do armazenamento residencial, e o Custo Anual Equivalente de cada projeto, para saber qual é o processo industrial mais viável dentro desses parâmetros. A análise demonstrou que a abordagem de bateria de segunda vida é aquela com o menor Custo Anual Equivalente, sendo assim o processo industrial mais viável. No entanto, o VPL mostrou que uma bateria reciclada seria a melhor opção numa aplicação superior a 10 anos, pois agrega mais valor devido ao maior resultado do VPL.

## Palavras-chave

Baterias de litio, baterias de segunda vida, reciclagem de baterias, reutilização de baterias, recondicionamento de baterias.

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# List of Abbreviations

LMO	Lithium manganese oxide
BEV	Battery electric vehicle
NCA	Lithium nickel cobalt aluminium oxide
NMC	Lithium nickel manganese cobalt oxide
BMS	Battery management system
GHG	Greenhouse gas emissions
ICE	Internal combustion engine
PHEV	Plug-in hybrid electric vehicle
HEV	Hybrid electric vehicle
FCEV	Fuel cell electric vehicle
SOC	State of charge
Li-ion	Lithium-ion
CAGR	Compound annual growth rate
USD	United States dollars
LIBs	Lithium-ion batteries
EOL	End-of-life
SoH	State-of-health
XRD	X-ray diffraction
EIS	Electrochemical impedance spectroscopy
CAPEX	Capital expenditure
OPEX	Operational expenditure
ROI	Return of investment
DOD	Depth of discharge
NPV	Net present value
EAC	Equivalent annual cost
WACC	Weighted average cost of capital
kg	Kilograms
kWh	Kilowatt hour
LCO	Lithium cobalt oxide
LFP	Lithium iron phosphate
°C	Degree Celsius
LiFePO <sub>4</sub>	Lithium iron phosphate
CO <sub>2</sub>	Carbon dioxide
NiMH	Nickel metal hydride
Ni-Cd	Nickel cadmium
H <sub>2</sub> O	Water
Na <sub>2</sub> CO <sub>3</sub>	Sodium carbonate
Li <sub>2</sub> CO3	Lithium carbonate
$Na_2S_2O_5$	Sodium metabisulphite
$H_2O_2$	Hydrogen peroxide
NaOH	Sodium hydroxide

# **Chapter 1**

Introduction

### 1.1 Overview

Currently, not every lithium battery that has reached its end-of-life is neither recycled nor reused or refurbished. The purpose of this thesis report is to study all the recycling methods and the possibility of reusing lithium batteries in order to know a viable industrial process aiming to reduce the carbon footprint that comes from disposing of lithium batteries. Also, it provides an insight into the market size and market share of lithium batteries. Understanding the circular value chain of lithium batteries, as shown in Figure 1-1, is provided further through the thesis report to understand the many uses that an industrial process can take advantage of recycling lithium batteries.

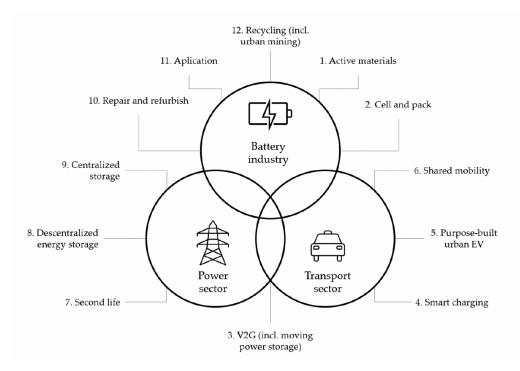


Figure 1-1. Circular battery value chain [1].

Lithium batteries have become the go-to technology when it comes to energy storage in the past few years due to their efficiency and capabilities to perform through a range of different applications; they are most commonly found in laptops, electric vehicles, residential storage, and others. This thesis report provides a more in-depth explanation of lithium battery technologies, advantages, disadvantages, waste management, and environmental issues. Also, a brief explanation on the sources of lithium can be found together with the countries with the largest lithium reserves.

In addition, recycling methods are thoroughly explained for both steps during the pre-treatment of the lithium battery, and the actual treatment through recycling. Therefore, the report offers extended understanding during the pre-treatment phase of processes like sorting, dismantling, and discharging, pyrolysis deactivation, and mechanical separation. Also, through the treatment process, methods like thermal treatment, pyrometallurgy, hydrometallurgy, and electrochemical extraction is provided. These recycling methods are expected to improve as technologies advance and become a requirement for every battery that reaches the end of its life cycle, due to the fact that only 10% of batteries are currently

being treated for recycling or reused [2].

Furthermore, the possibilities and benefits of giving lithium batteries a second life is also explained since a lithium battery reaches its end-of-life when its capacity reaches 80%. However, it can still be used for more stationary application, like energy storage as analysed in this report [3]. The European Union has been insisting on a battery passport for every battery in the market, and it will be a requirement for new manufacturers. The objective behind this is to understand what the battery has been through and how efficient and the status of the materials within them, to improve the recycling process, all of this will be provided through blockchain technology [4].

Past reports, projects, and case studies were thoroughly studied to gather the most efficient work regarding recycling, second life, and new lithium batteries. These past papers provide in-depth knowledge and information about the amount of material recovered during the recycling process and its efficiency and method deployed. Also, they provide the economic aspects of each process to understand how viable it is and how profitable it can be.

Lastly, three projects were analysed to determine and compare which is the most viable industrial process. These projects are residential storage, commercial storage, and solar farm storage, comparing them between the processes previously mentioned and gone over through the past papers, like recycling, second life, and new lithium batteries. Finally, a conclusion of the most viable industrial process is provided by calculating and comparing the Net Present Value of the residential storage project, and Equivalent Annual Cost of all three projects.

### 1.2 Solution

The main objectives of this thesis report are:

- 1. Understand the different recycling methods and which is the best one for an industrial process.
- 2. Calculate the Net Present Value and Equivalent Annual cost between a recycled, second life, and new lithium battery.
- 3. Compare and conclude which is the best method from the calculated results.

All the work was done at the Instituto Superior Tecnico (IST) and remotely, researching through articles, papers, case studies, thesis reports, and projects. Also, consulting and meeting weekly with the thesis supervisors, Professor António Quintino, and Professor Diogo Santos, in order to validate the advances and approaches taken through the work of the thesis aiming to complete the main objective previously listed.

## 1.3 Structure

This thesis report is structured in the following five chapters, as seen below.

Chapter	Description
1	Introduction of the thesis and its objective
2	State-of-the-art is explained to understand the importance of lithium batteries, their advantages and disadvantages, environmental issues, and their many different applications. Also, a complete explanation of every recycling method through pre-treatment and treatment of the lithium battery, as well as the importance of second-life batteries and how it works.
3	Theoretical development can be found where the past reports, projects, and case studies are thoroughly explained with the recycling and financial information about every application study throughout the report.
4	The results of the Net Present Value of residential storage, and Equivalent Annual Cost calculated by analysing the three different projects.
5	The main conclusion of the thesis, together with recommendations for future development, are present.

#### Table 1-1.Thesis structure.

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# **Chapter 2**

# State-of-the-Art

### 2.1 What are Lithium Batteries?

Lithium batteries are in a category of their own when compared to other batteries, because of their high energy density and low cost per cycle. They are classified into cells, modules, and packs, in addition, they are formed of four key components, which are [5]:

- Cathode: is the positive electrode that acquires electrons from the external circuit and is reduced during the electrochemical reaction. Also, it determines the voltage and capacity of the lithium battery and the source of its ions, as well as ensuring a stronger molecular bond capable of sustaining extreme charging conditions.
- Anode: allows the flow of the electric current through an external circuit, and the storage of ions in the anode when the battery is charged. This is the negative electrode and where oxidation occurs during cell discharge,
- Electrolyte: created from salts, solvents, and additives, it has the function of working as a channel of lithium ions between the anode and the cathode.
- Separator: it is essentially a physical barrier that separates the cathode and the anode. It works as an insulating material but allows ion transfer.

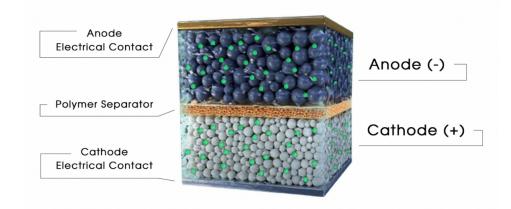


Figure 2-1.Scheme of the inside of a Lithium Battery [6].

The applications of lithium batteries are endless, it can be found in many places and objects, such as, our phone, computer, house if needed for storage, electric vehicle, and others; depending on the application lithium batteries can be found in a cylindrical, flat, or pouch cells kind of configuration. There are different lithium battery types based on cathode chemistry according to the application area. These are the following:

- Lithium cobalt oxide (LCO): used for small appliances like smartphones, laptops, tablets, and others.
- Lithium iron phosphate (LFP): useful for electric tools, medical equipment, gadgets, etc.
- Lithium manganese oxide (LMO): similar to lithium iron phosphate with the addition of battery electric vehicles (BEVs)
- Lithium nickel cobalt aluminium oxide (NCA): often found in BEVs like Tesla's Model 3 and

Model X

• Lithium nickel manganese cobalt oxide (NMC): every application mentioned above, with the addition to any other electric vehicles as well as battery storage power stations.

In addition, to have a better understanding of the comparison between each battery type previously stated, understanding its nominal voltage (V/cell), safety, maximum charge voltage (V/cell), energy density (Wh/kg), and life cycles, refer to Figure 2-2 [7].

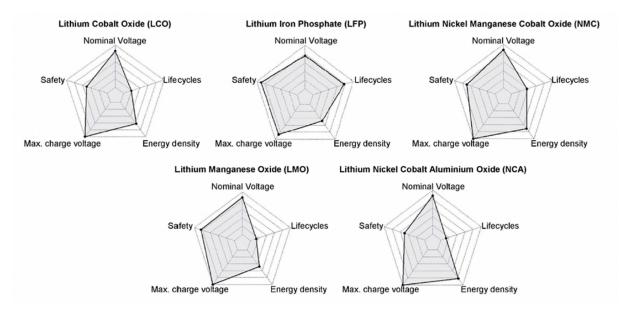


Figure 2-2. Different lithium batteries comparison [7].

However, it is a dominant battery in the renewable energy industry because of its safety, outstanding thermal stability, long cycle life, high current ratings, and tolerance. Lithium Iron Phosphate (LiFePO<sub>4</sub>) batteries are not prone to overheating, nor are they disposed to thermal runaway, ruling out any risk of over-heating or igniting when exposed to mishandling or severe weather conditions [8].

Lead-acid batteries are often replaced by lithium batteries since lithium batteries offer higher cell voltage. However, lithium batteries are unique over lead-acid batteries because an absorption charge is not needed to hold a constant voltage state. On the other hand, the discharge characteristics are also distinctive since it will maintain a much higher voltage during discharge than lead-acid batteries, typically under load [8].

Like in every application and technology, efficiency is a major factor. A lead-acid battery has an efficiency close to 80% from full to dead and back to full, better known as a round-trip. However, the round-trip of a lithium battery is 95% - 98%, making it a superior battery and offering significant improvements in energy storage for solar power systems to provide energy at night and during winter, as well as saving fuel from stopping the use of generators. Since lithium batteries do not possess absorption charge, resulting in the charge time of a lithium battery from completely dead, which does not cause any significant adverse effect, to full charge is about two hours [8].

The Battery Management System (BMS) is a system that monitors, evaluates, balances, and protects cells from being away from the known called safe operating area. It is integrated into all assemblies

because of the concern over safety and reliability of lithium batteries. The BMS protects the cells within the battery against over current, under/over voltage and temperature. A lithium battery cell has a nominal voltage of 3.2V; the cell can be permanently damaged if it falls below 2.5V or exceeds 4.2V. That is the function of the BMS; it monitors each cell to prevent them from damaging [8].

### 2.1.1 Advantages

There is a vast number of different applications where lithium batteries are taking place because of their many advantages, which are [9]:

- High energy density: this is the main advantage of LiFePO<sub>4</sub>. Systems such as mobile phones, portable computers, electric vehicles, and others that need to operate for a long period of time between charges while consuming a big amount of power, require high-density batteries, meaning they can have a high-power capacity while not being bulky.
- Self-discharge: a low self-discharge means that a lithium battery has the capability of retaining the charge longer than any other battery when left unused. Typically, it discharges around 5% in the first four hours and then approximately 1% or 2% per month.
- Low maintenance: lithium batteries rarely require maintenance. The only necessary maintenance is ensuring that every cell in the battery bank is charged equally. However, it is usually done automatically through a management system.
- Cell voltage: having a higher voltage in each cell can satisfy systems with one single cell by simplifying power management in applications such as smartphones.
- No requirement for priming: the cells found in a lithium battery bank are ready to go, meaning they do not require priming when receiving the first charge.
- Longevity: life cycles on lithium batteries are higher than any other battery, and they can be charged many times without causing any negative effect on their capacity and efficiency.

### 2.1.2 Disadvantages

Like every application and technology that has great advantages, it also has some disadvantages that are needed to be taken into consideration when wanting to choose between them. These include [9]:

- Cost: usually lithium batteries cost around 40% more to manufacture than nickel-cadmium (Ni-Cd) cells; hence when mass production is taken into account, any additional costs are a major issue.
- Transportation issues: airlines limit the number of lithium batteries that can be taken aboard the plane, meaning that its transportation is limited to ships.
- Protection: lithium batteries need protection from being overcharged, which could increase the risk of the battery exploding, and over-discharge, as they need to maintain the current within safe limits. The implementation of BMS allows the battery to be left on charge and, once fully charged, stop the supply immediately.

- Developing: even though these LiFePO<sub>4</sub> batteries have been around for many years, some consider it an immature technology. Although the technology does not remain constant, better solutions will become available.
- Ageing: even if the battery is unused, it is exposed to ageing, causing a reduction in capacity.

#### 2.1.3 Waste of Lithium Batteries

Lithium batteries have an approximate life expectancy of three years for small electronic devices, such as laptops, cell phones, tablets, cameras, etc., and between five to ten years in large electronic devices like electric vehicles and energy storage systems, making it a shorter lifespan compared to other batteries. It is estimated that 80% and 20% of lithium batteries are being used for small and large electronic devices, respectively. Also, previously in 2012, approximately ten thousand tons were disposed of. This value has been increasing and in 2020 was estimated to reach about two hundred and fifty thousand tons and expected to reach close to a half a million tons by 2025, due to the increase of use and demand of electric vehicles and energy storage systems [10].

Only up to 5% of lithium batteries are collected in Australia, the European Union, and the United States of America, due to the lack of awareness of consumers and the constant reselling of electronic devices or batteries instead of recycling them. In addition, even the most developed and advanced countries in sustainability with recycling trends lack efficiency, safety, and transportation of the disposed lithium batteries. Therefore, impactful improvements need to take place to tackle this issue [10].



Current state of waste Lithium batteries

Figure 2-3 Quality and collection of discarded lithium-ion batteries [10].

## 2.2 Environmental Issues

Significant environmental problems arise from the usage of lithium batteries, as well as natural resource pressure and pollution that comes from the exploration, extraction, and processing of metals that are part of the battery. Heavy metals such as cobalt and nickel are expected to increase in demand in the future for energy storage. Also, disposal of lithium batteries is a major concern, which is why recycling processes, and second life of these batteries are being studied and discussed in the renewable energy industry since approximately 90% of lithium batteries end up in landfills, making the materials impossible to recover for recycling purposes [11].

Lithium can be extracted in three different ways.

- 1. From hard rock, which is very common in Australia.
- 2. Sedimentary rock, under development in the United States of America.
- 3. Evaporation of brines, which is found beneath the salt flats on South America's Atacama Plateau.



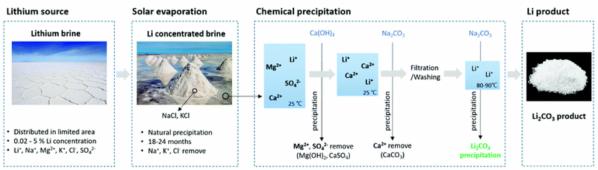


Figure 2-4. The several steps in the production of lithium [10].

