

## Article

# Characterization of Lithium-Ion Batteries from Recycling Perspective towards Circular Economy

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**Abstract:** Recycling processes of lithium-ion batteries used in electric and hybrid vehicles are widely studied today. To perform such recycling routes, it is necessary to know the composition of these batteries and their components. In this work, three pouch and three cylindrical LIBs were discharged, dismantled, and characterized, having their compositions known and quantified. The dismantling was performed using scissors, pliers, and a precision cutter equipment. The organic liquid electrolyte was quantified via mass loss after it evaporated at 60 °C for 24 h. The separators were analyzed using Fourier-transform infrared spectroscopy (FTIR), and the cathode and anode active materials were analyzed using a scanning electronic microscope coupled to an energy-dispersive spectroscope (SEM-EDS), X-ray diffraction (XDR), and energy-dispersive X-ray fluorescence spectrometry (EDXRF). All LIBs were identified by type (NCA, NMC 442, NMC 811, LCO, and two LFP batteries), and a preliminary economic evaluation was conducted to understand their potential economic value (in USD/t). Both results (characterization and preliminary economic evaluation) were considered to discuss the perspective of recycling towards a circular economy for end-of-life LIBs.

**Keywords:** electric vehicles; hydrometallurgy; economic evaluation; end-of-life battery; urban mining



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## 1. Introduction

Lithium-ion batteries (LIBs) have greater energy storage in smaller volumes due to their high energy density. Consequently, electronic equipment and electric cars have gained better operational conditions [1,2]. There are different types of LIBs, according to their geometry, such as cylindrical, in which the electrodes are wound, and prismatic and pouch, which have flat electrodes [3]. A typical pouch-type battery, for instance, is often used in portable electronic devices, such as cellphones, ultrathin computers, and tablets [4]. Pouch batteries have an important safety factor, since, in situations of overheating, the internal pressure does not generate harmful fragments during the rupture. The flexibility allows expansion of the battery avoiding rupture, being the safest type to be used in electric cars [5]. Prismatic cells are big cells and are subjected to far higher pressure than pouch or cylindrical cells, making them hazardous to open if the contents have degassed. To remove the contents of a prismatic cell, specific instruments are required [3,6].

Cylindrical batteries are tightly coiled and frequently joined into modules using epoxy glue (which is difficult to remove or recycle). Discharging this type of cell may be problematic due to the fuses at either end, which may be blown. In addition, the cell shape may be difficult to deconstruct for direct recycling, posing additional obstacles to separate the electrodes for recycling procedures [3,7].

The CO<sub>2</sub> net-zero initiatives motivated several countries to adopt sustainability policies, such as the incentive for purchasing electric vehicles. For instance, the USA has 438 government incentives to achieve this goal, such as tax incentives and subsidies for the

infrastructure needed to recharge electric cars [8]. Consequently, the increase in the use of electric vehicles will generate a proportional increase in battery production, raw materials extraction, and LIBs waste generation.

As a consequence, the number of vehicles worldwide has been increasing [9]. In 2021, around 71% of global lithium production was used in the lithium-ion battery market. This percentage rose to 80% in 2022 [10]. On the other hand, the lack of raw material to supply future demand, also known as critical raw materials, has caused international concerns [11,12]. The European Union added lithium to the list of critical raw materials in 2023 due to its risk of supply interruption. Co was already on the list, while Ni and Mn have been added as well, all of them being widely used for the production of LIBs [11].

Consequently, several countries, such as the ones in the European Union, have promoted an action plan to create a circular economy and increase the use of secondary raw materials. Regarding this, the recycling of spent batteries of electric vehicles is important for circular economy promotion towards supply–demand of critical metals such as lithium, cobalt, nickel, and manganese [11]. In order to establish a recycling process, it is important to understand what components a LIB presents.

One cell of the LIB has components such as electrodes (anode and cathode), and two current collector metallic foils that are attached by a binder to the electrodes. One current collector is made of metallic Al, which is attached to the cathode, and the other one is made of metallic Cu and is attached to the anode [13]. The binder is often made by polyvinylidene fluoride (PVDF). In addition, a cell also contains lithium hexafluorophosphate ( $\text{LiPF}_6$ ) dissolved in an organic solvent, such as the electrolyte [7,13]. The cathode is responsible for receiving the electrons and cationic ions during operation as a battery, whereas the anode is responsible for donating electrons and cationic ions to the cathode. A singular cell presents a polymeric separator, and an external structure that is usually made by steel, aluminum alloy or plastic [13–15].

