

THE CHALLENGES AND ENVIRONMENTAL JUSTIFICATION OF RECYCLING Li-ION ELECTRIC VEHICLES BATTERIES

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Abstract: Efforts to reduce CO₂ emissions in road transportation will lead to an estimated thirty fold increase of the number of EVs by 2030, and consequently an estimated 12.85 million tons of Li-ion batteries from EVs will go offline between 2021 and 2030. Therefore, end-of-life, waste management schemes should be put in place to successfully deal with the battery ‘waste’. Knowing full well that bringing down the levels of GHG emissions in the atmosphere is the only way to truly mitigate the destructive effects of climate change and the recycling industry has an important role to play in the effort, this paper aimed at reviewing different Li-ion EV battery recycling procedures. The findings showed that recycling Li-ion batteries helps prevent the shortage of critical minerals from a mass flow perspective, however, from an environmental perspective, the current available technologies are not recommended to recover the base materials from Li-ion batteries since they lead to more consumption of energy and higher air emissions than primary production.

Key words: GHG emissions; waste management; circular economy; materials recovery

ПРЕДИЗВИЦИТЕ И ОПРАВДАНОСТА НА РЕЦИКЛИРАЊЕТО НА ЛИТИУМСКИТЕ-ЈОНСКИ БАТЕРИИ КОИ СЕ СРЕЌАВААТ КАЈ ЕЛЕКТРИЧНИТЕ ВОЗИЛА

Апстракт: Напорите за намалување на емисиите на CO₂ од патниот транспорт ќе доведат до триесекратно зголемување на бројот на електрични возила (EVs) до 2030 година, а следствено на тоа околу 12.85 милиони тони литиумски батерии ќе завршат како отпад во периодот од 2021 до 2030 година. Токму затоа се јавува потребата од воспоставување методологија за управување со животниот век на EVs која би водела кон успешно справување со овој отпад. Знаејќи дека намалувањето на емисиите на стакленичките гасови во атмосферата е единствениот начин да се ублажат деструктивните ефекти од климатските промени, рециклирањето игра важна улога во овој процес. Па така, овој труд имаше за цел да ги согледа предностите и недостатоците на различните постапки за рециклирање на батериите од EVs. Резултатите од анализата покажуваат дека рециклирањето на литиумските јонски батерии е значајно поради тоа што на тој начин се штеди на критичните суровини, но од еколошка перспектива технологиите достапни во моментот не се препорачуваат за извлекување на базичните суровини од литиумските јонски батерии, бидејќи самите постапки за рециклирање водат до голема потрошувачка на енергија и до ослободување поголемо количество стакленички гасови во воздухот отколку што би се ослободило со примарното производство.

Клучни зборови: емисии на стакленички гасови; управување со отпад; циркуларна економија; поврат на примарните суровини

1. INTRODUCTION

Electric vehicles (EVs) seem to provide an elegant answer to the transport sector’s CO₂ emissions and efforts and policies aimed to reduce them

will lead to an estimated thirtyfold increase of the number of EVs by 2030 [1]. However, as production increases, the resulting waste at the end of the EVs useful lives should also be kept in check. The battery packs are one of the first things that go to

waste, retiring from their automotive application once their metric state of health drops to 80% [2, 3]. Projecting that 12.85 million tons of lithium-ion (Li-ion) EV batteries will go offline between 2021 and 2030 and being aware of the EV industry's mineral supply risk, reuse value, and the carbon offset potential of circular economies for Li-ion batteries, the success of the automotive industry and the electric mobility sector will depend on recycling.

Materials that can be recovered fall into one of these groups: lithium salts, cobalt, aluminium, and other (steel, plastic and various metals). In an analysis from 2014 [4], an optimistic assumption of 100% extraction of each of these materials is used and the results show that (in the current commodities market) the costs far outweigh the benefits of recycling Li-ion batteries [5]. However, the benefits of Li-ion battery recycling might lay with environmentalism and the evolving closed-loop supply chains in which the content recovered by recycling will be returned to the battery manufacturing process.