Major issues came to light a few years ago, back in 2016, in the lithium mine of Ganzizhou Rongda in Tibet. Protestors rallied together in Tagong, a nearby town, when the fish from the Liqi River, where these fishermen tend to go and fish to provide for their families and communities, were found dead from an outrageous toxic chemical leak coming from the lithium mine. Unfortunately, this was not the only incident in this town because of the high number of mining activity in recent years, for seven years before the previously mentioned incident, a similar accident happened where fish and many other livestock were found dead in the river [12].

This catastrophe is not only happening in Tibet. Unfortunately, it happens around the world in many lithium mines where the proper procedures, equipment, and trained workforce are not being considered when operating the mines [12].

The world's biggest lithium producer is Australia, and Chile in South America is coming in second. The holes are left for around 18 months, so the liquid evaporates before extracting the lithium, resulting in incidents like in Tibet mentioned above, harming local inhabitants and polluting rivers and land as well as its wildlife. Also, the amount of water consumption for the extraction of lithium is a concern; for every

tonne of lithium, half a million gallons of water is needed. Hence, through lithium mining, around 65% of the region's water is consumed in the mine, named Salar de Atacama, resulting in scarcity for the locals and mainly the farmers [12].

In addition, Chile is not the only South American country with this issue. In Argentina, at the Salar de Hombre Muerto, they have expressed concern over the lithium mining in their region since it contaminates the streams and irrigation of crops. Also, the damage to the soil that is left from the mining is noticeable and a concern for the farmers [12].

### 2.3 Applications

#### 2.3.1 Small Applications

Without knowledge, not every individual is aware that the portable electronic device they use every day has a lithium battery. It is a very common technology in small applications such as laptops, mobile phones, portable chargers, drones, smart watches, and others. Lithium batteries are the desired technology for these devices because of their fast charge, safety, and life cycle advantage.

### 2.3.2 Energy Storage

Lithium batteries are the most attractive type of battery for energy storage due to their high charge and discharge efficiency, high specific energy, and long-life cycle. Energy storage systems have a major role in the use of electricity, supplying electricity, and industrial deployment applications, especially at the grid-level. The energy demand tends to be unpredictable, although it can be forecasted with energy demand fluctuates daily, seasonally, and unexpectedly. Hence, to satisfy these changes in demand and continue the constant supply of energy when needed, especially at peak values, energy storage can help reduce the peak levels and load levelling, as seen in Figure 2-5. In order to provide stable and reliable power at a large scale the stability of both the voltage and frequency has to be taken into account, if a mismatch between power supply and demand is found, energy storage can maintain stability during short-term and long-term applications for voltage and frequency of power supply [13].

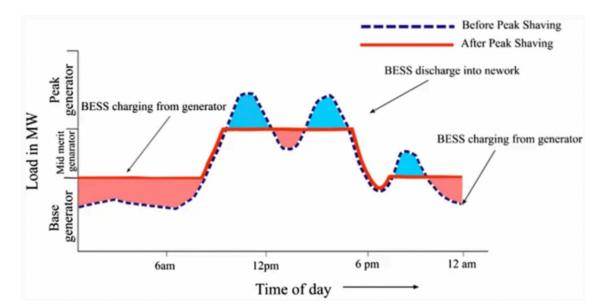


Figure 2-5. Peak load shaving using a BESS [13].

Also, it is understood that today's electrical grid infrastructure cannot fully support the injection of renewable energy. That is also why energy storage with lithium batteries is essential to fight climate change as the world reduces emissions to comply with the Paris Agreement. Due to renewable energy generation abundance and wide resources, lithium batteries have become a main choice for power generation along with the regulations for a greener and more sustainable world. Therefore, this industry has had extraordinary growth and is expected to keep growing. Hence in correlation to their growth is the necessity for efficient energy storage applications, giving more dependency on lithium batteries. In addition, energy storage with lithium batteries is not only necessary for large volumes of storage but also for small residential applications. Many household owners are installing stand-alone photovoltaic systems that require a lithium battery for energy storage, to provide electricity for their residence when photovoltaic energy production is non-existent [13].

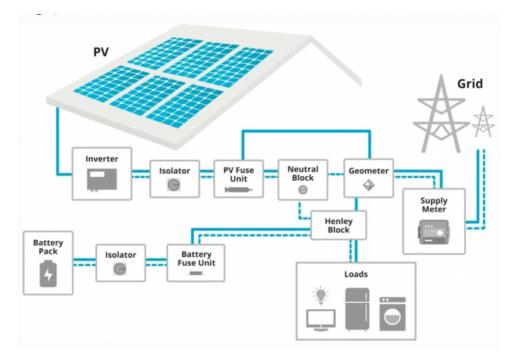


Figure 2-6. Scheme of a residential PV stand-alone system [13].

Furthermore, efficient power management is fundamental to ensure independent and cooperative function of the lithium batteries. The power management system can ensure reliable operation, stability, safety, and cost-effectiveness, hence when batteries are packed in the stack, power management must balance the voltage and current of each battery in the stack. One single lithium battery cell is incapable of satisfying the power grid's requirements, which is why lithium batteries are assembled in parallel to increase current capability and assembled in series to increase voltage. These different ways of assembling lithium batteries have an impact on their stability, operation, safety, and life cycle [13].

#### 2.3.3 Electric vehicle

The transportation industry is responsible for approximately 27% and 14% of the energy demands and greenhouse gas emissions (GHG) respectively, even though that fuel efficiency methods have been deployed, with a safer and a more sustainable way of production of vehicles has also been implemented, is not enough given that the commitment towards a more sustainable world and reducing greenhouse gas emissions is highly dependent on the transportation industry. Therefore, electric vehicles have been seen in the market more and more over the years and are expected to have exponential growth in demand in the following years, as regulations are being taken into place [14].

Also, electric vehicles' impact on the environment is beneficial, reducing and repairing the damage caused by internal combustion engine (ICE) vehicles throughout the years. In addition, electric vehicles are expected to be the major form of transportation in Europe and the world, from personal use to public transportation like buses.

Electric vehicles have a simple mechanism; the components that form part of the mechanism are the following [15]:

- Rechargeable battery: it is rechargeable via a plug and a battery charging unit that can be onboard or fitted in a charging station.
- Electric motor: also functions in reverse as a generator to charge the battery.
- Electric controller: manages the power supplied to the motor, hence controlling the vehicle's speed forward and reverse.

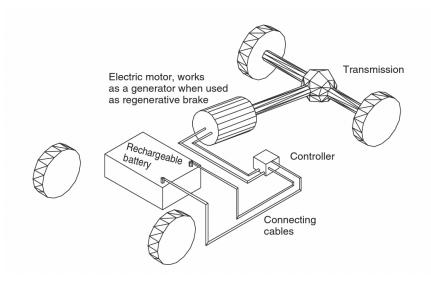


Figure 2-7. Electric vehicle schematic [15].

Furthermore, there are four main types of electric vehicles, and two of them have other kinds amongst them, which are:

- 1. Plug-in hybrid electric vehicle (PHEV)
- 2. Hybrid electric vehicle (HEV)
- 3. Battery electric vehicle (BEV)
- 4. Fuel cell electric vehicle (FCEV)

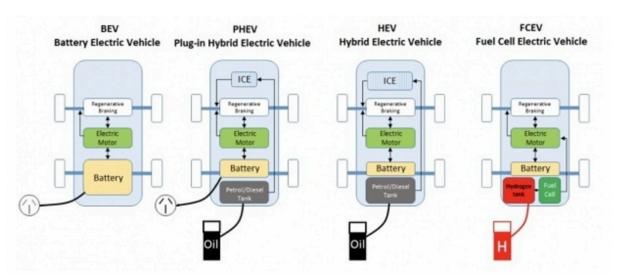


Figure 2-8. Types of electric vehicles [16].

#### 2.3.3.1 Plug-in hybrid electric vehicle (PHEV)

There is a similarity between plug-in hybrid electric vehicle (PHEV) and hybrid electric vehicle (HEV), given that both have an internal combustion engine (ICE) and a battery pack to provide driving power. However, PHEV has a battery system of at least 4kWh, capable of recharging the battery from an external source and can perform drives of up to 80 km in electric mode. The main advantage of PHEVs is the decrease in dependency on oil, better fuel economy, higher power efficiency, decreased greenhouse gas emissions and vehicle-to-grid (V2G) technology; these advantages occur because PHEV runs on both fossil fuels and electricity. In addition, PHEVs can increase their efficiency compared to HEVs, because they can have selective use of the internal combustion engine, allowing it to be used when the vehicle is at high speeds [17].

As previously mentioned, fossil fuels can be found in PHEV; not only gasoline can be used, but also diesel and ethanol. The way to charge the battery is by plugging in an electrical outlet of 120/240 V AC. Also, the battery can be recharged when the vehicle is exposed to braking. This happens due to the regenerative braking system it has that generates energy and directs it to the battery. In addition, it is mentioned in many studies that PHEV decreases the impact on CO<sub>2</sub> emissions due to the recharging of the battery through the electricity coming from the grid. Suppose this electricity is known to be from renewable sources. In that case, the reduction in greenhouse gas emissions could result in zero, making it a more environmentally friendly source of transportation, just like a fully electric vehicle. Plug-in hybrid electric vehicles offer reduction ranges of 25-55% in nitrogen oxide, 35-65% in CO<sub>2</sub> emission, and 40-80% in fossil fuel consumption [17].

Furthermore, PHEVs have three different kinds of designs; these are the following:

- 1. Series design: the electric motor is used to rotate the wheels, and the ICE turns a generator for the supply of electrical power to the electric motor, and the battery stores any excess energy.
- 2. Parallel design: similar design to HEV, where ICE and electric motor are in charge of driving the car by rotating the wheels independently or simultaneously with mechanical coupling.
- 3. Series/parallel design: gives the driver a choice to operate the vehicle in series or parallel mode.

In addition to the design, PHEVs offer two operation modes: the charge-depleting mode and the chargesustaining mode. These modes are often found mixed in PHEVs, and they can manage the battery discharge strategy and a more direct use depending on the size and capacity of the battery. A chargedepleting mode allows PHEVs to operate exclusively on electric power as far as the battery is fully charged, until the battery reaches a state of charge (SOC) low enough that the internal combustion engine must be engaged. On the other hand, a charge-sustaining mode combines the operations of two power sources in the vehicle with the objective of performing as efficiently as possible, limiting the state of charge of the battery from reaching a certain level [17].

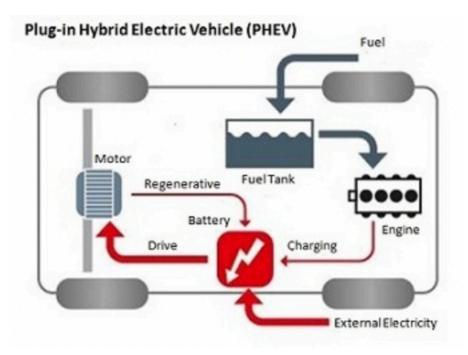


Figure 2-9. PHEVs process diagram [16].

There are battery requirements that need to be followed for PHEVs, such as a durable and larger battery type capable of experiencing deep discharge while performing a high number of cycles and battery life. Hence, issues like battery life, capacity, energy, weight, power, and cost need a solution and at times, a trade-off between them. The cost of a battery pack commonly used in a plug-in hybrid electric vehicle is high compared to a hybrid electric vehicle due to its larger size. In addition, the most common battery packs are nickel cadmium (Ni-Cd) and lithium-ion (Li-ion), where Li-ion batteries offer higher energy and power densities, therefore, a larger traveling range (approximately 600-900 km), and higher maximum speed (around 160 km/h) and acceleration. Furthermore, the energy consumption is estimated to be around 0.125 kWh/km [17].

#### 2.3.3.2 Hybrid electric vehicle (HEV)

As stated in the name, a hybrid has two different power sources: an internal combustion engine and an electric motor system connected to a battery. The purpose of the electric motor and battery is for a better fuel economy and performance compared to traditional ICE cars, and it is achievable since a higher efficiency source, electricity, compensates for a lower efficiency source, fossil fuel. Hybrid electric vehicles are very common in the current market since a number of years ago, with different control strategies dependent on the size of the ICE, and the electric motor, the ratio between both components results in the ratio known as hybridization. In addition, the electric motor can also be used as a generator to charge the battery and start the ICE when required [17].

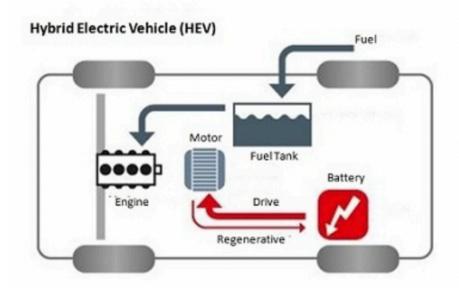


Figure 2-10. HEVs process diagram [16].

Hybrid electric vehicles have a simple mode of operation that consists of using the electric motor to drive the car at low speeds, and the ICE unit is engaged at high speed, faster acceleration, and at times to deliver power to recharge the battery. This combination of both components allows a natural switch between the electric side and the fuel side of the vehicle from driving to high speeds and coming to a stop and so on [17].

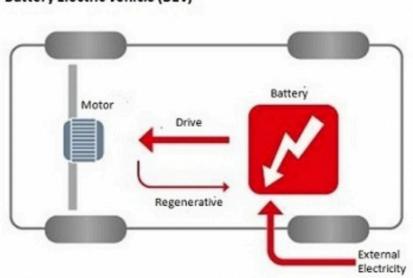
In addition, hybrid electric vehicles also have a regenerative brake that allows recharging of the battery as the car decelerates as it is braking. This technology puts the electric motor in generator mode to charge the battery; it is very effective when the vehicle is at low speeds and in constant stop-and-go driving situations. Nickel-metal hydride (NiMH) is the most common kind of battery in HEVs with an output voltage of 274 V, capacity of 6.5 Ah, and a weight of 53.3 kg, capable of permitting the vehicle a drivable range of 1.200 km maximum [17].

#### 2.3.3.3 Battery electric vehicle (BEV)

Due to the high demand for fossil fuels, given that the majority of vehicles in the world today run-on fuel, the battery electric vehicle, also known as the fully electric vehicle, is the main solution for the transportation sector in the future, due to their very low greenhouse gas emissions. The Paris Agreement states that the objective by 2030 is to reduce global warming by more than 2 degrees Celsius, and this objective can be achieved if BEVs represent 35% of the total number of vehicles by that year [18].

Battery electric vehicles are powered by an electric motor. Electricity is usually generated by on-board rechargeable battery packs and flywheels. Also, as previously mentioned, the battery can be charged like PHEVs, using a plug-in outlet with home electricity or from charging stations. In addition, an advantage, on top of the GHG emission reduction, is the vehicle's performance compared to gasoline vehicles, due to its built-in battery packs that offer quicker acceleration and faster speeds. However, due to the internal combustion engine missing, BEVs are not as effective in colder weather countries

because they struggle with the heating capability of the internal heating system of the vehicle [17].



Battery Electric Vehicle (BEV)

Figure 2-11. BEVs process diagram [16].

The mode of operation of fully electric vehicles is very simple, as shown in Figure 2-11 above. It consists of an electric motor, an electric motor controller, and a battery pack. The power is regulated and controlled coming from the battery pack and directed to the motor to allow the rotation of the wheels. The vehicle functions like regular gasoline vehicles; the power, voltage, and relative resistance depend on the pressure exposed to the acceleration pedal. In addition, BEVs could use AC or DC electric motor. An AC motor uses a three-phase motor running at 240 V with a 300 V battery voltage. On the other hand, a DC motor in a typical range of 20-30 kW and up to 40-60 kW requires a full battery voltage of up to 200 V [17].

Many car drivers are normally concerned about switching to a fully electric vehicle because of the limited electric traveling range, which is why the battery energy potential must be high enough to offer long driving ranges capable of covering daily driving routines, BEVs have a range of approximately 150-500km. Therefore, the battery, which lithium-ion is usually the chosen one, must provide high levels of power and energy, withstanding high discharge and high charging levels. These demands result in a high-cost battery pack, which is why BEVs are more expensive than regular ICE vehicles [17].