However, despite the fact that the literature presents knowledge about the main characteristics of LIBs, focusing mainly on the cathode and its treatment (the recovery of the metals in the cathode's active material), little is detailed about the characterization of those batteries prior to recycling [6,7,16,17]. This step is responsible for qualifying and to quantifying all the components of those batteries, and it fundamentally helps the next steps during the recycling process. Furthermore, the economic evaluations of each component of the battery are not thoroughly considered, and an economic study of the potential use of these materials is needed.

This present work aimed to characterize pouch and cylindrical-type LIBs and compare them to other configurations, providing detailed information. The LIBs were discharged by wires and opened for gravimetric analysis. Chemical analysis was carried out for cathode and anode to determine the critical metals content. After that, a preliminary economic evaluation was performed to observe the market trend among the types of the batteries studied. The results were compared with the literature. A preliminary economic and recycling techniques analysis were carried out in light of circular economy processes.

## 2. Materials and Methods

This work studied the characterization of LIBs and a preliminary technical and economic analysis of recycling to obtain critical raw materials. Figure 1 shows the flowsheet of the methodology used in this work, which was proposed by the literature [7]. In this study, three pouch cell LIBs from different manufacturers were studied, as shown in Figures 2 and 3. The batteries were identified as P1, P2, and P3. Three cylindrical batteries were also characterized—named as C1, C2, and C3—and further compared with pouch batteries. These commercial samples were supplied by a research center in battery development after use, without chemical or structural damage. In this study, the cells were first discharged and then manually dismantled to separate all fractions of the cells. The organic electrolyte (solvent) was evaporated at room temperature as previously reported and

determined [7,17–19], and the fractions were characterized by chemical and physical techniques. Then, a preliminary economic analysis was carried out, as well as a literature analysis.