Moreover, bringing down the global GHG levels is the only way to mitigate the destructive effects of climate change and the recycling industry has an important role to play in the effort. This paper aims at reviewing different Li-ion EV battery recycling procedures to find out how energy and carbon intensive each of these procedures is and pinpoint if the current material recovery methods are entirely environmentally friendly or not.

2. MATERIALS

Li-ion battery life

Li-ion batteries store high amounts of energy in addition to being lightweight and having a cost-efficient production, all of which have forecast them to have the highest potential for future energy storage technology. Lithium's use in HEVs and EVs is relatively new, but supporters claim 10–20 years battery life [6]. This is a longer life expectancy than other battery packs, a desirable feature for cost-conscious consumers, and they can charge and discharge quicker, while less likely to lose their charge when not used.

The most commonly used Li-ion batteries in EVs are the NMC (Lithium Nickel Manganese Cobalt Oxide) and NCA (Lithium Nickel Cobalt Aluminium Oxide) batteries (Table 1). Based on the type the share of the base metals like cobalt (5–

20%), nickel (5–10%), and lithium (5–7%) may vary [7]. NMC have two major advantages: high specific energy and low cost, but they are moderate in terms of specific power, safety, lifespan, and performance when compared to the other types of Li-ion batteries. The other commonly used type, the NCA battery, offers one strong advantage, and that is high specific energy. It is moderate in the rest of the characteristics like performance, cost, specific power, and lifespan, and its popular use in EVs and especially HEVs can confirm that. The main downside to this battery type is its low level of safety and potential to catch fire.

Another often-used Li-ion battery type is the Lithium-Iron-Phosphate (LFP) battery. It benefits from higher energy density than both NMC and NCA batteries, meaning that a single NMC/NCA battery cell will require twice as much room as an LFP battery, which is very important for vehicles with limited space. However, the charging efficiency of NMC/NCA lithium batteries is higher than that of LFP batteries. But at the end, the life cycle of a LFP battery is better than a NMC/NCA lithium battery. The theoretical life of a NMC lithium battery is 2000 cycles, but its capacity fades to 60% when it runs 1000 cycles; even the best-known Tesla NCA battery can only maintain 70% of its capacity after 3000 cycles, while the LFP battery will remain at 80% after 3000 charging cycles [9].

Table 1

Types of cathodes in Li-ion batteries, their developers and vehicle application [8]

Types of cathodes	Developers	Vehicle application
Nickel, cobalt, and aluminium (NCA)	Johnson Controls, Panasonic	Mercedes Benz S400 Hybrid, Tesla Model S
Manganese	LG Chem, NEC	Chevrolet Volt, Nissan Leaf
Iron-nano-phosphate	A123 Systems	Fisker Karma, Chevrolet Spark
Nickel, manganese, and cobalt (NMC)	EnerDel	THINK City electric vehicle

Battery components

Before understanding what a Li-ion battery is composed of, an explanation of a typical EV battery cell is required. The battery cell consists of two electrodes that sit in an ion-rich solution called electro-

lyte. A polymer film, called a separator, separates the two electrodes which are positioned very close to each other, thereby preventing a short circuit. The battery cannot function without any of these four main components (cathode, anode, electrolyte and separator) (Figure 1).

The cathode, being composed of layered transition metal oxides, determines the capacity and the voltage of the battery. If a cathode has a higher amount of lithium, then its capacity will be bigger. In Li-ion batteries, the cathode is made of lithium oxide since lithium in the element form is unstable. That means that lithium oxide is used as an active material which reacts chemically to produce electrical energy when the cell discharges. A layer of aluminium is used to hold the frame of the cathode coated with a compound made up of active material,

conductive additive, and a binder. The active material contains lithium ions; the conductive additive is added to increase the conductivity; and the binder acts as an adhesive which enables well setting on the aluminium substrate.

The anode is composed of graphite and other conductive additives. Its substrate is also coated with active material; the active material enables electric current to flow through the external circuit, while allowing reversible absorption or emission of lithium ions released from the cathode. Graphite is used in the anode because of its stable structure, and the substrate is coated with active material, conductive additive, and a binder.