#### 2.3.3.4 Fuel cell electric vehicle (FCEV)

A fuel cell electric vehicle (FCEV), also known as a zero-emission vehicle, can be understood as a sustainable form of hybrid vehicle because of the combination of a battery and fuel cell (hydrogen). Although this kind of vehicle is slowly increasing in demand and supply, it is expected to be a leading choice for drivers in the future. Due to the expected technological advances, FCEVs are anticipated to be the best-performing vehicle in the market in the future [19].

As seen in Figure 2-12, the mode of operation consists of the hydrogen tank supplying to the fuel cell to provide power to the electric motor, as well as assisting the battery pack. Also, it is understood that the

technology needs improvement due to the targets of having 65% peak efficiency at 25% rated power, and FCEVs currently offer around 57% peak efficiency at 25% rated power. In addition, the battery pack is like those found in typical BEVs [19].

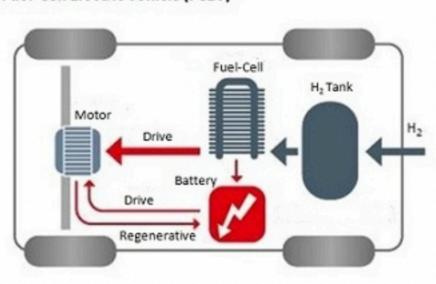




Figure 2-12. FCEVs process diagram [16].

### 2.4 Market

#### 2.4.1 Market Size

Due to the commitment that many countries around the globe have made, aiming to have a more sustainable world, the demand for lithium-ion batteries will continue to increase. In 2021, the global lithium-ion battery market size was valued at around 42 billion United States Dollars (USD), and a compound annual growth rate (CAGR) of 18.1% from 2022 to 2030. In the United States of America alone, the lithium-ion battery market size was 8.5 billion USD, as seen in Figure 2-13. A main factor in the increase of lithium-ion batteries demand is in correlation to the demand for electric vehicles worldwide. The United States of America ended 2021 with a revenue share of 75% in the electric vehicle market, because of the benefits, supportive federal policies, tax breaks, awareness, and market presence. In addition, not only do electric vehicles increase the demand for Li-ion batteries worldwide, but also decrease the demand for lead-acid batteries, due to the contamination and environmental hazard that lead has over lithium [20].

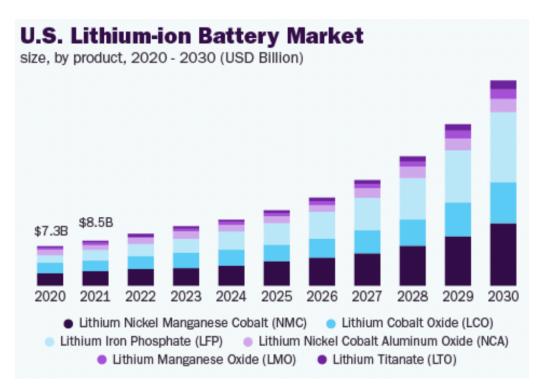


Figure 2-13. USA lithium-ion battery market size [20].

### 2.4.2 Market Share

As previously mentioned, there are various applications where lithium-ion batteries are leading and being used. Consumer electronics, like mobiles, tablets, laptops, portable batteries, etc., was the largest market share for LIBs, with a revenue share surpassing 40% in 2021. Even though sustainable vehicles are increasing in demand, they came in second place in 2021 for the market share. However, it is expected to have the biggest global lithium-ion battery market share in the future, as it is forecasted to have a lucrative and exponential growth. The same growth is expected to happen in energy storage systems application, due to the increase of renewable energies worldwide, sustainable commercial buildings, institutions, etc. Also, given that stand-alone photovoltaic systems will be found in almost every home in the future, it is expected an increase in market share for energy storage systems with lithium-ion batteries in the future. To better understand the global lithium-ion battery market share, please refer to Figure 2-14 [20].

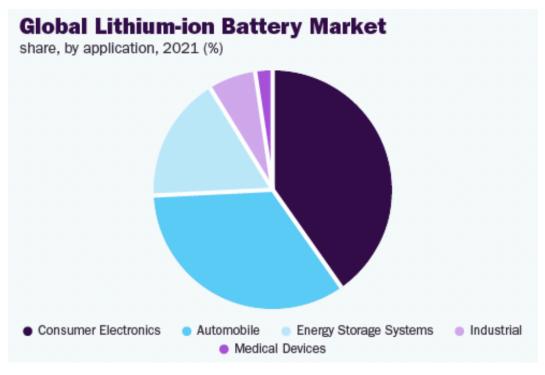


Figure 2-14. Global lithium-ion battery market share [20].

### 2.5 Recycling Lithium Batteries

There are high levels of unawareness amongst people on the importance of recycling batteries. Many individuals still throw away their batteries, as mentioned previously, about 90% of batteries end up in landfills and around 10% of the remaining batteries are recycled. Specialists in the environment and batteries suggest recycling used batteries, since it will recover the expensive materials in their composition. It can be seen in the price fluctuation of nickel and cobalt; these are expensive cathodes that are used regularly. Now looking at lithium batteries (LIBs), the concentration of lithium, nickel, manganese, and cobalt are high, meaning that if these materials are recovered through recycling, they can have the same benefits as natural ore [2].

Also, it is understood that recycling will decrease the electronic waste that is present in landfills nowadays. Countries similar to Congo are responsible for approximately 50% of the cobalt used in batteries worldwide, previously mentioned countries like Australia, Chile, Argentina, and Tibet, are responsible for the mining of lithium. Resulting in environmental damage, social conflicts and human rights abuse which will be reduced from the recycling of batteries by reducing the dependency on these materials. Therefore, there will be an improvement on security and safety of the mines, and this will show benefits in the social and environmental impact that have been affected from the mines [2].

On the business side of recycling batteries, major companies with high reputation and knowledge are developing and commercializing many recycling processes to recover the materials in batteries and capture the most amount of value possible, as seen in Figure 2-15.

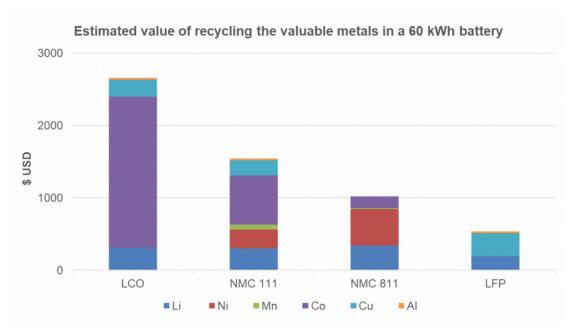


Figure 2-15. Estimated material value in a 60-kWh battery [21].

Currently, there are three main points to which lithium batteries are being sent at their end-of-life stage: previously mentioned landfills controlled by municipalities, waste-to-energy facilities, and recycled focus facilities. The objective is to eliminate the first two destinations and aim that every end-of-life (EOL) lithium battery goes straight to the recycling facilities. Many different recycling processes of lithium batteries are now taking place and under development as well. These recycling processes for lithium batteries have different unit operations. Firstly, the battery must face deactivation, which is done in three ways [22]:

- 1. Discharge: reduces stored energy, preventing the activation of further reactions, hence reducing the hazardous level.
- 2. Thermal treatment: all organic compounds of the battery cell are decomposed along with the volatilization of the electrolyte.
- 3. Freezing electrolyte: the electrolyte is exposed to freezing conditions to prevent any electrochemical reaction and short circuits.

These unit operations can also be divided into three methods of treatment [22]:

- Mechanical treatment: battery cells or battery modules are crushed to open them, continued by the extraction of the valuable materials within them, followed by the classification and separation processes of aluminium foil, copper foil, separator, and electrode active materials. Also, modules and battery cell casting are recovered alongside the connecting components.
- 2. Pyrometallurgical treatment: many components that are part of the battery cell and the battery cell itself are melted. Through this process, the lithium and aluminium remain in the slag, while metals such as nickel, cobalt, and copper are recycled from the battery cell cast. However, an extra procedure is needed to recover lithium, which is when hydrometallurgical treatment takes place.
- 3. Hydrometallurgical treatment: as previously mentioned, used to recover lithium from slag after

the pyrometallurgical treatment, as well as from the separated electrode active material from the mechanical treatment. Also, this treatment includes the discharge of main metals, extraction, and separation, to finally recover the products from the separated steams by crystallization and precipitation.

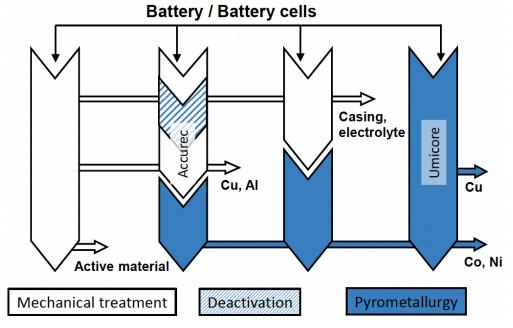


Figure 2-16. Battery recycling treatment combinations [22].

In addition, safety regulation plays a major role in the life cycle of lithium batteries, especially during the collection, storage, and dismantling each end-of-life lithium battery. However, mistreatment of the batteries in certain circumstances may cause leakage of lithium battery components and thermal runaway as it can catch on fire and result in an explosion. As seen in Figure 2-17, a visual explanation of the recycling process can give a better understanding.

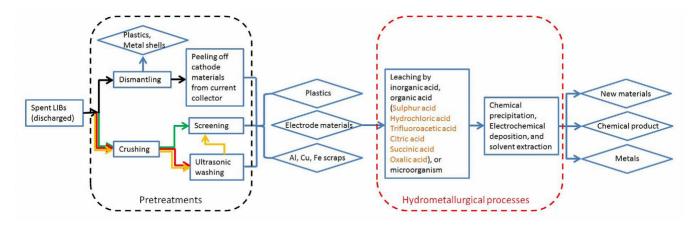


Figure 2-17. LIBs flowchart of the recycling process [23].

#### 2.5.1 Sorting, disassembly, and discharging

The standard separating processes are known to be through hand picking to magnetic separation, xray, electromagnetics, and lastly by UV-based sorting. The most common sorting technique across the European Union is a combination of manual and visual sorting. For example, in Germany, approximately 50% of the portable batteries that were collected in 2013 were sorted as previously mentioned. In addition, industrial batteries that have a challenge during sorting procedure due to their size, weight, and unique non-standard designs. Therefore, it cannot have an automated sorting process, resulting in high handling costs because employees must manually sort it. This ensures the importance of labelling batteries and improving battery sorting methods [7].

The next step after sorting is the disassembly of lithium batteries. The main advantage of dismantling before recycling lithium batteries is that the residual value of components are more accessible and profitable. Valuable materials like copper, steel, aluminium, plastics, and any other metals are more accessible for recycling after the disassembly of lithium batteries. It also reduces the volume flow and variety of material for a more efficient recycling process. Standardization is a major challenge in the disassembly process since every EV battery must be manually dismantled by trained employees and appropriate tools to handle high voltages and energy density. All these challenges can be avoided by having a more sustainable, eco-friendly battery design, which can be adapted to disassembly automation, also for manufacturers and recyclers to exchange and share their knowledge and achieve this faster [7].

In addition, it is expected that disassembling of lithium batteries allows discharging and diagnosis of the current state-of-health (SoH) of the battery modules. The information that can be obtained from the state-of-health of a battery is for decision making to ensure that the battery qualifies for any kind of reuse, either by repairing it or refurbishing it, known as second life battery in the automotive industry. Unfortunately, a standard that determines the SoH of a battery is not yet available. However, there are technical solutions in place. There are two main ways to discharge a battery [7]:

- 1. The current method uses ohmic resistors, especially for large energy storage and electric vehicle batteries. The remaining lithium batteries are either evaporated or stored for further use in an on-site energy storage project.
- 2. The use of electrically conductive liquid, mainly used for small batteries.

## 2.5.2 Pyrolysis for deactivation

Before mechanical treatment, pyrolysis is done on lithium batteries, with the objective of deactivating battery cells for safety measures. Any active materials of the anode and cathode are separated from the aluminium foil or copper through the removal of organic binder. The energy content is controlled as organic components are removed, and the halogenated substances are reduced because they are removed through the exhaust gas stream. Once pyrolysis is completed, lithium batteries can be temporarily stored, allowing the mechanical treatment to take place, reducing the risk of thermal runaways [7].

For the aluminium foil to be recovered in its metallic form, pyrolysis under vacuum is necessary. An experiment was carried out where the process is placing the furnace under the vacuum and filled with high purity nitrogen. Previously, some experiments noticed that at 600 °C under atmosphere,

unfortunately, the results were not as expected, since the cathode could be detached, while the electrode faced deformation, since it became fragile and oxidised slightly at such high temperature. During the pyrolytic process, mass losses are found, and can be divided into two stages [7]:

- 1. Mass loss, a result of the volatilisation of moisture and electrolyte leftover, happens through the initial heating stage at a temperature of 30 °C to 150 °C.
- 2. Decomposition of organic binder happens at two different temperature stages, from 250 °C to 450 °C, and from 550 °C to 600 °C.

Pyrolysis facilitates the liberation of electrode material from the conductor foil for the next processes, as well as the temperature during pyrolysis impacts on the separation efficiency. However, the optimal temperature to achieve the most efficient separation of cathode and anode, is 500 °C and 550 °C, respectively [7].

#### 2.5.3 Mechanical separation and pre-treatment

After the previously explained deactivation and discharge process, lithium battery packs are crushed or thermodynamically or chemically inhibited to prevent fire or explosions. The main objective is to separate valuable fractions of iron, copper, and aluminium alloys from the black matter, which comes from the active materials of the lithium batteries cathode and anode, hence, it possesses valuable compounds, high in lithium, copper, nickel, manganese, etc. these valuable compounds are then recovered through the metallurgical treatment. Mechanical pre-treatment uses rotary shears at low revolutions per minute (rpm) with double shaft shredders, or impact crushing with high revolutions per minute using hammer mills. The shredding may be accompanied with sieving, magnetic, and density separation depending on the particle size after the first separation step [7].

In addition, if thermal deactivation of the battery cell was not wanted, crushing them can be done by shredding in inert atmosphere, cryogenic crushing, or electrohydraulic shredding. Unfortunately, it is very costly, and needs a high level of investment and operational cost, although the recycling efficiency is higher through pre-treatment of the battery cell. Also, since thermal deactivation was not performed, this allows for materials such as plastic and the electrolyte, to be recovered more easily. These methods, both shedding in inert atmosphere and cryogenic crushing, are not dependent on the state-of-charge or state-of-health of the lithium batteries [7].

Furthermore, shredding in inert atmosphere is a safer method, since it prevents the battery from catching on fire or even exploding, this method is performed in a closed chamber filled with carbon dioxide, nitrogen, or argon, before any comminution happens. On the other hand, cryogenic crushing occurs when batteries are cooled down between -175 °C and -195 °C with the use of liquid nitrogen, dry ice, or argon, taking advantage of the low temperatures; resulting in the materials of which the battery is composed to be brittle and electrochemically inactive. The brittleness level of the materials allows for a safer and more efficiently grinding, since it can be beneficial for further metallurgical processes [7].

Moreover, another technique under mechanical separation is known as flotation separation, which consists of physicochemical separation that works with the differences between the hydrophobic

graphite anode materials and the hydrophilic cathode materials. A bubbling process occurs during flotation, allowing the graphite, chemically known as a hydrophobic particle, to be attached to the air bubbles and rise to form a foam layer converting it into a launder, leaving behind any hydrophilic particles. This method results in approximately 90% and 10% recovery of anode and cathode materials, respectively [24].

In summary, mechanical pre-treatment and mechanical separation are performed as an auxiliary method to improve the recovery efficiently of targeted materials before metal leaching process. Unfortunately, in lithium batteries, its components are not separated completely throughout this process because of their composition, which happens to collide with each other [24].

### 2.5.4 Thermal treatment

Lithium batteries could also face thermal pre-treatment in two different ways. One is by removing organic components, binders, and electrolytes from end-of-life lithium batteries. The second way is by applying pyrolysis to produce compounds or electrode materials as the temperature and atmosphere are controlled. Thermal treatment is a simple method that can be performed on a large scale, although smoke is produced due to combustion; hence, purification is needed to prevent pollution and have a more sustainable process. Efficient thermal treatment minimizes the problems during the solid-liquid separation procedure after leaching, and the efficiency of recovery increases [24].