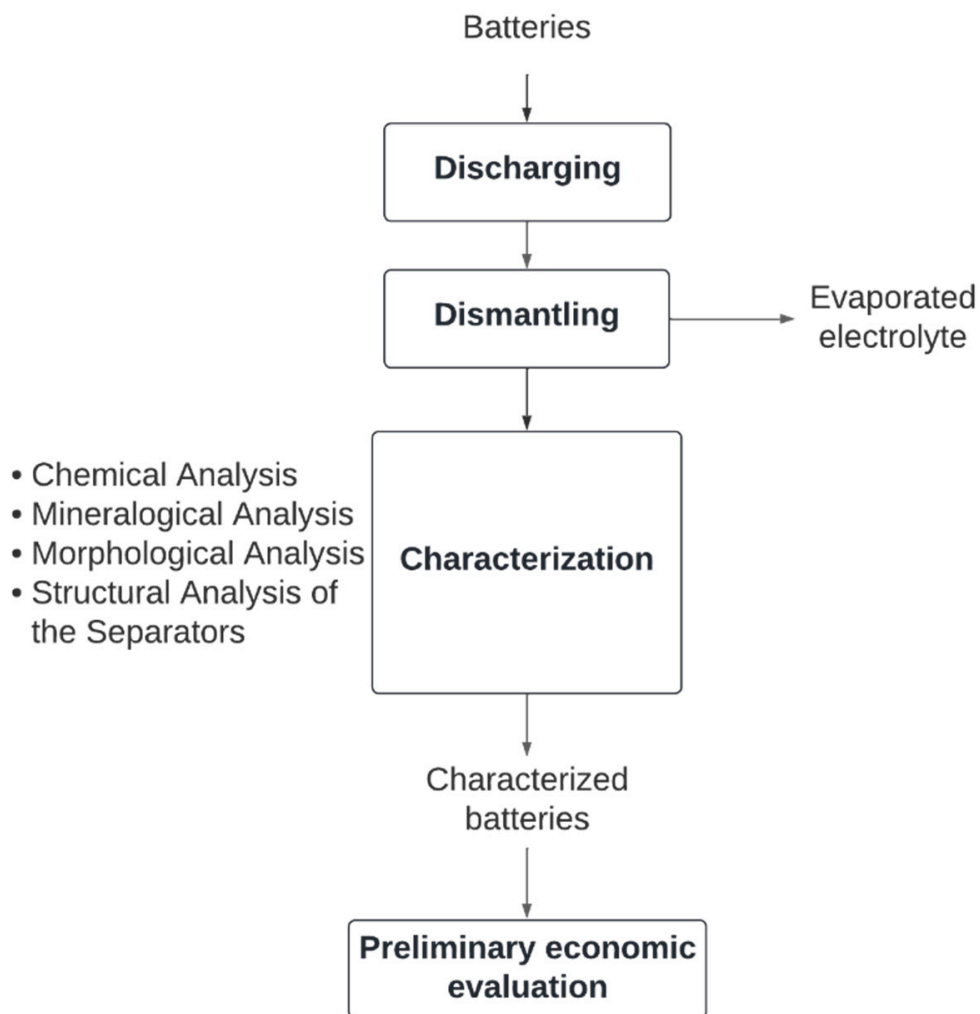
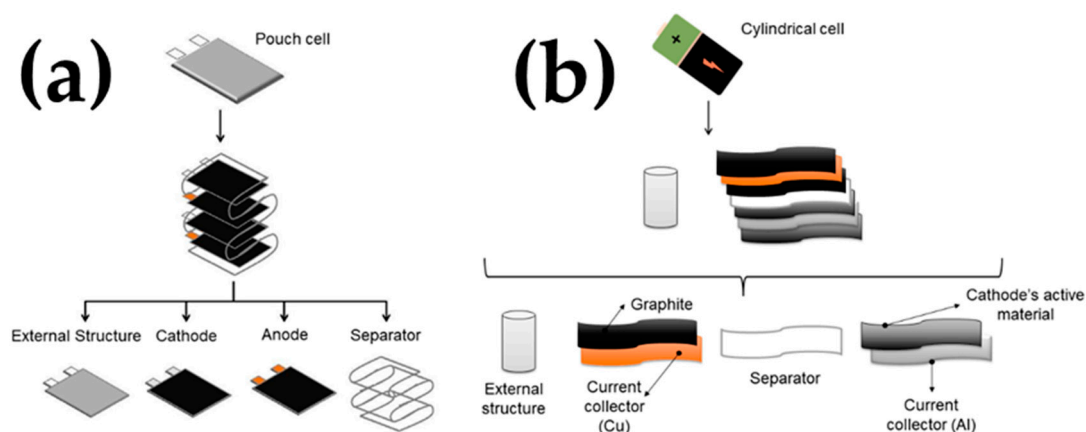


Figure 1. Flowsheet of the process used in this work.



Figure 2. The three pouch type LIBs studied in this work (P1, P2, and P3), and the three cylindrical batteries found in the literature (C1, C2, and C3).