The entire product is saturated in the electrolyte solution, consisting of lithium-salts, additives and organic solvents.

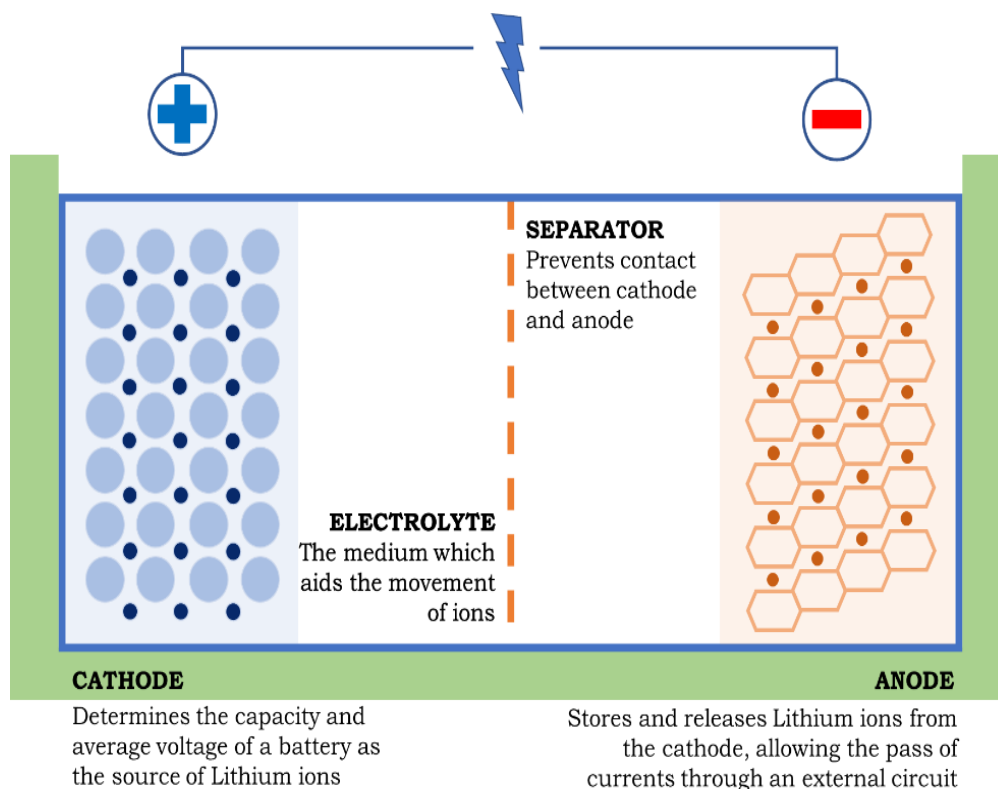


Fig. 1. Li-ion battery components

The entire product is saturated in the electrolyte solution, consisting of lithium-salts, additives and organic solvents. The absolute barrier between the electrodes, the separator, together with the electrolyte determines the safety of the battery since it acts as a physical barrier between the electrodes. The separator is made of synthetic resin such as polyethylene and polypropylene, and has microscopic holes on it, through which it lets the ions pass.

Li-ion batteries can overheat, so they are built with safety vents, thermal interrupters, and other features, such as a centre pin to provide structural stability and to prevent short circuits. At the end of the battery components' production, every part is sealed in a casing (usually made of steel or aluminium) for a battery cell to be created. Once the battery cell is complete, several cells are arranged to form a battery module and the EV battery is literally a "battery pack" that consists of many battery modules.

3. METHODS

The recyclability of a material mainly depends on its ability to reacquire the properties it had in its original state, and in the Li-ion batteries' case, achieving this is complicated. Currently, the automotive advanced-battery recycling field involves mostly projects using test batteries, batteries reclaimed from wrecked vehicles and a few thousand EV batteries that have failed for a variety of reasons. Anyhow, eventually each cell of every produced battery will be unable to support any application and therefore it should get recycled. If not recycled, EV batteries may be dumped, incinerated, or exported,

by which they will contaminate the environment and threaten public health. The problem is that due to the expensive needs of the process and complex subprocesses, EV battery recycling is not yet achieved at a high scale.