Also, both positive and negative electrode materials, during thermal treatment with high pure nitrogen environment, increase the recovery efficiency, and controlling the temperature between the melting point of the current collector and thermal decomposition can result in the burnt off of the binder and carbonaceous conductor [24].

## 2.5.5 Pyrometallurgy

The iron and steel industries have done the pyrometallurgical method to extract metals from different ores, minerals, and raw materials, some of the same processes can be applied to the recycling of lithium batteries by using high temperatures to cause reactions in the cathode and anode to make lithium soluble in H<sub>2</sub>O by evaporating organic materials. One of the main mechanisms is known as calcination, from conventional pyrometallurgical operations, is a thermal process that takes place in either low oxygen or free of oxygen atmosphere at 700 °C and applied to solid materials for the removal of volatile components, while the solid material experiences a change in its phase or decomposition reactions. Following water leaching through filtration, that water is then evaporated, leaving the lithium chemical available. A visual explanation is available in Figure 2-18 [10].

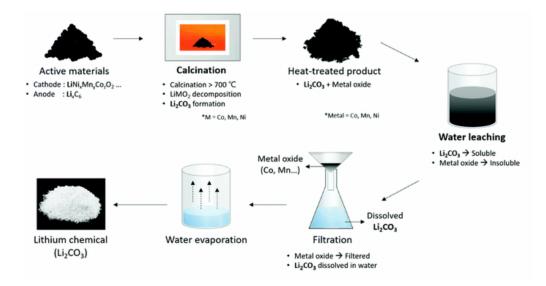


Figure 2-18. LIBs recycling by pyrometallurgy [10].

Also, it is mainly performed to pre-treat metals that contain carbonate minerals. Concerning the recycling of lithium batteries, the main point of interest is the decomposition of lithium metal oxides into a metal oxide and lithium carbonate, as the oxygen is released. In addition, to further decomposition of lithium carbonate into carbon dioxide and lithium oxide [7].

On the other hand, roasting is a pyrometallurgical method that takes place in an atmosphere rich in oxygen and usually applied to sulfide materials, for a more efficient lithium extraction. Applying this method on lithium batteries is highly dependent on the pre-treatment that the battery went through [7].

In addition, another well-established process is known as Umicore, including both pyrometallurgical and hydrometallurgical processes, which will be explained later. In this process, end-of-life lithium batteries are fed together with coke and slag formers to a shaft furnace, which is divided into three zones with different temperatures each, these are [7]:

- Zone 1: known as the preheating zone, the electrolyte of the LIBs is evaporated at a slow rate at temperatures that do not exceed 300 °C to have a safer process and reduce the risk of an explosion.
- 2. Zone 2: where the temperature reaches 700 °C, using the C-rich phases from the lithium battery packs, like plastics, hence the energy efficiency is enhanced by lowering the energy requirement of both the melting and reduction processes.
- 3. Zone 3: lithium and aluminium are oxidized and slagged, while copper, cobalt, and nickel are reduced to finally have a metal alloy.

The off-gas, leaves the furnace at a temperature below 700 °C, then it is heated to 1,150 °C using a plasma torch and post combusted afterwards. Then, the gas's temperature is lowered to approximately 300 °C and it is cleaned for further use. The main advantage of the Umicore process is that it does not require any pre-treatment; however, it demands a high level of energy and slagging of lithium [7].

Furthermore, another drawback of the pyrometallurgy process when recycling lithium batteries is that it requires extra procedures after calcination, as explained above. Also, it requires unique equipment to

perform calcination and may cause the emission of harmful gases throughout the process [10].

#### 2.5.6 Hydrometallurgy

The most used recycling method for the extraction of lithium from lithium batteries uses leaching, crystallization (selective absorption), extraction, and precipitation. Also, the purpose of the hydrometallurgical process is to re-synthesize battery materials to create a closed loop recycling. A more in-depth explanation of each step within the hydrometallurgical process is provided below [7].

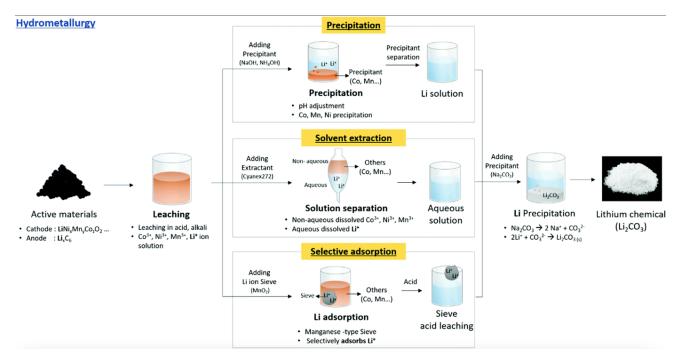


Figure 2-19. LIBs recycling by hydrometallurgy [10].

#### 2.5.6.1 Leaching

Leaching of the black matter, is the first step of a standard hydrometallurgical treatment, where the metal components are transitioned to dissolve in inorganic or organic acids to prepare for precipitation, solvent extraction, and selective absorption. Also, the purpose is to minimize the impurities found in organic residues and to prepare a variety of the purest possible product metals [7].

In addition, kinetic data has a very important role in the leaching process, since it has a high level of dependency on the heterogeneous phase transition between solid, liquid, and gas, plus with the addition of reduction agents results in an acceleration of the process. Furthermore, regarding solid-liquid ratio, parameters taken into account, like the temperature of the reaction and the concentration of the leaching agents, have a very important role [7].

Nowadays, leaching has been performed and studies show how beneficial the chemical properties of metals presented in aqueous solution to separate and recover a product, such as lithium, can be offered to the market. Finally, the efficiency in this very first step depends on the nature of the cathode, the reducing agent and its concentration, and acid used, as well as the time through the leaching process

and at what temperature, stirring speed and liquid-solid ratio [7].

#### 2.5.6.2 Precipitation

Following the leaching process, the most used method to separate lithium is known as precipitation, it uses the difference in the solubility of metal compounds provided by thermodynamics and E-pH diagrams, the compounds found within that region are the ones precipitated. In addition, transition metals like oxalates or hydroxides, which are materials low in solubility, are the ones precipitated as well [10].

The precipitation method is the most economical, efficient, and safest compared to solvent extraction and selective absorption within the hydrometallurgical treatment. The increase in temperature results in a decrease in solubility, making precipitation easier. The same happens when the pH increases, resulting in the separation of hydroxides after precipitation, like cobalt, manganese, and nickel. For example, a saturated sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) solution added to the solution to create lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>) reportedly extracts close to 98% of lithium [10].

#### 2.5.6.3 Solvent extraction

This process is shorter than precipitation, in order to separate lithium from the leached cathode material, solvent extraction requires a two-phase system, as well as relative solubility to separate ions from nonpolar and polar liquids. Lithium is removed from the stratified solution, and metals such as manganese, nickel, and cobalt are removed from nonpolar extractants. Recent development created a compound called Cyanex 936P, that can separate lithium from alkaline metals. This new compound has a lot of potential regarding lithium battery recycling for the extraction of lithium. Also, the efficiency of the solvent extraction process depends on pH conditions and solvent concentration [10].

#### 2.5.6.4 Solvent absorption

Solvent absorption removes lithium, which is the smallest metal ion, by using lithium-ion sieves, which are inorganic adsorbents that originate from the technology used to extract lithium from lithium brines, to absorb the dissolved lithium. Even though the hydrometallurgical process has high recovery rates and minimum use of equipment, it is not the case for this method since it requires a vast volume of acid and base for leaching, resulting in additional chemical cost, and the cost of disposing of the used solution is also high [10].

### 2.5.7 Electrochemical Extraction

The purpose is to separate lithium from pre-treated active material by placing the active material in water and using a ceramic solid electrolyte to have separated lithium. In addition, it depends on the purity of lithium, given that high purity lithium is the best-case scenario, but most of the brines do not consist of this level of purity since they are relatively low. A main advantage of electrochemical extraction, also known as electrochemical ion pumping, is the versatility available, which permits brines with distinct components and concentrations by changing its input, as well as that through the application of current to complete the process, other chemicals are not required and needed to regenerate the active material. Electrochemical extraction consists of four main steps, which are [25]:

- 1. Lithium cations from an artificial brine and an electrolyte are selected to be placed between a lithium captured electrode through current.
- 2. The solution is then exchanged with a recovery one for the release of lithium.
- 3. Lithium cations are released into the recovery solution, simply by changing the direction of the current.
- 4. The lithium brine is flushed in the cell so the cycle can start again from the first step.

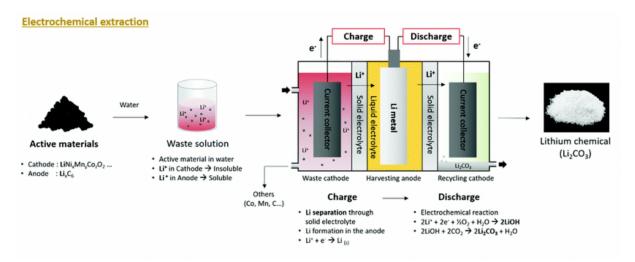


Figure 2-20. Electrochemical extraction scheme [10].

Also, the percentage of extraction applying electrochemical extraction is high, around 75% to 95% of not only lithium carbonate but also lithium metal. The purity level of both lithium carbonate and lithium metal are very pure, resulting in 99.6% and 99%, respectively. Unfortunately, this recycling method has been performed at a laboratory scale and is expected to be ready for commercialization in the near future [10].

# 2.6 Second life battery

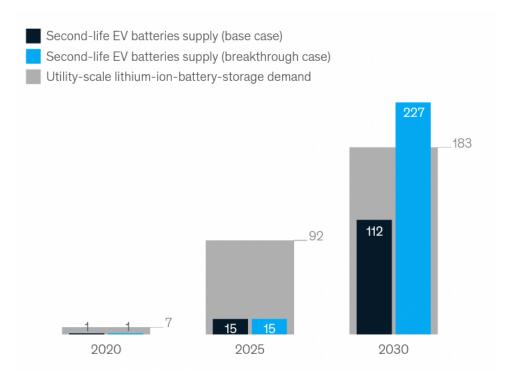
After a battery has reached its end-of-life, it does not mean they are no longer useful. In addition, to recycling lithium batteries they can also have a second life, by reusing them in various ways, and adding economic and sustainable benefits. When the battery of an electric vehicle reaches a capacity of 70% or 80%, meaning that these batteries are eligible for stationary systems, such as energy storage systems to store energy produced by renewable sources. Allowing reusable batteries like second life will reduce carbon footprint and increase renewable energy on the grid. Also, it decreases the disposable cost of lithium batteries, and by 2030, in Italy, it is estimated that approximately 60 kilotons of electric vehicle batteries will be disposed of per year [3].

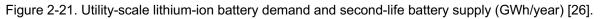
For a circular economy, the efficient use, recycling, and reuse of storage systems and batteries increased over the years, resulting in the investment of specific projects, research, and development regarding second-life batteries (SLB) [3].

However, there are challenges that second-life batteries must overcome, these are [26]:

- Large number of battery-pack designs with customizable sizes, electrode chemistry and format. Therefore, the lack of standardization of batteries limits how easily it can be set up for secondlife use.
- 2. As technology advances, the cost of a new battery continues to decrease, diminishing the economic advantage of second-life batteries. Currently, there is a 30 to 70 % cost advantage of second-life batteries over new ones, and it is projected to decrease to 25% by 2040.
- 3. There are no guarantees of quality and performance of second-life batteries, requiring standards for second-life batteries.
- 4. Lack of regulations give regional differences regarding if a battery must be recycled or reused.

Figures 21 and 22 give a visual representation of a utility scale of lithium-ion battery demand with second-life battery supply, and the second-life geographical battery supply, respectively [26]. As for Figure 2-21, it displays the increase of demand for lithium-ion battery-storage at a utility-scale for the years 2020, 2025, and 2025, as well as a base case representation and a breakthrough case, meaning the best possible scenario, done by McKinsey and Company. It shows that by 2030 in the breakthrough case the utility-scale demand can be covered completely and even more, by second-life batteries from electric vehicles.





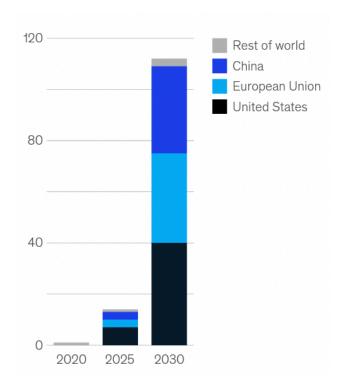


Figure 2-22. Second-life battery supply by geography (GWh/year) [26].

# 2.7 Battery Passport

The European Union is developing a regulation on electric vehicles batteries known as a battery passport, which is a technology platform that allows stakeholders in the supply and value chains to share information amongst themselves to know better about the battery and its history to increase safety, makes the most out of the usage of the battery with the remaining lifecycles and to get the most during the recycling process at the end-of-life [4].

The objective is to enable via IoT to have the data on battery minerals, battery pack and key components, such as modules, and sales. This data can be shared between interested and qualified stakeholders on a distributed blockchain. In addition, it will provide real-time data on the interested battery, which is immutable. Also, the European Commission and the Global Battery Alliance understand and acknowledge the importance of battery passports in the electric vehicle battery circular economy [4].

Furthermore, this initiative will let battery manufacturers build a more standardized battery. As previously mentioned, this will greatly benefit any recycling process, increase efficiency, and reduce costs.

## 2.8 Future trends

A new approach to recycling lithium batteries, specifically found in electric vehicles, has been developed by researchers from the University of Leicester in the United Kingdom, which involves ultrasonic waves to remove important chemicals, like lithium and nickel, from end-of-life batteries. [27]

Since current methods, previously explained, are somewhat inefficient, slow, and time-consuming, this new ultrasonic wave method separates the elements faster, cheaper, and more eco-friendly, this technology is already applied in the food preparation industry and by dentists. It functions by breaking down adhesive bonds between coating layer and substrate. The researchers had outstanding results when removing graphite, lithium, nickel, cobalt, and manganese. Also, efficiency values were very high when tested on the most common battery types [27].

# **Chapter 3**

Development

## 3.1 Introduction

To know a viable industry process on recycling lithium batteries, three different battery selections are studied in this dissertation. The batteries are, a recycled battery, a second-life battery, and manufacturing a new lithium battery. Different case studies and theoretical papers were analysed to gather the most crucial information to compare the different scenarios.

Throughout this section, different findings will be displayed, such as the process selected for recycled lithium battery, the amount of recovered material limitations and steps to provide second-life batteries, and the economics behind the processes used to understand the business side of it. This will give all the needed information to perform a full analysis in the next section of the report with the assumptions and applications studied.

# 3.2 Recycled Lithium Battery

An experiment was performed at the University of Queensland to know the recyclability of lithium-ion battery materials. The raw materials and chemicals that took place in the experiment were [28]:

- Spent iPhone battery lithium cobalt oxide (LCO) powder.
- Commercial lithium cobalt oxide powder.
- 98 wt.% of sulphuric acid.
- Sodium metabisulphite (Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub>) with a purity level of 97%.
- 30 wt.% of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>).
- 2 wt.% nitric acid.
- NaOH and Na<sub>2</sub>CO<sub>3</sub> 1M

In addition, economic aspects were also studied from a different source to understand the market value of recycled materials and the expenses behind performing recycling methods on a large scale.

### 3.2.1 Recycling experiment

The iPhone batteries underwent pre-treatment, acid leaching, selective precipitation, cathodic material resynthesis, and fabrication of new lithium-ion batteries from the recovered product. The first step was pre-treatment to ensure the safety of the battery for the recycling process. The lithium batteries need to have a voltage below 3.0 V to be safe for dismantling, unpacking and unfolding component layers of the four spent iPhone batteries used in the experiment were conducted [28].



Figure 3-1. iPhone batteries [28].

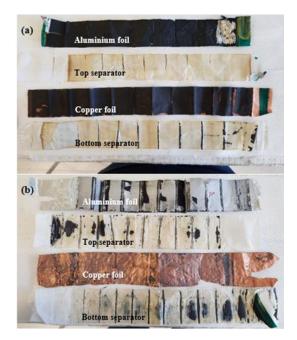


Figure 3-2. Materials extracted from iPhone batteries [28].

