Analysis of energy demand for recycling electric vehicle batteries through a flexible hydrometallurgical process

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ABSTRACT

The decarbonization of the transport sector through electrification is limited if we consider the life cycle of lithium-ion batteries from manufacture to disposal**.** Such batteries, when reaching the end of their useful life, must be recycled because, by recovering these critical metals, it increases the circularity of the materials and the production of batteries again, in addition to reducing CO_2 emissions and the impacts associated with mining activities. Currently, recycling processes use pyrometallurgy techniques, employing high temperature, high energy consumption and higher CO_2 emissions. Hydrometallurgy has stood out as a promising process for recycling batteries, with high efficiency, use of low temperatures, lower emissions, and lower energy consumption. This work aims to analyze the energy demand and the yield of recycled materials through the developed hydrometallurgical process, since it aims to minimize the emission of waste in nature with a flexible process for recycling lithium-ion batteries. Different types of batteries were processed, physical separation and leaching recovered more than 90% of the minerals of interest with a reduction in energy consumption of up to 65% compared to pyrometallurgical techniques.

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INTRODUCTION

The concern about climate change and its mitigation has increased research effort to avoid the imminent consequences of greenhouse gases (GHG) emission. The United Nations Framework Convention on Climate Change (UNFCCC) is implementing agreements, since COP92 in Rio, proposing limitations in emissions; Kyoto Protocol, inducing pollution reduction by carbon pricing, emission credit trades and stimulating investments in developing countries; Paris Agreement, limiting the temperature increase implementing a five year cycle action plan by countries and inducing forest conservation; and proposing sustainable development goals (SDGs) with 17 targets to sustainable development. [1-3]

The electrification is one of the movements to reduce CO_2 emission through the World. However, the energy matrix in all countries should move from fossil fuels to renewable energy sources, increasing the contribution to the decarbonization process. [4] Lockdown proved the impact of anthropogenic activities, with the decline of 6% of fuel emissions, which was almost five times larger than 2009 financial crisis reduction. [5-7]

Zhang (2020) discusses electric vehicles (EV) contribution as a clean technology capable to decarbonize electricity in long term, due to transport sector impact in emissions and energy consumption. [8] Low carbon technologies in transportation involves the substitution of petroleum fuels by alternative fuels, implementation of automated vehicles, selection of freight transportation in rails or low carbon vehicles. [9]

Lithium-ion batteries (LIB) can influence emissions in all steps of life cycle, due to energy intensive manufacturing activities by mining sector, efficiency of technologies by cathode composition and the end-of-use approach. According to Guilherme et al. (2023), an electric vehicle demands more than a double or even triple carbon footprint than an equivalent combustion vehicle due to battery size. The mining activities linked to battery production consumes also high level of water and move several tons of earth to enable the production of one battery. In this way, recycling these batteries can reduce primary raw materials exploration, and mitigate impacts as toxicological problems and environmental pollution of inadequate disposal. [10-11]

Moisé and Rubinová (2023) foresee that emission related with EV manufacturing could decrease by 14% to 23% by 2040, if LIBs are recycled to re-input raw materials. LIB recycling methods involves pyrometallurgical or hydrometallurgical processes recovering 70% of cathode value. Pyrometallurgical process are energy intensive, generates toxic gases, has limited materials recovery with hydrometallurgical post process to recover more metals; advantages consist in the direct processing of battery modules without prior physical steps and application in sorted feedstock LIB waste. Hydrometallurgy tends to recover more metals and materials with required purity to be secondary raw material for LIB production. [12-14]

Pyrometallurgical recycling can reduce up to 34% of energy consumption in cell production, while hydrometallurgy can reduce up to almost 70%. [15] Consecutively, GHG emissions are reduced more than 60% by existing pyrometallurgical routes, and as expected hydrometallurgy can reach lower carbon footprint due to its lower energy requirements. [16] In this way, this work aims the development of a flexible recycling process by hydrometallurgical route with lower energy demand. Different types of batteries were evaluated, as well as different cathode materials to design the route.