Separating the mixture of chemical components of the anode and cathode materials in Li-ion batteries requires advanced chemical and physical methods which make the recycling process particularly challenging. However, these methods enable separation into pure materials with no or limited contamination in a safe and efficient process. Sometimes as Figure 2 demonstrates combination of multiple methods ensures a higher rate of recyclability.

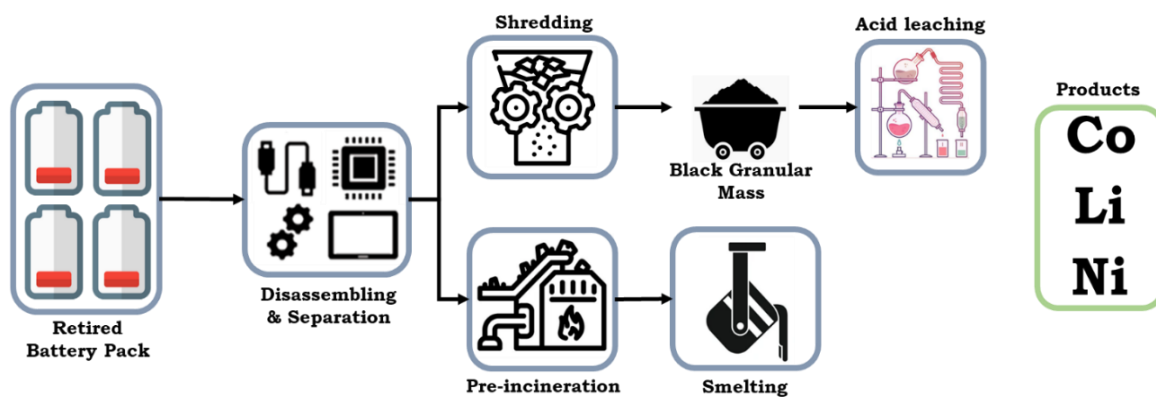


Fig. 2. Flow-chart of the Li-ion batteries' recycling process

Physical Li-ion battery recycling processes

Among the physical processes for the recycling of spent Li-ion batteries, two recycling methods have been especially popular in the past decade and include thermal treatment or mechanical separation of the components.

The thermal processes are generally associated with the production of steel, ferro-alloys, or other metal alloys. They usually consist of furnace heating in a controlled atmosphere between 100 and 150°C to separate out the insoluble organic additives and adhesives. Heating time is not standardized, but it does not exceed two hours [10]. These types of processes often include smelting, meaning recovering basic elements or salts at high temperatures. The smelting process is operational on a large scale, since multiple types of batteries can be smelted including Li-ion batteries. Smelting enables recovery of the valuable metals only up to a point, but the idea is that later the metals can be sent for refining in order for the product to be suitable for multi-purpose use. When smelting, only 25% of the materials are restored, and include mostly copper, cobalt, and

nickel. The other materials, including lithium, are contained in the slag.

Another type of thermal process involves pre-incinerating the battery to regain the electrolyte and a small amount of graphite, which together comes down to about 40% of the battery set. A traditional pyrometallurgical method, often used in the industry, is the ultra-high temperature method (UHT), which involves smelting and purification. During UHT, spent Li-ion batteries are usually smelted with other types of batteries (e.g., Nickel Metal Hydride), or ores and industrial wastes.

In a different thermal processing method known as roasting reduction (RR) method, a pre-treated battery pack is roasted at a mild temperature lower than 1000°C under oxygen-free conditions, by which the cathode materials are converted to lithium carbonate, metal, and metal oxide by carbothermal reduction. Most used RR methods are the ones where the batteries are roasted under N₂ atmosphere (RR-N₂) and vacuum conditions (RR-Vac).

While thermal treatment is advantageous, as the operations necessary are simple and convenient,

the process has been linked to high emissions of dioxins, chloride compounds and mercury, making it necessary to install special equipment to purify the gases and smoke caused by combustion. Additionally, pyrometallurgy, and primarily smelting, generates more GHG emissions than it saves.