An example of the LCO powder gathered from the iPhone batteries previously mentioned is shown in Figure 3-3. The powder was tested by SEM-EDS and atomic absorption spectroscopy (ASS) for qualitative and quantitative analysis. These analyses determine any signs of damage found in the lithium cobalt oxide powder, resulting from the battery's charging and discharging. In addition, it also shows grain degradation, leading to a high surface area and accessibility of lithium cobalt oxide powder with leaching solutions [28].



Figure 3-3. LCO powder [28].

Finally, the cathodic material mass and raw LCO powder composition is provided in Table 3-2. The cathodic material was fully dissolved and achieved liquid state for better analysis. The stoichiometry ratio of LCO should be 1.1 theoretically, and the experiment performed at the University of Queensland achieved a stoichiometry ratio of approximately 1.07, which is very close to the theoretical value. Such high weight concentration of cobalt in LCO powder shows the level of waste if these kinds of batteries are not recycled [28].

Battery Number	Mobile	Cathodic material mass (g)
IP3-001	iPhone 3	4.9778
IP3-002	iPhone 3	5.0249
IP5-001	iPhone 5	4.0291
IP6-001	iPhone 6	5.2015

Table 3-1.Dismantled LIBs [28].

Table 3-2	Raw	LCO	powder	composition	[28].
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Element	Lithium	Cobalt
wt. %	8.3	65.8

Furthermore, the acid leaching experimental results of metals from active cathode material were positive. There was an increase of cobalt leaching from 67.6% to 98.5%, and of lithium from 74.6% to 98.9%, increasing the acid concentration from 1 mol/L to 4 mol/L. Given that there was a similarity on the results of acid concentrations between 3 mol/L and 4 mol/L, the experiment was conducted with an acid concentration of 3 mol/L to minimise risks, reduce cost and for environmental benefits [28].

Cobalt and lithium leaching efficiency also changed with the addition of different reducing agents. However, the most significant change was when hydrogen peroxide dosage, was used as the reducing agent, from 0 to 4 wt.%, showing an increase in leaching efficiency for cobalt from 68.74% to 99.77% and for lithium from 39.19% to 99.89%. Therefore, hydrogen peroxide is understood to positively affect the metal recovery [28].

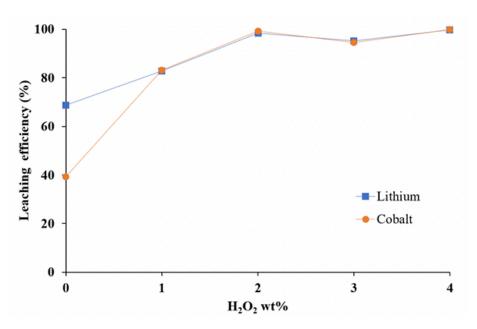


Figure 3-4. Leaching efficiency of hydrogen peroxide [28].

In addition, the effect that temperature had on metal leaching was significant. The experiment displayed results of higher leaching efficiency when the temperature increased from room temperature (25 °C), where only 76.31% and 79.65% of cobalt and lithium were leached, respectively, and when the temperature raised to 80 °C, the leaching efficiency of cobalt increased to 98.76% and of lithium to 99.26%, as shown in Figure 3-5. On the other hand, changes such as pulp density and time did not significantly affect leaching efficiency [28].

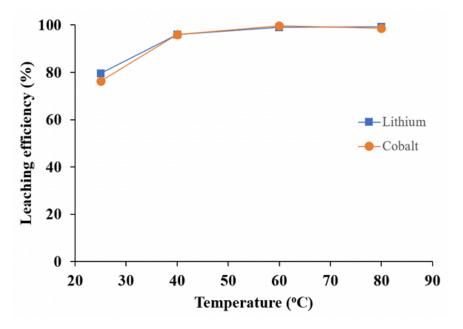
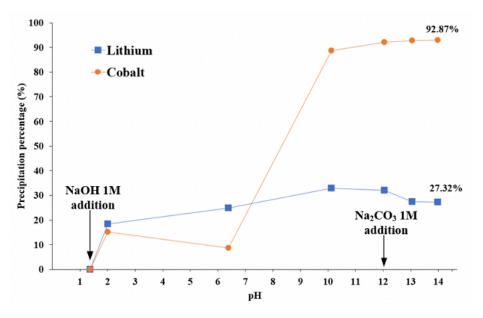
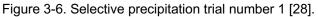


Figure 3-5. Leaching efficiency at room temperature [28].

Then, the following step was precipitation to recover the wanted metals. The achieved leachates are

transparent and light red, which means that there is a high presence of  $Co^{2+}$ . The experiment ran twice to try the most efficient recovery of cobalt and lithium by adding NaOH 1M at pH equal to 1 and adding Na<sub>2</sub>CO<sub>3</sub> 1M at pH equal to 12. However, only cobalt resulted in a high precipitation percentage reaching 92.9% and lithium only 31.9%. Figure 3-6 and Figure 3-7, as well as Table 3-3, provide a better representation of the difference in precipitation percentage between cobalt and lithium on both tries [28].





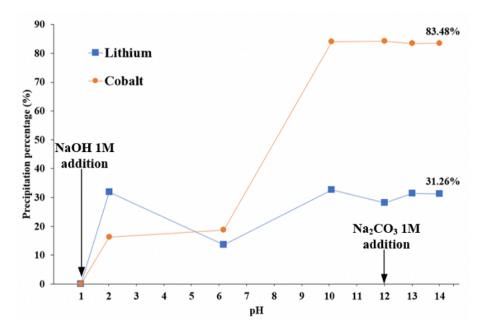


Figure 3-7. Selective precipitation trial number 2 [28].

Try	Metal	Leaching efficiency (%)	Precipitation efficiency (%)	Overall recovery efficiency (%)
First	Cobalt	99.1	27.3	27.1
First	Lithium	99.3	92.9	92.2
Casand	Cobalt	99.4	31.3	31.1
Second	Lithium	99.2	83.5	82.8

Table 3-3. Selective precipitation recovery efficiency [28].

Also, the experiment was analysed by X-ray Powder Diffraction (XRD), an analytical technique for phase identification of crystalline material and information on unit cell dimensions. It was found many similarities between recycled and commercial lithium cobalt oxide powder. These peak intensities can relate to the purity levels of the powder and LCO formation reaction in the recycled sample. A visual representation of the spikes is found in Figure 3-8 [28].

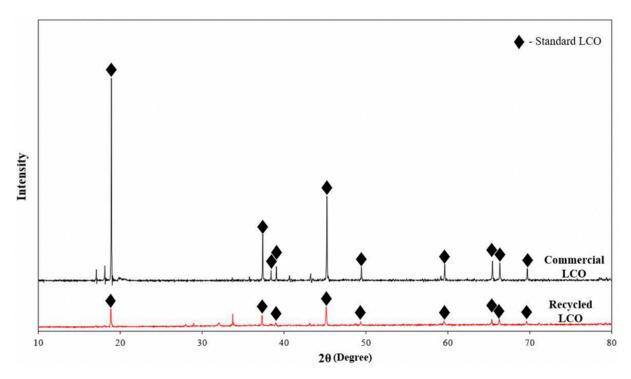


Figure 3-8. LCO powder purity levels [28].

The experiment concluded with the fabrication of two new lithium-ion batteries, one with recycled LCO material and the other with commercial LCO powder. Both batteries were exposed to an electrochemical impedance spectrum (EIS) to study the response of an electrochemical cell to an applied AC potential. Figure 3-9 gives a visual representation of the results of the EIS test [28].

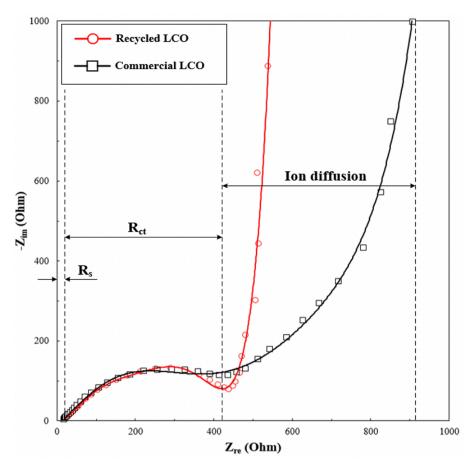


Figure 3-9. EIS test results of electrochemical cell response [28].

Figure 3-9 shows the similarity in electrolyte resistance ( $R_s$ ) and charge transfer resistance ( $R_{ct}$ ). Electrolyte resistance comes from an ionic solution which depends on its concentration, type of ions, covered area, and temperature. Also, charge transfer resistance measures how easily an electron shifts from one atom or compound to another. On the other hand, the major difference was seen in the ion diffusion kinetics of the battery, which consists of a number of extend, narrow, parallel channels through which the fluid moves in streamline flow, in which the recycled LCO has a steeper line meaning a low diffusion resistance, hence a pure capacitive response at low frequencies [28].

Also, a cycling performance test was performed, but as expected, the commercial LCO battery performed better than the recycled LCO battery. The commercial battery shows a decay in performance after 40 cycles, and the recycled one after just 3 cycles [28].

#### 3.2.2 Recycling process economics

The market value of the main elements of lithium-ion batteries is provided in Table 3-4. Taking a reference in the prices per element can give a better understanding of the costs and savings performed through recycling process [1].

Material	Price (€/Ton)
Cobalt	61,550
Nickle	20,171
Lithium	30,930
Copper	9,688
Aluminium	2,658
Iron	90.50
Manganese	5.4

Table 3-4. Market value of lithium battery materials [1].

The recycling value can be calculated by adding up different variables, adding up to a total of  $\leq 26/kWh$  with a recycling fee of  $\leq 10/kWh$ , such as [1]:

- Depreciation
- New battery
- Environmental treatment
- Materials
- Equipment

- Recycling plant location
- Transportation
- Electricity
- Workforce
- Maintenance
- Taxes

A recycling plant must operate for 320 days per year, 20 hours a day, and has a useful life span of 10 years. For example, American Manganese, which is a company focusing on lithium recycling in the USA, states that nickel, manganese, and cobalt (NMC) batteries weigh approximately 7.3 kg/kWh, with materials ranging from €15/kg up to €95/kg. Depending on the type of battery recyclers are paid to recycle a specific battery, it is expected that NMC batteries have higher values, are purchased to be recycled, and can create more profit. Table 3-5 provides the value of each kind of battery; a negative value means that the recycler gets paid to recycle the battery [1].

Battery	Value (€/kg)
LCO	2.00
NMC (111)	0.20
NMC (622)	0.00
NCA	0.00
LMO	-1.00
LFP	-2.20

Table 3-5. Recycling values of lithium batteries for recyclers [1].

In addition, capital expenditure (CAPEX), operational expenditure (OPEX), and return of investment (ROI) must be considered as well. CAPEX are the costs that are needed to improve efficiency, capacity, and innovation in the recycling process, like in machinery, equipment, materials, etc. Table 3-6,

Table 3-7, and Table 3-8 provides values in direct capital expenditure, indirect capital expenditure, and the total capital expenditure respectively [1].

Direct CAPEX	Value (€ M)
Infrastructure	9
Land and buildings	34
Front end	6
Hydromet	24
Utilities	12
Installation	11
Subtotal	96

Table 3-6. Direct CAPEX values [1].

Table 3-7.	Indirect CAPEX values [1].
------------	----------------------------

Indirect CAPEX	Value (€ M)
Project management	45
Insurance & taxes	9
Subtotal	54

CAPEX values [1].

Table 3-8.

CAPEX	Value (€ M)
Direct CAPEX	96
Indirect CAPEX	54
Contingency (10%)	15
Total	165

Also, a 2018 estimate of the value generated from recycling lithium batteries in 10 to 20 years was around 20€/kWh. For the energy storage system application, a recycling process covers the decommissioning costs, Table 3-9 mentions the costs of lithium-ion battery in 2018 and 2038 for a 1000 kWh storage system [1].

Table 3-9. Lithium-ion battery costs for a 1000 kWh storage system [1].

Process	2018 Cost (M€)	2018 €/kWh	2038 Cost (M€)	2038 €/kWh
Workforce and Equipment	8,250	50.69	13,409	13.41
Transport	8,000	191.49	13,367	13.37
Recycling	10,000	33.79	-33,418	-33.42
Total Cost (2.6% inflation in 20 years)	26,250	275.97	-6,642	-6.64

Furthermore, operational expenditure (OPEX) comprises payroll, maintenance, and workforce hiring

costs. An operation conducted in Germany of lithium-ion battery recycling resulted of  $\in$  1,560/ton, and out of that total it breaks down as shown in Table 3-10 [1].

Process	OPEX Percentage (%)	OPEX Cost (€/ton)
Reagents and consumables	33.4	521.04
Workforce	26.4	411.84
Administration	22.9	357.24
Utilities	12.8	199.68
Maintenance	4.5	70.2

Table 3-10. Operational Expenditure [1].

Lastly, the return of investment (ROI) must also be considered for a final economic analysis to understand the efficiency and profitability of the investment made for a recycling lithium battery process. A situation was studied by China Everbright Securities where they calculated the return of investment of unused LFP batteries, conducting a comparison between second-life batteries or recycled, relative to new batteries (Table 3-11) [1].

	1	r
Electricity price (€/kWh)	Used LFP Battery (%)	Unused LFP Battery (%)
0.06	6.10	-
0.07	8.29	1.70
0.08	10.35	3.86
0.09	12.37	5.91
0.09	14.32	7.88
0.10	16.24	9.77

Table 3-11. ROI comparison on LFP batteries [1].

As seen in the table above, a used LFP battery has a higher return of investment than an unused LFP battery, better catalogued as a new battery, as the prices of electricity increase. This shows how beneficial used batteries are to new batteries when analysing their ROI.

## 3.3 Second life battery

As previously mentioned, electric vehicles are expected to continue their growth in sales, and their batteries have a life expectancy of 8 to 10 years, which is when they reach 80% of their original capacity, hence no longer meeting the power range demand required by drivers. Therefore, second-life battery application needs to be taken into consideration to expand their life expectancy and usage, and at the same time, reduce waste. These end-of-life lithium batteries can have a second-life battery in

applications life, residential and commercial energy storage, grid stabilisation, portable energy storage, powertrains for low-speed vehicles, and others [26].

Also, it is expected that in 2025 second-life batteries can decrease in value between 30% and 70%, and stationary applications can surpass 200-gigawatt hours per year by 2030, resulting from a market evaluation of around €30 billion. However, this application could face challenges as new lithium batteries are expected to be cheaper overtime, and by 2040 the cost gap between a second-life battery and a new one could be 25% approximately [26].

### 3.3.1 Second-life battery economics

Electric vehicle owners may benefit from getting money for their spent batteries. A new industry that will focus only on re-purposing lithium batteries can be assembled, creating business opportunities. This new industry would oversee assessing, rearranging, and repackaging end-use batteries suitable for second-life application. Lastly, consumers will benefit the most from second-life batteries from a business standpoint since they will purchase a battery for energy storage system application at a cheaper price than a new battery, as shown in Figure 3-10 [29].

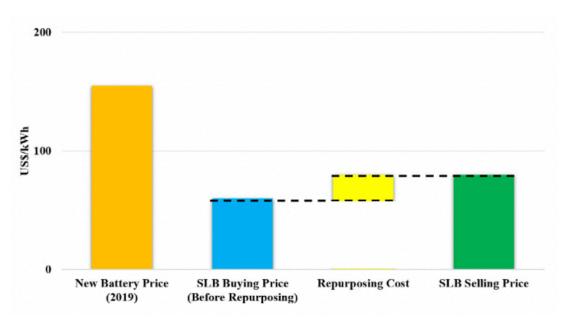


Figure 3-10. Battery price comparison [29].

Given that new battery prices are dropping, second-life batteries will always be cheaper than new ones. The price for a second-life battery must also include the cost of a new battery. The buying price can be calculated with the following formula [29].

$$SLB = BAT_{new} \times f_{health} \times (1 - f_{reuse} - f_{discount})$$
 Equation 3-1

Where;

*SLB*: buying second-life battery price in n<sup>th</sup> year.