METHODS AND MATERIALS

The process was carried out using different types of Li-ion batteries: cylindrical, pouch and prismatic. Figure 1 presents the cells used in this study. The packs and modules were disassembled manually to release the cells. Further, the batteries were discharged using electrical resistance, where they were connected in series and then to a wire or resistance. After the voltage achieved zero (0V), the batteries were comminuted, and the cathode was separated by physical treatment.

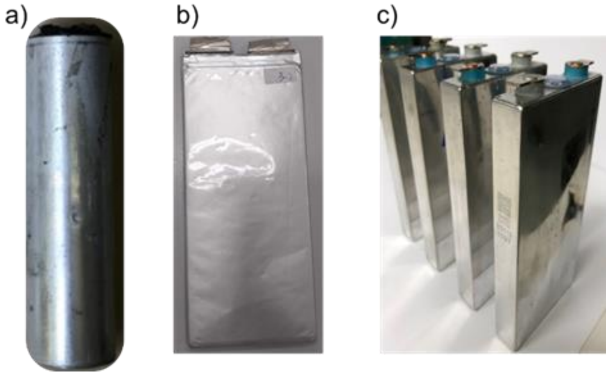

Figure 1. Photo of the types of cells (a) cylindrical (b) pouch (c) prismatic used in electric vehicles. [17]

The active material was leached using H_2SO_4 varying the parameters to achieve efficiencies up to 95% for the most valuable metals. Then, the leach liquor was treated by separation and purification steps to obtain high purity products: using a precipitant agent for Mn recovery, precipitation for Al removal, solvent extraction for Co separation and further precipitation, and Ni and Li precipitation.

The experiments were carried out in laboratory scale. The experimental conditions were set considering the purity up to 90% for the products. The recycling process was designed to be flexible, simultaneously processing all types of Li-ion batteries. In leaching, the experiments were carried out in 500mL glass reactor under heating and magnetic stirring with 10-100g of material after physical treatment. Separation and purification steps were carried out in glass reactor under temperature and pH control. Precipitation experiments were performed using precipitant agents, and solvent extraction for Co separation was carried out with a selective organic compound.

The calculation of the energy consumption of the Tupy hydrometallurgical process for recycling batteries includes the consumption of electrical energy in all stages of the process, on a laboratory scale. This consumption is estimated based on the power levels of the equipment and the time of use, as well as the impact of utilization factors related to scale-up.

RESULTS AND DISCUSSION

Two batteries were dismantled to release cylindrical and pouch cells and prismatic cells were received separate from module. The module of cylindrical cells was divided in cells (56% wt.), aluminum structure (38% wt.), and electronic compounds (6% wt.), with total weight of 46 kg. Pouch module was composed by cells (46% wt.), plastic structure (23% wt.), internal aluminum structure (12% wt.), and electronic compounds (19% wt.), with 9 kg of total weight. In both modules, structures represent similar weight proportion and are composed mainly by aluminum structure.

To characterize the cells obtained, after discharge, disassembly was performed separating cathode, anode, polymeric separator, case and electrolyte. Figure 2 shows weight composition of the three lithium-ion battery cells.