The goal of the mechanical separation processes is to separate materials according to their different properties such as density, conductivity, magnetism, etc. So far, the most efficient and cost-effective way of mechanical recycling is by disassembling. The main challenge in the disassembly process stems from the large number of battery pack designs, which vary in size, electrode chemistry and form. The second major challenge is that the current technology is highly assisted by humans, which ends up being very high-priced. Mechanical separation processes are usually applied as pre-treatment, meaning they are the first applied to treat the outer cases of the batteries and will later lead to a hydrometallurgical or a pyrometallurgical recycling process.

A Li-ion battery recycling company from Canada, called Li-cycle, uses a mechanically dominant recycling process designed to have high recovery rates of the materials. In a crucial first step, the battery is discharged to allow safe tear-down. Next, the battery packs are placed on a disassembly line.

Opening the battery is done in a box filled with argon. The opening of the battery case is made around the edges of the battery – avoiding cutting the stacks of modules. Afterwards, the individual components of the battery are being separated which in most cases, is done manually. Once opened, the outer casing that consists of mostly metals and plastics is sent for conventional recycling.

Next, the modules move down a conveyor belt leading to a shredder. The shredder always remains closed, so the shredding process takes place in a vacuum or in a nitrogen atmosphere to prevent ignition. The rollers of the shredder grind the materials into tiny pieces and the mass is collected in plastic containers. This mass includes a mixture of valuable materials such as cobalt, nickel, and lithium, and passes through an array of magnets and a cyclone filter to extract these materials. All that remains at the end is a fine grey/black powder, called the black mass.

Chemical Li-ion battery recycling processes

The chemical battery recycling processes are mainly hydrometallurgical methods involving acid or base leaching, solvent extraction, filtration, chemical precipitation, bioprocesses and electrochemical processes, or a combination of the mentioned. The separation and extraction of metals based on reaction in aqueous medium, and particularly sulfuric acid (HM-SA) and citric acid

(HM-CA), is known as hydrometallurgical recycling. The function of the acid or base leaching is to put the metals in a solution. Once in a solution, the metals can be recovered by chemical precipitation through altering the pH of the solution or adding some reaction agent, or by electrolysis. The solution can also be separated by solvent extraction using an organic solvent, which binds to the metallic ion, separating the metal from the solution. The metal separated by solvent extraction can then be recovered by electrolysis or by chemical precipitation.

To put the chemical processes in perspective, we can analyse the example of implementing a chemical process onto already mechanically shredded Li-ion batteries such as the one described earlier. In fact, to ensure recycling different groups of materials, combining multiple methods of recycling almost always allows a higher rate of recyclability.

The process following the mechanical shredding involves a bespoke hydrometallurgy or wet chemistry process (an acid bath), used to process the black mass into battery-grade materials. During this process, different chemicals get added to the black mass which leach out different elements. These chemicals either make their way into the final products or are reused in the process – so this does not actually produce any waste water. This way, the black mass is separated into its components (lithium, cobalt, nickel). Once the materials have left the shredder, the liquid electrolyte is evaporated under a very low temperature so that there is no risk of ignition, and afterwards, it is condensed. The electrolyte can also be leached out in water or runs out as a clear fluid and flows into a large metal tank to find use as a base material in the chemical industry.

Comparison of the different battery recycling processes

The pyrometallurgical processes and the incineration of plastics have the largest impact on global warming potential over a 100-year time period (GWP 100) and electricity generation has the largest impact to human toxicity potential (HTP) and terrestrial ecotoxicity potential (TETP) [10]. The effects of electricity generation vary country to country and these effects could be reduced by implementing a larger proportion of energy generation from renewable sources. The hydrometallurgical processes indicate that the landfill of gypsum and residue has a large impact on GWP 100 and TETP.

As one of the main components of a battery, the recovering of the lithium cobalt oxide (LCO) is a process that is a big part of the recycling process. Roughly, just over a quarter of LCO can be recovered from a functional unit (FU) of one tone of spent Li-ion battery packs by the UHT, HM-SA, HM-CA, RR-N₂ and RR-Vac method (284.2, 275.5, 261.0, 273.0, 274.3 kg, respectively) [11].