 $BAT_{new}$ : new battery price of similar capacity in n<sup>th</sup> year.

 $f_{health}$ : SoH of the battery (%), if SoH not determined assume 3%.

 $f_{reuse}$ : second-life battery re-purposing cost (%), assume 15%.

 $f_{discount}$ : discount factor.

The re-purposing process, which means adapting the used battery for its new use as a SLB, can be divided into different costs. A study was performed to find the estimated cost of dismantling electric vehicle batteries at every part of the process. The battery chosen was from Smart For-Four battery with a capacity of 17.6 kWh. Also, the time and workforce required was catalogued throughout the process. The study presented its cost analysis in three stages: whole battery pack, module, and cell level. Transportation cost was neglected from the calculation. Refer to Figure 3-11 to understand the second-life re-purposing cost categories, Figure 3-12 for the cost distribution, and

Table 3-12 to know the study's cost analysis through the different steps from battery pack, module, and cell [29].



Figure 3-11. SLB re-purposing cost categories [29].

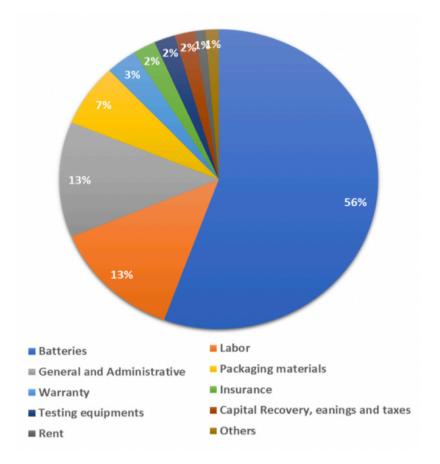


Figure 3-12. Re-purposing cost to selling price of second-life battery [29].

ltem	Battery pack (€)	Module (€)	Cell (€)
Removal from EV	117	117	117
Assessment	442	442	442
Disassembly to module	-	500	500
Disassembly to cell	-	-	275
Total	588	1,058	1,333
Cost / kWh	32	60	76

Table 3-12. Costs analysis [29].

In addition, a report by the Global Battery Alliance in 2018 mentioned that the selling price of the secondlife battery ranges from  $\in 60$  to  $\in 300$  per kilowatt-hour, and they projected a price drop in 2030 to  $\in 43$ /kWh selling price. Also, it is estimated that the break-even point is achieved if a second-life battery is 60% cheaper than a new lithium battery. However, in certain applications, second-life batteries are more economically favourable if the system is dependent on 50% depth of discharge (DOD), which means the capacity left on the battery, and competitive with new lead-acid batteries [29].

# 3.4 New lithium battery manufacturing

Back in 2010, the average price of a lithium-ion electric vehicle battery pack was €1,200 per kilowatthour, decreasing to €132 per kilowatt-hour in 2021, which is a price drop of 89% within eleven years. In addition, the cost of manufacturing a new lithium-ion EV battery cell component breakdown is shown in Table 3-13. The largest electric vehicle battery manufacturers have headquarters in Asia, and 80% of all cell manufacturing takes place in China [30].

EV Battery Cell component	Cost (%)
Cathode	51
Manufacturing and depreciation	23
Anode	12
Separator	7
Electrolyte	4
Housing and other materials	3

Table 3-13. Electric vehicle lithium-ion battery component breakdown [30].

In addition, the CAPEX cost of manufacturing new lithium batteries includes equipment, land, and building cost, for a total cost of UDS $\in$  4.6 billion for a 50 GWh facility. Out of the total cost, 85% comprises equipment cost. Also, the average operating cost is approximately around  $\in$  2.5 billion for the same facility previously mentioned. [31].

## 3.5 Residential Solar Panel

Stand-alone systems have been increasing worldwide, specifically across the United States of America, Europe, and Asia. Due to the benefits of reducing your electricity bill, benefiting from generating and consuming your own energy, helping the fight again climate change, and tax break benefits.

The number of panels required to be installed in a house varies between projects, depending on the surface area for the installation, available direct irradiance, household consumption, location, and other factors. However, the industry has been able to advance and make a standardization of the number of panels needed for installation depending on each of these factors as displayed below.

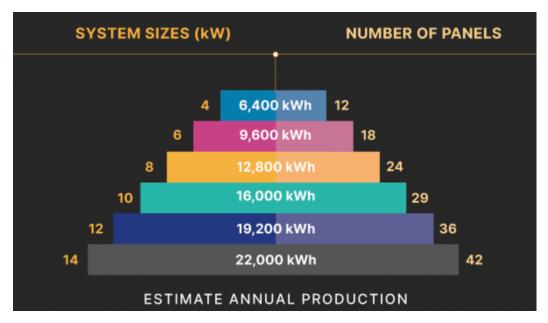


Figure 3-13. How many solar panels are needed to be installed depending on the system [32]

A common household typically needs between 20 to 24 solar panels to cover over 100% of the electricity consumption. This gives the homeowner an estimated annual production of approximately 12,800 kWh [32].

# 3.6 Analysis methods

The following methods were used in this thesis to analyse the most viable industry process between recycling lithium batteries, second-life batteries, and manufacturing a new lithium battery. The software used to perform the needed calculation was Microsoft Excel.

## 3.6.1 Net Present Value (NPV)

Net present value (NPV) is the difference between cash value of inflows at the present and the cash value of outflows at present as well over a period of time, in months or years, most typically years. The main use of net present value is in capital budgeting and investment planning to analyse how profitable a project can be or the investment. Also, the net present value results from the present value of future payments [33].

The formula to calculate net present value is the following:

$$NPV = \sum_{t=1}^{n} \frac{R_t}{(1+i)^t}$$
 Equation 3-2

#### Where;

 $R_t$ : net cash inflow – outflows during a single period t.

- i: discount rate
- t: number of periods

Therefore, NPV can let you know the time value of money and used to compare similar investment alternatives, like recycling batteries, second-life batteries, and manufacturing new batteries. Net present value depends on a discount rate that comes from the cost of the capital required for the project or investment. Also, if a net present value is negative, the project or investment must be discarded [33].

For the purpose of the analysis for this thesis report, it considers both the electricity generated and the electricity stored during the project's lifetime. Then, the investment cost will need to be deducted to get the total NPV value. This investment cost is the sum of the solar panels' system plus the cost of the type of battery that is being analyzed, either recycled, second-life, or a new lithium battery. The net cash inflow-outflow ( $R_t$ ) is calculated by multiplying the price of electricity in  $\in$ /kWh times the capacity of the specific project analysed and deducting the investment costs in solar panels and battery, resulting in the total savings in  $\in$ .

The discount rate, which is equal to the weighted average cost of capital (WACC), is a combination of the cost of capital from all possible sources, including debt, preferred shares, and common shares. The formula is the following [34].

$$WACC = \frac{E}{V} \times Re + \left(\left(\frac{D}{V} \times Rd\right) \times (1 - T)\right)$$
 Equation 3-3

Where;

- E : market value of company's equity
- D : market value of company's debt
- V : value of capital
- $\frac{E}{r}$ : percentage of capital that is equity
- $\frac{D}{T}$ : percentage of capital that is debt
- *Re* : cost of equity
- Rd : cost of debt
- T: tax rate

### 3.6.2 Equivalent Annual Cost (EAC)

The equivalent annual cost (EAC) is the annual cost of owning, maintaining, and operating the lithiumion battery for its lifetime. This method allows companies to compare the cost-effectiveness of many assets with different lifespans. Other companies use equivalent annual cost by calculating the optimal life of an asset, in this case a battery, to determine the best option between buying or renting, how the maintenance cost will impact the battery, the necessary cost savings by purchasing a new battery, and determining the cost of keeping the existing one [35].

Also, the cost of capital is the required return needed to make a budgeting project, like building a new factory, or in this case the most viable recycling method for a lithium battery. It also includes the cost of debt and equity. The lower the EAC value, the better the project is. The formula to calculate the equivalent annual cost is the following [35]:

$$EAC = \frac{AP \times i}{1 - (1 + i)^{-n}}$$
 Equation 3-4

Where;

AP : asset price

i: discount rate

n: number of periods

Furthermore, as explained before in the NPV, the asset price (AP) is the total cost of the battery, divided by the annuity, which is shown further along the report, giving the result in  $\in$ . Also, the discount rate would be the same as explained in the NPV above, which equals the WACC.

In addition, there is a relationship between net present value and equivalent annual cost, since the equivalent annual cost is equal to the net present value divided by the annuity factor, by considering the cost of capital and the number of years. The annuity factor formula is the following: [35]

Annuity Factor = 
$$\frac{1-\frac{1}{(1+r)^t}}{r}$$
 Equation 3-5

Where;

r : discount rate

t: number of periods

# **Chapter 4**

# Analysis

# 4.1 Viable industrial process analysis

### 4.1.1 Introduction

This section of the thesis report compares the findings between residential storage, commercial storage, and solar farm storage applications. Also, the assumptions taken throughout the analysis process will be explained. For residential storage, a lithium battery with a capacity of 65 kWh was analysed. The project at the Johan Cruyff Arena in Amsterdam of 2.8 MWh was used as a reference for commercial storage. Lastly, for the solar farm storage, the project called Tomatoh Abira Solar Park 2, which consists of a 64.6 MW solar farm with 19 MWh battery energy storage capacity, was used as a reference. The comparison between each application was analysed by calculating the Net Present Value for only the residential storage project taking into account the solar panel installation, and for the Equivalent Annual Cost a comparison between each project previously mentioned was calculated between different lifespans, as explained in the previous chapter. [36], [37].

#### 4.1.2 Assumptions

An assumption regarding the manufacturing price of recycled batteries was done with the following information gathered. An article by the World Electric Vehicle Journal named Factors Determining the Manufacturing Costs of Lithium-Ion Batteries for PHEVs, calculated the battery packs from the data created by simplified design programs. The costs were estimated to be fixed with the addition of weighted items. Some of the unit costs that were taken into account are the following [38]:

- Cell materials
- Cell purchased items
- Module and battery
- Direct labour
  - Electrode processing
  - Cell assembly
  - Formation cycling
  - Module and battery assembly
  - Rejected cell and scrap recycling
  - Receiving and shipping
- Administration

However, a summary chart was provided, as seen in Figure 4-1, where the assumption of the manufacturing cost was calculated [38].

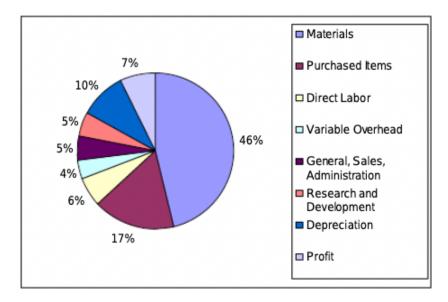


Figure 4-1. Summary of Unit Cost for Manufacturing Baseline Batteries [38].

With the information provided in Figure 4-2, the manufacturing cost could be calculated. Since materials are 46% of the unit cost for manufacturing baseline batteries, purchased items, direct labour, variable overhead, general sales, administration, depreciation, and profit will take part in the manufacturing cost, adding up to 49%. Research and development were neglected, since this section will correspond to a much lower percentage in recycled batteries. Also, the cathode taken into account for the pricing of the recycled lithium battery was LMO instead of LCO, from the case study mentioned in Chapter 3, to have an accurate value due to the fact that the unit cost of LCO is 35 €/kg, and LMO is 10 €/kg [39].

In addition, as shown in Table 3-13 in Chapter 3, knowing the percentage of what makes up the cost of lithium-ion batteries, the same distribution was assumed for each application making it a distribution regarding the weight of the battery, in kilograms, where 1 kWh battery would weight 9 kilograms approximately.

Furthermore, it was assumed that the manufacturing of recycled batteries would be entirely based on recycled materials, even though some companies in the industry use both new and recycled materials to cut costs, sometimes even by half.

Also, for Net Present Value and Equivalent Annual Cost calculations the discount rate, which is equal to WACC, it was assumed that the cost of debt was 3.5%, cost of equity as 8%, the proportion of debt vs. equity was 50% each, and the tax to be equal to 20%; giving a discount rate of 5.4%. The lifetime will be studied from a 10-year application up to 30 years, showing the results every 5 years. These lifespans are for both the NPV calculation for the residential storage project and the EAC calculation for every type of battery analysed.

In addition, the currency conversion from United States Dollars (USD\$) to Euros ( $\in$ ) is approximately 1, due to the market's current price as of the 14<sup>th</sup> of July of 2022.

### 4.1.3 Recycling battery analysis

As previously mentioned, the cost distribution per weight would be the following, knowing that 1 kWh is 9 kg [40].

Description	Cost distribution for a 9 kg battery
Cathode	4.6
Manufacturing	2.1
Anode	1.1
Separator	0.6
Electrolyte	0.4
Battery housing	0.3

Table 4-1. Distribution of material costs of a battery in kilograms.

Then, after gathering the unit prices of the recovered materials from recycling batteries, as seen in Table 4-2. The cost of a new battery from recycled materials is shown in Table 4-3, by multiplying the unit price times the weight distribution of what makes the battery, as shown in Table 4-1. Given the unit prices of what makes the battery, to get the manufacturing unit price following what was explained in the assumptions section it resulted to adding up the unit costs that added up to  $\in$  14.9/kg, which is 46% as seen in the pie chart on Figure 4-1, and the 49%, which forms part of the manufacturing unit cost results to  $\in$  15.9/kg [39].

Material	Unit cost (€/kg)	
Aluminium	1.3	
Plastics	0.1	
LMO	10	
Electrolyte solvents	0.15	
Graphite	0.28	

Table 4-2. Battery materials unit costs [39].

Table 4-3. Unit cost of the material distribution for a new battery.

Description	Unit cost (€/kg)
Cathode	10
Manufacturing	15.9
Anode	0.28
Separator	0.1
Electrolyte	0.15
Battery housing	1.3

Therefore, to calculate the price of the recycled battery for each of the applications studied for this analysis given that 1 kWh is 9 kg, Table 4-4 gives the weight of each application as well as the price following the distribution and unit cost of the materials as previously shown. Refer to Annex B for a more complete calculation breakdown.

Application	Battery capacity (kWh)	Weight (kg)	Price (€)
Residential Storage	65	585	4,730
Johan Cruyff Arena	2,800	25,200	203,715
Solar Farm	19,000	171,000	1,382,355

Table 4-4. Total price of recycled batter per application.

#### 4.1.4 Second life battery analysis

Given that the range of price for second-life lithium battery is  $40-160 \notin kWh$ , a  $65 \notin kWh$  price was chosen to calculate the price of second-life lithium battery for each application, because of the kind of battery that would be recovered to use as a second life to keep the same market difference as the new lithium-ion battery for this analysis. Hence, the total price of the second-life battery will be the multiplication of the battery price ( $\notin kWh$ ) by the capacity (kWh). Table 4-5 shows the total price for each application studied in this analysis.

Table 4-5. Total price of second-life battery.

Application	Second-life battery price (€/kWh)	Battery capacity (kWh)	Price (€)
Residential Storage	65	65	4,225
Johan Cruyff Arena	65	2,800	182,000
Solar Farm	65	19,000	1,235,000

#### 4.1.5 New lithium battery analysis

The same procedure as for second-life lithium battery was performed for the new lithium battery analysis given that the price of a new lithium battery is  $132 \notin kWh$ , as previously mentioned in Chapter 3, which correlates to the difference in price between a new lithium battery and a second life one shown in Figure 3-10 in Chapter 3, which shows a 47% difference in price. There is a 51% difference in price between 65  $\notin kWh$  and 132  $\notin kWh$ , as chosen for the analysis, and the difference is close enough to the market. Also, the same calculation for the total price of a new lithium battery was carried out for the second-life battery. Therefore, the price of new lithium batteries is shown in Table 4-6.

Application	New lithium battery price (€/kWh)	Battery capacity (kWh)	Price (€)
Residential Storage	132	65	8,580
Johan Cruyff Arena	132	2,800	369,600
Solar Farm	132	19,000	2,508,000

Table 4-6. Total price of a new lithium battery.