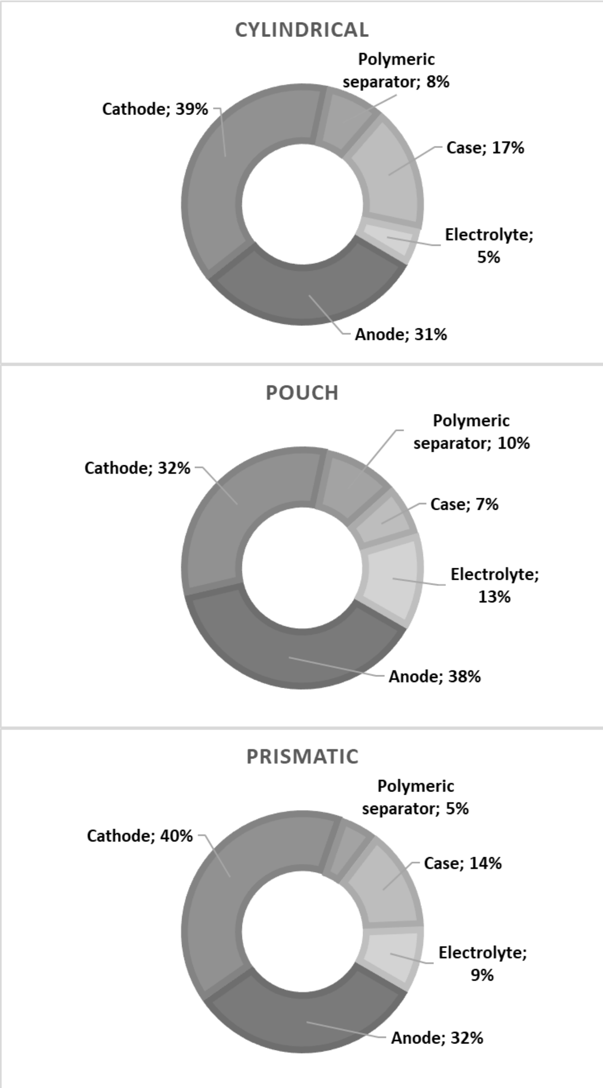

Figure 2. Weight composition of cylindrical, pouch and prismatic Li-ion battery cells.

It was observed that the anode and cathode are more than 2/3 of cell weight. While anode is composed by graphite material with low lithium concentrations, cathode contains valuable metals, as Ni, Co, Li, Mn, Fe and Al, depending on the chemistry of active material (NCA – $LiNiCoAlO_2$; NMC – $LiNiMnCoO_2$; LCO – $LiCoO_2$; LFP – $LiFePO_4$). [18] Separators are polymeric made and avoid short-circuit between cathode and anode. The case from pouch cells was lighter than others due to its unique structure mounted with aluminum foils. Electrolytes are a mix of organic compounds and Li salts, which was removed by evaporation at room temperature after disassembling.

Figure 3 presents LAREX-Tupy hydrometallurgical process flowchart. [17] Battery cells are firstly comminuted in knife mill or shredder, and components are separated by physical methods. The active material contained in cathode was leached reaching up to 99% of Li, Ni, Co, Mn, while 60% of Al was leached. Inert graphite and unreacted aluminum were retained in filter after solid/liquid separation. Pregnant leaching solution (PLS) was treated to separate and recover metals as oxides, carbonates and hydroxides by precipitation of Mn, Al, Li and Ni, while Co was separated by solvent extraction. Purity of products obtained was superior to 90% and recoveries of proposed process is at least 90% of cathode active material.