If only LCO recovery was considered, the energy consumption of all recycling methods would be significantly lower than that for industry production of an equivalent weight of virgin LCO (38,367–41,777 MJ).

Assuming a recovery rate of 85% for copper (Cu) and aluminum (Al) the carbon intensiveness of the production of virgin copper and aluminum was also compared. Looking at Figure 3, each pair of bars is related to a recycling method, where the bottom bar represents the GHG emissions required for each recycling method and Cu and Al recovering processes for 1 FU, the top bar is the GHG emissions for an equivalent weight virgin LCO and primary Cu and Al production in industry, and the difference represents the energy saving. The numbers indicate the reduction rate of GHG emissions from 1 FU for each method.

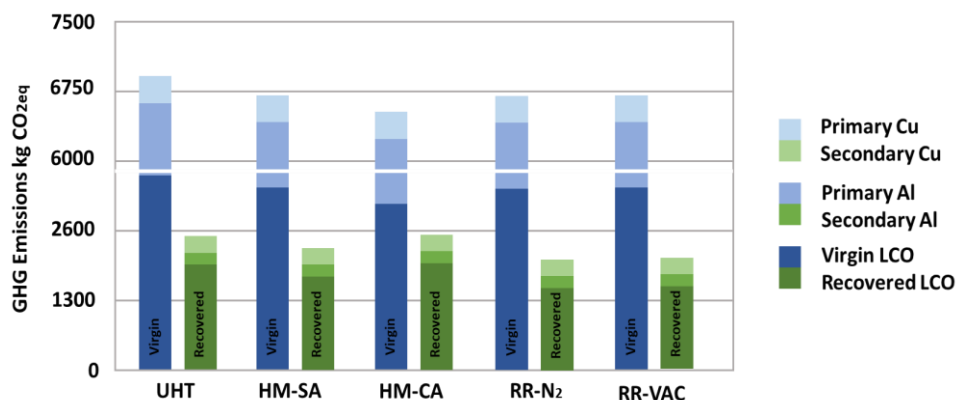


Fig. 3. Contribution analysis of GHG emissions for different recycling methods

4. RESULTS

The choice of the recycling process is very important with regard to the environmental impact and its carbon footprint. The general consensus about recycling is that it does require energy, but it usually takes a fraction of the energy as it does to mine and smelt raw ore. The process's lower energy consumption also means that recycling produces fewer greenhouse gas (GHG) emissions than mining operations. However, this is often not the case when it comes to the recycling of Li-ion EV batteries. In fact, by comparing the leading recycling process today, we found that some methods are extremely energy intensive and therefore could significantly contribute to global warming.

The environmental effects of recycling batteries can be reduced by choosing a location for the recycling plant in the nearby area of collecting the batteries, namely by saving CO₂ emission due to minimal transport fuel costs. Furthermore, most Li-ion batteries are manually disassembled at present (due to the diversity of cell types, cell chemistries, and pack structures produced by various manufacturers) and the mechanical separation and disassembly process is generally a low-carbon intensive process. This is because the process of mechanical separation

The RR methods manifest higher reduction in energy consumption and GHG emissions as compared with UHT, HM-SA, and HM-CA. This suggests that the RR method is more suitable for recycling spent Li-ion batteries than the UHT and hydrometallurgical methods (HM-CA and HM-SA). The method of HM-CA has been noted as one of the most energy intensive methods present in the industry, by having an energy consumption of 20,892 MJ per one FU during the whole process, while RR consumed the least amount of energy (4833 MJ per one FU). The RR method has the lowest total rate of GHG emissions (1525 kg CO₂-equivalent per one FU), while the HM-CA method exhibited the highest amount of GHGs (2351 kg CO₂-equivalent per one FU) among the analyzed methods [11].

ration is mainly associated with labor needs and not with fuel-powered machinery.

Furthermore, the metals used in the batteries are a non-renewable resource, meaning, there will not be any metals left to dig up once all the deposits are depleted by mining. Therefore, recycling helps in reducing the rate at which we need to mine metals to meet demand.