#### 4.1.6 PV solar panels analysis

As mentioned in Chapter 3, a typical stand-alone system consists of 20 to 24 solar panels providing an estimated annual production of 12,800 kWh. Considering these parameters and the market value for an 8-kW system size, it varies between  $\in$ 7,500 to  $\in$ 11,200, and the total value used for this analysis would be the value of the two values giving a total system cost of  $\in$ 9,250, taking into account installation, inverters, cabling, and any other cost [41].

## 4.2 Net Present Value results

Firstly, the NPV of the solar panels was calculated, for a 30-year system, taking into account a yearly degradation of the solar panels of 0.6%, the cost of electricity is  $0.24 \in /kWh$  [42], which is assumed to increase 2% for the first 5 years and then stays stagnant for the remainder of the project's lifetime. The cash-flows will be the savings created by both the generation from the photovoltaic system in the house and the energy stored in the battery that will be consumed at night, hence that amount that is used from the battery rather than buying it from the grid will also account as savings. These savings are calculated by the multiplication of the generation of electricity in kWh times the price of electricity in  $\in /kWh$  in the solar panels case, and for the battery would be the same process but multiplying the capacity of the battery times the cost of electricity, everything resulting in euros. Also, as previously mentioned, the discount rate equals WACC, which results in 5.8%.

The degradation of the batteries varies depending on the type of battery, 2% yearly for a recycled battery, 2.5% for a second-life battery, and 1.5% for a new lithium battery, affecting the total energy storage capacity. Then, after having calculated the entire savings from both the stand-alone photovoltaic system, and the battery, the discount factor, the only step left would be to add both the cost of the PV system plus the cost of the type of battery to be subtracted from the NPV results for a complete NPV value. Table 4-7 shows the results of the NPV calculation for the residential storage project with a recycled battery, second-life battery, and new lithium battery.

	PV with Recycled Battery	PV with Second Life Battery	PV with New Lithium Battery
	Total System Cost (€)	Total System Cost (€)	Total System Cost (€)
	13,980.00	13,475.00	17,830.00
Life span (yrs)	NPV (€)	NPV (€)	NPV (€)
10	30,317.14	30,504.82	26,625.80
15	51,524.27	51,511.69	47,933.06
20	71,857.65	71,644.80	68,366.57
25	91,317.27	90,904.17	87,926.32
30	109,903.14	109,289.78	106,612.32

After analysing the NPV results, it can be found that the recycled battery and second-life battery are the best options for this kind of project, since they are the ones that provide the most value and have the highest NPV. Furthermore, among all projects, the second-life battery is the one that adds the most value for residential storage use with a stand-alone PV system with 24 solar panels producing an estimated 12,800 kWh yearly, only for the first 10 years. For an application longer than that, the recycled battery becomes the best option, and the margin of difference between the two becomes greater as the years go by. Refer to Annex B for a full breakdown of the calculations.

#### 4.3 Equivalent Annual Cost results

Lastly, to complete the analysis, the Equivalent Annual Cost results must be compared to determine the most viable industrial process for each application, given that they have different demands. The first step would be to calculate the annuity factor for the different years of the application. The result is shown in Table 4-8.

Life span (yrs.)	Annuity Factor
10	7.57
15	10.10
20	12.05
25	13.55
30	14.69

Table 4-8.Annuity Factor.

Hence, to calculate the EAC of each project for the different type of battery, it is needed to take the cost of the battery, which was previously shown for every type of battery and project size and divide it by the

annuity factor for the different years during the life span. Table 4-9, Table 4-10, and Table 4-11 show the results for the recycled battery, second life battery, and new lithium battery, respectively.

	EAC of Recycled Battery (€)			
Life span (yrs.)	Residential Storage	Johan Cruyff Arena	Solar Farm	
10	624.51	26,896.93	182,515.31	
15	468.10	20,160.57	136,804.20	
20	392.53	16,905.58	114,716.73	
25	349.18	15,038.94	102,050.20	
30	321.86	13,862.25	94,065.51	

Table 4-9. EAC result of Recycled Battery (in €).

Table 4-10. EAC result of Second Life Battery (in €).

	EAC of Second life Battery (€)			
Life span (yrs.)	Residential Storage	Johan Cruyff Arena	Solar Farm	
10	557.84	24,029.85	163,059.71	
15	418.13	18,011.56	122,221.28	
20	350.62	15,103.53	102,488.26	
25	311.90	13,435.87	91,171.95	
30	287.50	12,384.61	84,038.40	

Table 4-11. EAC result of New Lithium Battery (in €).

	EAC of New Lithium Battery (€)			
Life span (yrs.)	Residential Storage Johan Cruyff Arena Solar		Solar Farm	
10	1,132.84	48,799.08	331,136.65	
15	849.12	36,577.31	248,203.21	
20	712.02	30,671.79	208,130.00	
25	633.41	27,285.14	185,149.19	
30	583.85	25,150.28	170,662.60	

As seen above, across every project, and even at the latest lifetime, of 30 years, the second-life battery has the lowest EAC value, making this type of battery the most viable industrial application. However, understanding that not every battery has the same lifespan, and 30 years may be too long, a recycled battery could be a better option depending on the project's lifetime. Since a recycled battery would last longer than a second life for a 20-year project, whereas a second-life battery could only last 15 years, a recycled battery has a lower Equivalent Annual Cost in that scenario, making it a more viable application. Refer to Annex B for an entire display of the calculations.

# **Chapter 5**

## Conclusion

Improving the recycling methods, reuse, and manufacturing of batteries are essential to decrease lithium battery waste. The increase in investment and development of recycling technologies represents an excellent opportunity to reduce the carbon footprint from manufacturing new lithium batteries and offer a sustainable solution to dispose of batteries. In addition, given that only 10% of lithium batteries are recycled, improving recycling methods and finding solutions for reuse or refurbishing is a great opportunity for a more sustainable world and a business opportunity at the commercial and industrial level.

Regarding recycling methods, direct recycling is expected to be the most profitable method over hydrometallurgical, and pyrometallurgical recycling. Refer to Annex A for a visual representation of the net recycling profit for commercial EV battery packs in countries such as China, South Korea, United States of America, Belgium, and the United Kingdom. Also, a table displaying the number of materials that can be recovered through each method is available in Annex A.

Three projects were analysed in this thesis report; these were residential, commercial, and solar farm storage. Within each project, three different applications were considered, recycled lithium battery, second life battery, and new lithium battery. In addition, the analysis was completed by conducting a Net Present Value analysis of the residential storage project, considering the installation of solar panels. Also, an Equivalent Annual Cost was calculated for every type of battery and all three projects at different years of their 30 years lifespan.

The NPV results showed an increase in value when adding either a recycled battery or a second-life battery for the residential project. However, out of the two best options, for this kind of project with the same scenario of 24 solar panels generating an estimated 12,800 kWh/year, the second-life battery had the higher NPV for a 10-year lifetime project only, but for a project with a longer lifetime a recycled battery has a higher NPV making it a better option for residential storage with a lifetime longer than 10 years. The difference in value kept becoming higher and higher as the project requested a higher lifetime.

Furthermore, the EAC of residential, commercial, and solar farm storage was also analysed and followed the same result as for the NPV. The recycled and second-life batteries showed the most prospect, although the second-life battery has the lowest EAC value, making it the most viable process for each of the project. However, given that with technology advances and understanding that not every battery has the same lifetime cycles, a recycled battery will have the edge over the second life battery, when the latter cannot provide a long lifetime, resulting on the recycled battery to have the lower EAC value in that scenario where it can have a longer life cycle.

In conclusion, adding a recycled or second-life battery to a house for energy storage with an existing solar system installed is a good investment since the NPV result is positive. However, the most viable industrial process among the projects studied in this dissertation is the second-life battery, because this application has the lowest Equivalent Annual Cost.

In the upcoming years, the battery passport is expected to be implemented more throughout the European Union and the World, providing a more efficient recycling process and better data gathering

regarding the status of the battery. Also, technological advances are expected to increase over the years, improving recycling methods and reducing carbon footprint. Data and research show that recycling batteries will overtake second-life batteries, and new batteries, as the most optimal and viable industrial process, since the majority of lithium supply is already in the household or being use by consumers. Hence, when people become aware and successful collection programs are implemented, along with technological advances, we could have a 100% recycled lithium battery and have a full circular value chain.

# Annex A

## Recycling benefits and profits

#### A.1 Material recovery

Direct	Hydrometallurgical	Pyrometallurgical
Copper	Copper	Copper compounds
Steel	Steel	Iron compounds
Aluminium	Aluminium	Co <sup>2+</sup> in output
Graphite	Graphite	Ni <sup>2+</sup> in output
Plastics	Plastics	Lithium compounds
LCO	Lithium carbonate	Aggregate (from slag)
NMC (111)	Co <sup>2+</sup> in output	
NMC (622)	Ni <sup>2+</sup> in output	
NMC (811)	Mn <sup>2+</sup> in output	
NCA	Electrolyte solvents	
LMO	Electrolyte salts	
LFP		
Electrolyte solvents		
Electrolyte salts		

Table 5-1. Recoverable materials through different recycling technologies [39].

#### A.2 Net recycling profits

An analysis was performed of the recycling process for EV LIBs in which a net recycled profit was calculated for direct, hydrometallurgical, and pyrometallurgical recycling (Figure 5-1) [43].

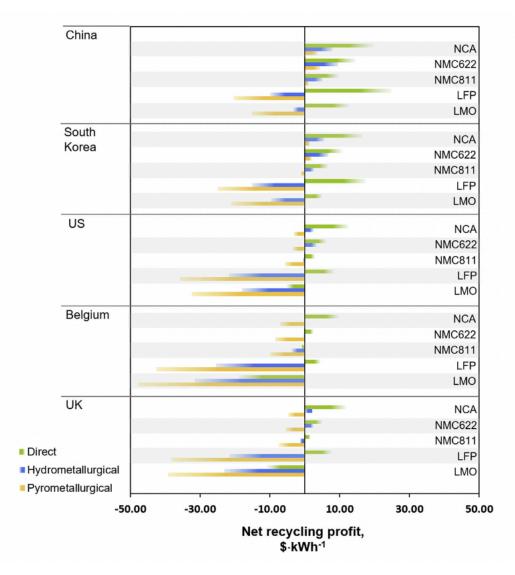


Figure 5-2. Net recycling profit for commercial EV battery packs [43].

# **Annex B**

## **Complete analysis calculations**

#### **B.1 NPV Calculations**

		Battery Weight (kg)		
Material	Percentage	585	25200	171000
Cathode	51%	298.35	12852	87210
Manufacturing	23%	134.55	5796	39330
Anode (graphite)	12%	70.2	3024	20520
Separator	7%	40.95	1764	11970
Electrolyte	4%	23.4	1008	6840
Battery housing (steel)	3%	17.55	756	5130

Table 5-2. Calculation of Recycled Battery cost.

Material	Unit Cost (Euro/kg)	Residential Storage (€)	Johan Cruyff Arena (€)	Solar Farm (€)
Cathode	10	2,983.50	128,520.00	872,100.00
Manufacturing	12.60	1,695.53	73,038.42	495,617.85
Anode (graphite)	0.28	19.66	846.72	5,745.60
Separator	0.1	4.10	176.40	1,197.00
Electrolyte	0.15	3.51	151.20	1,026.00
Battery housing (steel)	1.3	22.82	982.80	6,669.00
	TOTAL	4,729.11	203,715.54	1,382,355.45

Table 5-3. Recycled Battery manufacturing cost assumption.

Manufacturing Unit Cost Assumption			
11.83 46%			
12.60152174	49%		

Table 5-4. Discount rate calculations (WACC).

Cost of debt	3.5%
Cost of equity	8%
Debt vs. equity	50%
Tax	20%
WACC	5.4%

			Residential Storage	
Year	Degradation	Cost per kWh	Generation	Savings
1	0.6%	0.2400	12800	3072.00
2	0.6%	0.2448	12723.2	3114.64
3	0.6%	0.2497	12646.4	3157.76
4	0.6%	0.2547	12569.6	3201.35
5	0.6%	0.2598	12492.8	3245.43
6	0.6%	0.2598	12416	3225.47
7	0.6%	0.2598	12339.2	3205.52
8	0.6%	0.2598	12262.4	3185.57
9	0.6%	0.2598	12185.6	3165.62
10	0.6%	0.2598	12108.8	3145.67
11	0.6%	0.2598	12032	3125.72
12	0.6%	0.2598	11955.2	3105.77
13	0.6%	0.2598	11878.4	3085.81
14	0.6%	0.2598	11801.6	3065.86
15	0.6%	0.2598	11724.8	3045.91
16	0.6%	0.2598	11648	3025.96
17	0.6%	0.2598	11571.2	3006.01
18	0.6%	0.2598	11494.4	2986.06
19	0.6%	0.2598	11417.6	2966.11
20	0.6%	0.2598	11340.8	2946.16
21	0.6%	0.2598	11264	2926.20
22	0.6%	0.2598	11187.2	2906.25
23	0.6%	0.2598	11110.4	2886.30
24	0.6%	0.2598	11033.6	2866.35
25	0.6%	0.2598	10956.8	2846.40
26	0.6%	0.2598	10880	2826.45
27	0.6%	0.2598	10803.2	2806.50
28	0.6%	0.2598	10726.4	2786.54
29	0.6%	0.2598	10649.6	2766.59
30	0.6%	0.2598	10572.8	2746.64

Table 5-5. Solar Panels for Residential Storage, NPV calculations.

Table 5-6. Recycled Battery for Residential Storage, NPV calculations.

					Residentia	al Storage
Year	Degradation (%)	Degradation	Cost per kWh	Discount rate	Capacity	Savings
1	2.00	98.00	0.2400	5.40%	65	1528.80
2	2.00	96.00	0.2448	5.40%	65	1527.55
3	2.00	94.00	0.2497	5.40%	65	1525.64

4	2.00	92.00	0.2547	5.40%	65	1523.05
5	2.00	90.00	0.2598	5.40%	65	1519.73
6	1.00	89.00	0.2598	5.40%	65	1502.85
7	1.00	88.00	0.2598	5.40%	65	1485.96
8	1.00	87.00	0.2598	5.40%	65	1469.08
9	1.00	86.00	0.2598	5.40%	65	1452.19
10	1.00	85.00	0.2598	5.40%	65	1435.31
11	1.00	84.00	0.2598	5.40%	65	1418.42
12	1.00	83.00	0.2598	5.40%	65	1401.53
13	1.00	82.00	0.2598	5.40%	65	1384.65
14	1.00	81.00	0.2598	5.40%	65	1367.76
15	1.00	80.00	0.2598	5.40%	65	1350.88
16	1.00	79.00	0.2598	5.40%	65	1333.99
17	1.00	78.00	0.2598	5.40%	65	1317.10
18	1.00	77.00	0.2598	5.40%	65	1300.22
19	1.00	76.00	0.2598	5.40%	65	1283.33
20	1.00	75.00	0.2598	5.40%	65	1266.45
21	1.00	74.00	0.2598	5.40%	65	1249.56
22	1.00	73.00	0.2598	5.40%	65	1232.67
23	1.00	72.00	0.2598	5.40%	65	1215.79
24	1.00	71.00	0.2598	5.40%	65	1198.90
25	1.00	70.00	0.2598	5.40%	65	1182.02
26	1.00	69.00	0.2598	5.40%	65	1165.13
27	1.00	68.00	0.2598	5.40%	65	1148.24
28	1.00	67.00	0.2598	5.40%	65	1131.36
29	1.00	66.00	0.2598	5.40%	65	1114.47
30	1.00	65.00	0.2598	5.40%	65	1097.59

Table 5-7. Second Life Battery for Residential Storage, NPV calculations.