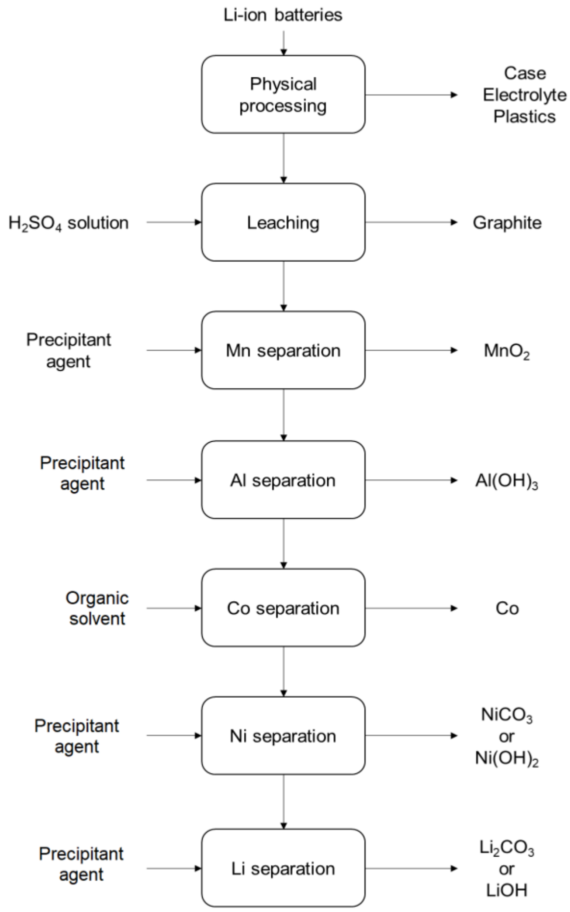

Figure 3. LAREX-Tupy hydrometallurgical process flowchart for Li-ion battery recycling.

The first metal removed from leach solution was Mn, in acid pH conditions and using a precipitant agent, it is possible to remove the Mn with precipitation efficiency of 99% with a purity of 96% (Co was in part co-precipitated). Al was removed from PLS by pH adjustment adding sodium carbonate or hydroxide, reaching efficiency of

100%, while Li, Ni and Co losses by co-precipitation were lower than 2%.

Subsequently solvent extraction process separated Co from PLS, while Ni and Li remained in solution. Co was stripped from organic phase by an acid solution and then possibly recovered by oxalate and carbonate precipitation, or in metallic form by electrowinning. Li and Ni remaining in solvent extraction aqueous solution were precipitated as carbonate or hydroxide.

Differently of pyrometallurgical process that reduce Co, Ni, Cu and Fe in alloys separating them from Li, Al and Mn in slag phase, hydrometallurgical process can recover metals individually. [13] Besides that, capital investment and energy consumption in pyrometallurgical process is higher intensive. [19-20] Figure 4 presents direct energy consumption comparing literature data of pyrometallurgical routes and LAREX-Tupy hydrometallurgical process in different technology readiness levels (TRL).

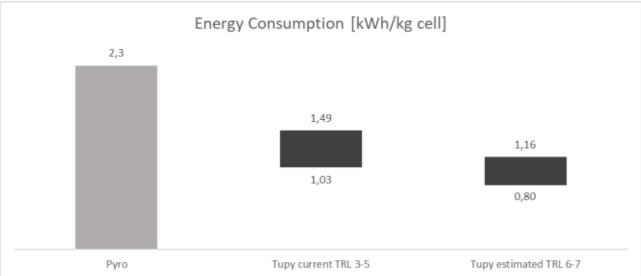

Figure 4. Energy consumption of different recycling process for LIBs from literature (Adapted from Dai et al., 2019; Xu et al., 2021) [21-22], and comparison with LAREX-Tupy process.

It was observed that the pyrometallurgical process has a higher energy consumption than the hydrometallurgical one. Besides that, LAREX-Tupy hydrometallurgical technology developing process approximates of referenced hydrometallurgical consumption and can decrease this value with increasing TRL. Describing consumption by recycling step processes, physical processing is responsible for a larger share of energy consumption, due to comminution energy intensive step. Figure 5 shows energy consumption by steps of recycling process.

In addition to an energy consumption, consecutive carbon footprint is obtained presenting the hydrometallurgical process as a competitive technology to reduce emissions and contribute to a less environmental impact. Figure 6 presents the conversion of energy consumption in equivalent CO_2 emission of above processes discussion, including comparison of nations with different power generation matrix (renewable and non-renewable).

In a circular economy proposal of recovering metals from LIBs, reinserting them to battery market can potentialize GHG emission reduction of electrification, within a hydrometallurgical route, as a greener solution. It was also observed in Figure 6, the lower impact in emission of Brazilian scenario, due to its hydroelectric plants share in electricity generation. Therefore, LAREX-Tupy process is a flexible route with high extraction efficiency, which can meet sustainable development goals and recycle different EV batteries.