Taking all of the abovementioned data in consideration, not all of the currently available technologies are environmentally justified to recycle Li-ion batteries. Occasionally, the presented recycling processes lead to slightly less consumption of energy than primary production of virgin ores, but the carbon intensity of each individual process is still significantly high.

5. DISCUSSION

The sustainability of the recycling processes could not be taken for granted as it could vary depending on the materials and the impact of environmental policies and awareness is certainly not exhausted. A key concept that is directly connected to sustainability and should be used in the EV battery waste industry is the concept of circular

economy. The circular economy is a regenerative economic system within which production resources, waste, waste emission and energy outflow are significantly reduced by slowing down, rounding off and extending the energy and material cycles (life cycles) in production. The main challenge in this concept lies in the way we use the raw materials, i.e. what we will choose to use as a raw material to get a certain product and how we will close the life of that product. The circular economy, unlike the linear one is related to production, consumption, the way we design things, how to maintain them, how to use, fix and reuse them, and finally move on to recycling them. This means that we should not look at the situation as if it was waste management, but like it is more about resource management – how to manage resources and how to maintain their value through the recycling process.

When handling EV batteries, it is extremely important to follow the rules of the well-known waste management hierarchy, meaning that the used batteries are not waste but rather a resource that can be reused. Ideally, they would be collected, inspected, repaired, and returned in working condition for use in a different electronic device. If this cannot be achieved, the next step is repurposing. At the moment when the energy from these batteries is absolutely depleted and they are no longer efficient and/or safe to use, they should be treated as hazardous waste. Collection, transport as well as the recycling process itself and the further use of recovered resources should be sustainable and in line with standards that consider environmental protection. The recycling process should be non-invasive, safe and efficient. In this process, only a small part of the components should end up as waste, and most of the raw materials should be recovered. In regard to the carbon intensiveness of the different processes, it is important to consider which type of energy source is used to power the processes. Higher carbon intensiveness and GHG emissions are mostly related to coal-fired power plants when compared to renewable energy sources.

6. CONCLUSIONS

The aim of this paper was to investigate the different processes that are currently used for recycling Li-ion batteries disposed from EVs and to compare them with a focus on the associated environmental impact regarding their energy intensiveness and carbon emissions. The recycling processes were discussed with regard to their characteristics, advantages and disadvantages.

The findings indicate that the processes are not always easy to implement, nor are they always truly environmentally justified. Yet, keeping in mind that the ecological issues are the primary drivers to select between either virgin ores or recycled materials, decision makers need to be aware of the possible undesirable environmental effects if recycling processes are not introduced and encouraged in the automotive and battery production industry.

The main challenge that relates to the recycling of Li-ion EV batteries lies with the fact that neither of these procedures has been implemented on a wider scale, which in turn has a lot to do with EVs still lacking larger serial production numbers. Furthermore, some recycling procedures have a greater environmental impact over others, but it is important to combine several kinds of recycling processes to recover the main targeted metals (copper, aluminum, cobalt, manganese, and lithium) since just a single recycling process such as dismantling, thermal treatment, acid leaching, solvent extraction or chemical precipitation limits the percentage of recovery of the base materials.

While the recycling of Li-ion batteries helps prevent the shortage of critical minerals from a mass flow perspective, from an environmental perspective, the current available technologies lead to significant consumption of energy and higher air emissions than primary production. For the pyrometallurgical processes, the largest impacts are caused by plastics incineration for global warming potential, electricity generation for human toxicity potential and terrestrial ecotoxicity potential. For hydrometallurgical processes, the largest impacts are caused by landfill for global warming potential, terrestrial ecotoxicity potential, and electricity generation for human toxicity potential. To decrease the environmental impacts of recycling Li-ion batteries, processes that utilize low temperatures and are capable of recovering plastic should be used. Additionally, it is necessary to carry out a preliminary mechanical separation before valuable metals which make up Li-ion batteries can be recovered. Mechanical separation reduces the volume of waste and improves recovery efficiency of the target metals.

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