					Residentia	al Storage
Year	Degradation (%)	Degradation	Cost per kWh	Discount rate	Capacity	Savings
1	2.50	97.50	0.2400	5.40%	65	1521.00
2	2.50	95.00	0.2448	5.40%	65	1511.64
3	2.50	92.50	0.2497	5.40%	65	1501.30
4	2.50	90.00	0.2547	5.40%	65	1489.94
5	2.50	87.50	0.2598	5.40%	65	1477.52
6	1.00	86.50	0.2598	5.40%	65	1460.63
7	1.00	85.50	0.2598	5.40%	65	1443.75
8	1.00	84.50	0.2598	5.40%	65	1426.86
9	1.00	83.50	0.2598	5.40%	65	1409.98
10	1.00	82.50	0.2598	5.40%	65	1393.09
11	1.00	81.50	0.2598	5.40%	65	1376.20

12	1.00	80.50	0.2598	5.40%	65	1359.32
13	1.00	79.50	0.2598	5.40%	65	1342.43
14	1.00	78.50	0.2598	5.40%	65	1325.55
15	1.00	77.50	0.2598	5.40%	65	1308.66
16	1.00	76.50	0.2598	5.40%	65	1291.77
17	1.00	75.50	0.2598	5.40%	65	1274.89
18	1.00	74.50	0.2598	5.40%	65	1258.00
19	1.00	73.50	0.2598	5.40%	65	1241.12
20	1.00	72.50	0.2598	5.40%	65	1224.23
21	1.00	71.50	0.2598	5.40%	65	1207.34
22	1.00	70.50	0.2598	5.40%	65	1190.46
23	1.00	69.50	0.2598	5.40%	65	1173.57
24	1.00	68.50	0.2598	5.40%	65	1156.69
25	1.00	67.50	0.2598	5.40%	65	1139.80
26	1.00	66.50	0.2598	5.40%	65	1122.92
27	1.00	65.50	0.2598	5.40%	65	1106.03
28	1.00	64.50	0.2598	5.40%	65	1089.14
29	1.00	63.50	0.2598	5.40%	65	1072.26
30	1.00	62.50	0.2598	5.40%	65	1055.37

Table 5-8. New Lithium Battery for Residential Storage, NPV calculations.

					Residentia	al Storage
Year	Degradation (%)	Degradation	Cost per kWh	Discount rate	Capacity	Savings
1	1.75	98.25	0.2400	5.40%	65	1532.70
2	1.75	96.50	0.2448	5.40%	65	1535.51
3	1.75	94.75	0.2497	5.40%	65	1537.82
4	1.75	93.00	0.2547	5.40%	65	1539.60
5	1.75	91.25	0.2598	5.40%	65	1540.84
6	1.00	90.25	0.2598	5.40%	65	1523.96
7	1.00	89.25	0.2598	5.40%	65	1507.07
8	1.00	88.25	0.2598	5.40%	65	1490.18
9	1.00	87.25	0.2598	5.40%	65	1473.30
10	1.00	86.25	0.2598	5.40%	65	1456.41
11	1.00	85.25	0.2598	5.40%	65	1439.53
12	1.00	84.25	0.2598	5.40%	65	1422.64
13	1.00	83.25	0.2598	5.40%	65	1405.75
14	1.00	82.25	0.2598	5.40%	65	1388.87
15	1.00	81.25	0.2598	5.40%	65	1371.98
16	1.00	80.25	0.2598	5.40%	65	1355.10
17	1.00	79.25	0.2598	5.40%	65	1338.21
18	1.00	78.25	0.2598	5.40%	65	1321.32

19	1.00	77.25	0.2598	5.40%	65	1304.44
20	1.00	76.25	0.2598	5.40%	65	1287.55
21	1.00	75.25	0.2598	5.40%	65	1270.67
22	1.00	74.25	0.2598	5.40%	65	1253.78
23	1.00	73.25	0.2598	5.40%	65	1236.90
24	1.00	72.25	0.2598	5.40%	65	1220.01
25	1.00	71.25	0.2598	5.40%	65	1203.12
26	1.00	70.25	0.2598	5.40%	65	1186.24
27	1.00	69.25	0.2598	5.40%	65	1169.35
28	1.00	68.25	0.2598	5.40%	65	1152.47
29	1.00	67.25	0.2598	5.40%	65	1135.58
30	1.00	66.25	0.2598	5.40%	65	1118.69

	PV with Recycled Battery	PV with Second Life Battery	PV with New Lithium Battery
	Total System Cost (€)	Total System Cost (€)	Total System Cost (€)
	13,980.00	13,475.00	17,830.00
Life span (yrs)	NPV (€)	NPV (€)	NPV (€)
10	30,317.14	30,504.82	26,625.80
15	51,524.27	51,511.69	47,933.06
20	71,857.65	71,644.80	68,366.57
25	91,317.27	90,904.17	87,926.32
30	109,903.14	109,289.78	106,612.32

#### B.2 EAC Calculations

Table 5-10.	Annuity factor	calculations.
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Life span (yrs.)	Annuity
10	7.57
15	10.10
20	12.05
25	13.54
30	14.69

	Residential Storage	Johan Cruyff Arena	Solar Farm
Cost of battery	€4,730.00	€203,715.00	€1,382,355.00
Life span (yrs.)	EAC	EAC	EAC
10	€624.51	€26,896.93	€182,515.31
15	€468.10	€20,160.57	€136,804.20
20	€392.53	€16,905.58	€114,716.73
25	€349.18	€15,038.94	€102,050.20
30	€321.86	€13,862.25	€94,065.51

Table 5-11. Recycled Battery EAC calculations.

Table 5-12. Second Life Battery EAC calculations.

	Residential Storage	Johan Cruyff Arena	Solar Farm
Cost of battery	€4,225.00	€182,000.00	€1,235,000.00
Life span (yrs.)	EAC	EAC	EAC
10	€557.84	€24,029.85	€163,059.71
15	€418.13	€18,011.56	€122,221.28
20	€350.62	€15,103.53	€102,488.26
25	€311.90	€13,435.87	€91,171.95
30	€287.50	€12,384.61	€84,038.40

Table 5-13. New Lithium Battery EAC calculations.

	Residential Storage	Johan Cruyff Arena	Solar Farm
Cost of battery	€8,580.00	€369,600.00	€2,508,000.00
Life span (yrs.)	EAC	EAC	EAC
10	€1,132.84	€48,799.08	€331,136.65
15	€849.12	€36,577.31	€248,203.21
20	€712.02	€30,671.79	€208,130.00
25	€633.41	€27,285.14	€185,149.19
30	€583.85	€25,150.28	€170,662.60

### References

- [1] M. C. C. Lima, L. P. Pontes, A. S. M. Vasconcelos, W. d. A. Silva Junior and K. Wu, "Economic Aspects for Recycling of Used Lithium-Ion Batteries from Electric Vehicles," *Energies*, vol. 15, p. 2203, 2022.
- [2] K. Tomaszewski, "Lithium-ion Battery Recycling: Benefits and Risks Analyzed," 2020. [Online]. Available: https://www.cirbasolutions.com/news/lithium-ion-battery-recycling-benefits-and-risksanalyzed/.
- [3] "What is a second life battery?," Enel X, 2022. [Online]. Available: https://corporate.enelx.com/en/question-and-answers/what-is-second-life-battery.
- [4] "European Compliant EV Battery Passport," 2021. [Online]. Available: https://everledger.io/industry-solutions/batteries/ev-battery-passport-eu-regulations/.
- [5] J. Unwin, "What are lithium batteries and how do they work?," 06 February 2020. [Online]. Available: https://www.power-technology.com/analysis/what-are-lithium-batteries-madeof/#:~:text=A%20lithium%20battery%20is%20formed,are%20stored%20in%20the%20anode.. [Accessed June 2022].
- [6] "Solid-state batteries: the new frontier of electrification?," 2022. [Online]. Available: https://www.flashbattery.tech/en/how-solid-state-batteries-work/. [Accessed 2022].
- [7] S. Windisch-Kern, E. Gerold, T. Nigl, A. Jandric, M. Altendorfer, B. Rutrecht, S. Scherhaufer, H. Raupenstrauch, R. Pomberger, H. Antrekowitsch and F. Part, "Recycling chains for lithium-ion batteries: A critical examination of current challenges, opportunities and process dependencies," *Elsevier*, vol. 138, pp. 125-139, 2022.
- [8] "What is Lithium Battery Technology?," Northern Arizona Wind & Sun, [Online]. Available: https://www.solar-electric.com/learning-center/lithium-battery-technology.html/. [Accessed June 2022].
- [9] "Lithium Ion Battery Advantages & Disadvantages," Electronicsnotes, 2022. [Online]. Available: https://www.electronics-notes.com/articles/electronic\_components/battery-technology/li-ion-

lithium-ion-advantages-disadvantages.php. [Accessed May 2022].

- [10] B. Hyuntae and K. Youngsik, "Technologies of lithium recycling from waste lithium ion batteries: a review," *Royal Society of Chemistry,* vol. 2, pp. 3234-3250, 2021.
- [11] C. Costa, J. Barbosa, R. Gonçalves, H. Castro, F. Del Campo and S. Lanceros-Méndez, "Recycling and environmental issues of lithium-ion batteries: advances, challenges and opportunities," *Elsevier*, vol. 37, pp. 433-465, 2021.
- [12] J. Murray, "Is the Nobel Prize-winning lithium-ion battery really having a positive impact on the environment?," NS Energy, 2019. [Online]. Available: https://www.nsenergybusiness.com/features/lithium-ion-battery-environmentalimpact/attachment/untitled-design/. [Accessed June 2022].
- [13] T. Chen, Y. Jin, H. Lv, A. Yang, M. Liu, B. Chen, Y. Xie and Q. Chen, "Applications of Lithium-Ion Batteries in Grid-Scale Energy Storage Systems," *Springer*, vol. 26, pp. 208-217, 2020.
- [14] "World Energy Resources," World Energy Council, 2016. [Online]. Available: https://www.worldenergy.org/. [Accessed July 2022].
- [15] "Types of Electric Vehicles EV Architecture," John Wiley & Sons, Ltd., 2012.
- [16] "Types of Electric Cars and Working Principles," Omazaki, 2022. [Online]. Available: https://www.omazaki.co.id/en/types-of-electric-cars-and-working-principles/. [Accessed May 2022].
- [17] A. Paullikkas, "Sustainable options for electric vehicle technologies," *Elsevier*, vol. 41, pp. 1277-1287, 2015.
- [18] C. Iclodean, B. Varga, N. Burnete, D. Cimerdean and B. Jurchis, "Comparison of Different Battery Types for Electric Vehicles," *IOP Publishing Ltd*, vol. 252, 2017.
- [19] J. Kurtz, S. Sprik, G. Saur and S. Onorato, "Fuel Cell Electric Vehicle Durability and Fuel Cell Performance," National Renewable Energy Laboratory, 2019.
- "Lithium-ion Battery Market Size, Share & Trends Analysis Report By Product (LCO, LFP, NCA, LMO, LTO, NMC), By Application (Consumer Electronics, Energy Storage Systems, Industrial), By Region, And Segment Forecasts, 2022 2030," Grand View Research, 2020.
- [21] N. Nhede, "The importance of Li-ion recycling and what will drive it?," Smart Energy International, 17 September 2020. [Online]. Available: https://www.smart-energy.com/storage/the-importance-

of-li-ion-battery-recycling-and-what-will-drive-it/. [Accessed June 2022].

- [22] C. Hanisch, J. Diekmann, A. Stieger, W. Haselrieder and A. Kwade, "Recycling of Lithium-Ion Batteries," *Handbook of Clean Energy Systems*, 2015.
- [23] H. Bin, P. Zhefei, X. Su and L. An, "Recycling of lithium-ion batteries: Recent advances and perspectives," *Journal of Power Sources,* vol. 399, pp. 274-286, 2018.
- [24] P. Makwawimba, M. Tang, Y. Peng, S. Lu, L. Zheng, Z. Zhao and A.-g. Zhen, "Assessment of recycling methods and processes for lithium-ion batteries," *iScience*, vol. 25, no. 5, 2022.
- [25] A. Battistel, M. S. Palagonia and D. Brogioli, "Electrochemical Methods for Lithium Recovery: A Comprehensive and Critical Review," *Wiley Online Library*, vol. 32, no. 23, 2020.
- [26] H. Engel, P. Hertzke and G. Siccardo, "Second-life EV batteries: The newest value pool in energy storage," 30 April 2019. [Online]. Available: https://www.mckinsey.com/industries/automotive-and-assembly/our-insights/second-life-evbatteries-the-newest-value-pool-in-energy-storage. [Accessed July 2022].
- [27] A. Reis, "New method to recycle lithium-ion batteries," European Scientist, 01 July 2021.
  [Online]. Available: https://www.europeanscientist.com/en/energy/new-method-to-recycle-lithium-ion-batteries/. [Accessed August 2022].
- [28] P. Anh, "The recyclability of lithium-ion battery materials," The University of Queensland, 2019.
- [29] M. Mohammed, J. Woon Lee, G. Ramasamy, E. E. Ngu, S. P. Thiagarajah and Y. H. Lee, "Feasibility of utilising second life EV batteries: Applications, lifespan, economics, environmental impact, assessment, and challenges," *Elsevier*, vol. 60, no. 5, pp. 4517-4536, 2021.
- [30] G. Bhutada, "Breaking Down the Cost of an EV Battery Cell," Visual Capital, 22 February 2022.
  [Online]. Available: https://www.visualcapitalist.com/breaking-down-the-cost-of-an-ev-battery-cell/. [Accessed July 2022].
- [31] T. Sarkar, B. Verma, E. Mandal and M. Dixit, "Indigenisation of Lithium-ion Battery Manufacturing: A Techno-economic Feasibility Assessment," *CSTEP*, 2018.
- [32] J. Marsh, "How many solar panels do I need for my home?," EnergySage, 13 September 2022.
  [Online]. Available: https://news.energysage.com/how-many-solar-panels-do-i-need/.
  [Accessed September 2022].
- [33] J. Fernando, "Net Present Value (NPV)," Investopedia, 05 August 2021. [Online]. Available:

https://www.investopedia.com/terms/n/npv.asp. [Accessed August 2022].

- [34] C. Team, "WACC," CFI, 23 January 2022. [Online]. Available: https://corporatefinanceinstitute.com/resources/knowledge/finance/what-is-wacc-formula/. [Accessed September 2022].
- [35] W. Kenton, "Equivalent Annual Cost EAC Definition," Investopedia, 23 August 2020. [Online]. Available: https://www.investopedia.com/terms/e/eac.asp#:~:text=Understanding%20the%20Equivalent% 20Annual%20Cost,are%20the%20most%20relevant%20variable.. [Accessed August 2022].
- [36] A. Colthorpe, "Solar farm fitted with batteries to meet grid output control requirements goes online in Japan," Energy Storage News, 7 July 2020. [Online]. Available: https://www.energystorage.news/solar-farm-fitted-with-batteries-to-meet-grid-output-control-requirements-goesonline-in-japan/. [Accessed August 2022].
- [37] C. Hammerschmidt, "Second life for traction batteries in Amsterdam football arena," eeNews Power Manage,ent, 29 June 2018. [Online]. Available: https://www.eenewspower.com/en/second-life-for-traction-batteries-in-amsterdam-footballarena/. [Accessed August 2022].
- [38] P. Nelso, D. Santini and J. Barnes, "Factors Determining the Manufacturing Costs of Lithium-Ion Batteries for PHEVs," *World Electric Vehicle Journal*, 2009.
- [39] Q. Dai, J. Spangenberger, S. Ahmed, L. Gaines, J. Kelly and M. Wang, "EverBatt: A Closedloop Battery Recycling Cost and Environmental Impacts Model," Argonne National Laboratory, 2019.
- [40] Aentron, "Aentron," [Online]. Available: https://aentron.com/products/1kwh-batteries/. [Accessed 21 September 2022].
- [41] S. P. Melbourne, "WHY CHOOSE AN 8KW SOLAR ENERGY SYSTEM FOR YOUR HOME," Trione Energy, [Online]. Available: https://www.trione.com.au/why-choose-8kw-solar-systemfor-your-home/. [Accessed September 2022].
- [42] EPRS, "Electricity prices for household and non-household consumers," European Parliamentary Research Service, 2021. [Online]. Available: https://epthinktank.eu/2022/06/16/monitoring-the-energy-situation-in-the-eu-june-2022/electricity-prices-for-household-and-non-householdconsumers/#:~:text=The%20average%20EU%20price%20for,per%20kWh%2C%20without%2 0taxes).. [Accessed September 2022].

[43] L. Lander, T. Clever, M. A. Rajaeifar, V. Nguyen-Tien, R. J. Elliot, O. Heidrich, E. Kendrick, J. S. Edge and G. Offer, "Financial viability of electric vehicle lithium-ion battery recycling," *iScience*, vol. 24, no. 7, 2021.