Besides emission reduction, a comparison of material recovery from pyrometallurgical and hydrometallurgical processing of a NMC cell also demonstrates the impact of a hydro process for a circular economy. From Figure 6 for Brazilian power mix, pyrometallurgy presents almost 2 times higher CO_2 emission than hydrometallurgy for the same volume of processed cells. However, taking in consideration that pyrometallurgy recovers around 60% of processed materials and hydrometallurgy 90%, the CO_2 emission for the same level of recovered materials presents CO_2 emissions of 3 times higher for pyrometallurgy. [24-26]

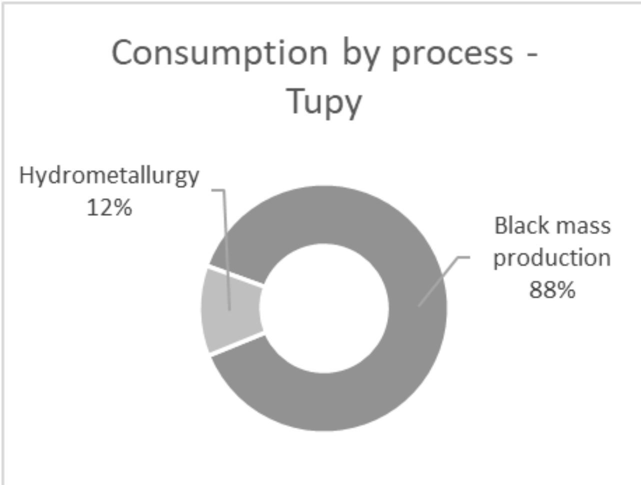

Figure 5. Energy consumption in recycling steps of LAREX-Tupy process.

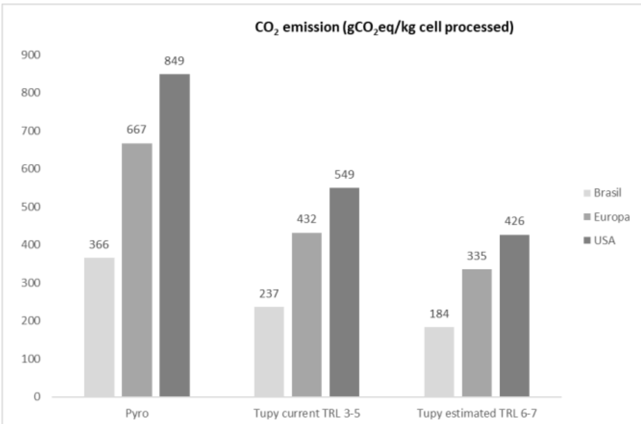

Figure 6. Impacts in GHG emission of pyrometallurgical process compared with Tupy process in Brazil, Europe and USA. [23]

Combining the results from recovery, the carbon impact of pyrometallurgy is even more than hydrometallurgy, without considering lower purity of products from pyrometallurgical processes.

CONCLUSION

LAREX-Tupy process is a recycling hydrometallurgical route with flexibility to treat all LIB configurations (cylindrical, pouch and prismatic), which has lower energy consumption if compared to pyrometallurgical processing, and lower GHG emission, enabling the recovery of battery metals (Ni, Co, Li, Mn, Al) with high purity (>90%) and high recovery efficiencies (>90%). Implementation of the process at a higher TRL, in a Brazilian scenario of electrification, allows meeting the objectives of sustainable development, reducing CO_2 emissions, enabling the reintroduction of recycled minerals in the battery chain, what increases the circularity of strategic minerals. In addition, it may favor the mitigation of environmental issues related to the manufacture and life cycle of batteries.

Evaluations of energy demand show reductions of 36 to 44% in current TRL level for hydrometallurgy compared to pyrometallurgy. It can be even improved up to 50-66% in the next TRL increase loop.

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