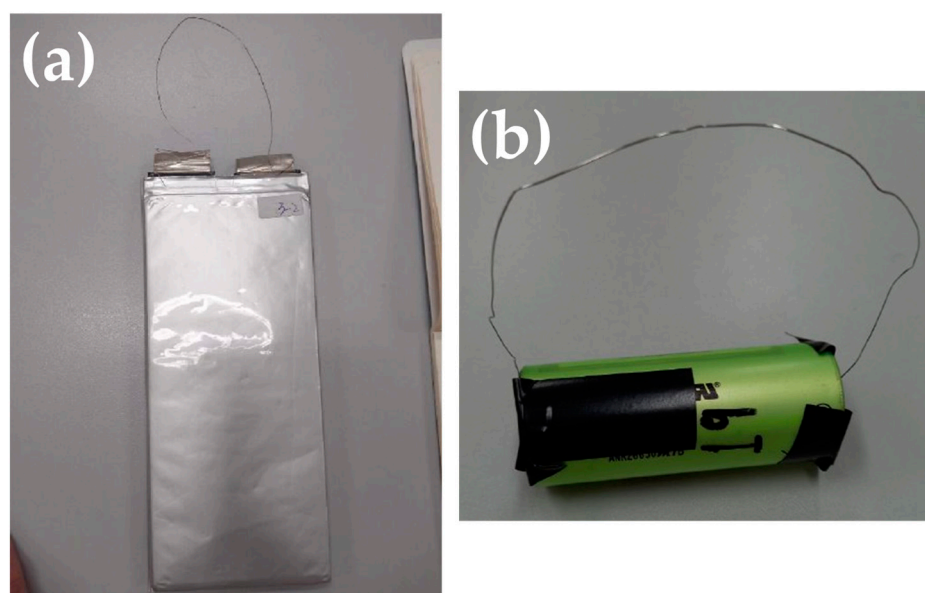


**Figure 3.** Scheme of the separated components from a (a) pouch battery cell and from a (b) cylindrical battery cell.

### 2.1. Discharging and Dismantling the Batteries

The batteries were discharged using a Ni-Cr wire with  $4\ \Omega$  resistance connected across their positive and negative terminals, using the methodology we previously proposed [7,20–22]. This procedure was adopted to avoid losses of material and emission of harmful gases by corrosion in the discharging process using alkali (as NaOH), salts solutions (as NaCl, NaSO<sub>4</sub>, FeSO<sub>4</sub>, and ZnSO<sub>4</sub>), or thermal reactions [23–25].

After completely discharged (from 3.7–4.0 V to 0 V after around 24 h), the external structures of the batteries were removed manually using scissors (pouch LIBs) or using the Buehler IsoMet 1000 precision cutter (Buehler, Lake Bluff, IL, USA) at a speed of 875 rpm (cylindrical LIBs). The batteries were then dismantled and separated into an external structure, cathode (for this study, the cathode will be considered as active material + Al foil), anode (for this study, the anode will be considered as active material + Cu foil), and separator. The cathode, anode, and separator were named in this study by adding a C, A, or S, respectively, in front of the names of the batteries. Figure 4 shows a schematic of the components, separated from cell into fractions.



**Figure 4.** Battery discharge by connecting the electrodes with a resistance wire: (a) a pouch cell and (b) a cylindrical cell.

## 2.2. Sampling and Preparation

After manual dismantling, all components were separated and dried at 60 °C for 24 h. Active material samples were obtained using a spatula to scrape them from the current collector. A mass equivalent of 1 g of this material was then digested in a 250 mL beaker using 100 mL of aqua regia (HNO<sub>3</sub>:HCl = 1:3, *v/v*) for 24 h at 60 °C, under constant stirring. After acid digestion, the mixture was filtered, and the liquor and the residue generated were analyzed. The Al and Cu foils were quantified after acid treatment (4% HNO<sub>3</sub>) under ultrasound for 0.25 h (15 min) and washed with ethanol 99.5% under ultrasound for 0.17 h (10 min) at room temperature.

## 2.3. Analytical Procedures

The solid samples were dried at 60 °C for 24 h before the analysis. The mineralogical assessment of the active materials from the cathode and the anode was performed in X-ray diffraction equipment with incident CuK $\alpha$  radiation and equipped with a graphite monochromator and nickel filter at a 2 $\theta$  angular range from 15° to 85° (XRD Rigaku Miniflex300 (Rigaku, Mumbai, India)). The morphology and the semiquantitative elementary analysis of the samples was carried out in scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDS PhenomWorld ProX (Thermo Fisher Scientific, Waltham, MA, USA)). The concentration of metals was analyzed by an energy-dispersive X-ray fluorescence spectrometer (EDXRF Epsilon 3-XL (Malvern Panalytical, Malvern, Worcestershire, UK)) and by an inductively coupled plasma optical emission spectrometer (ICP-OES 700 Series (Agilent Technologies, Santa Clara, CA, USA)). Lithium quantities were analyzed by flame photometer (Model 910 (Analyser, Sao Paulo, Sao Paulo, Brazil)). Separator analyses were carried out by Fourier-transform infrared spectroscopy (FTIR Tensor 27 (Bruker, Billerica, MA, USA)) from 4000 cm<sup>-1</sup> to 400 cm<sup>-1</sup>.

## 2.4. Preliminary Economic Analysis

After the characterization analysis, a preliminary economic evaluation was carried out in order to know which of the pouch batteries had the highest market trend, according to their composition. Hence, the battery with the highest value of USD/t of battery was considered as the one with the highest trend in the market. The prices of each element or product, in USD/t, for each battery were obtained from the London Metal Exchange (LME), the SMM, and the Trading Economics websites. The values of the batteries were obtained using the prices from those websites (December/2023) and further compared with the literature.

A literature analysis was carried out to evaluate recycling approaches for these batteries in light of the circular economy; that is, techniques that can make possible the recovery of most of the materials, including plastics and organic electrolytes. Efficiencies and cost-effective analyses were performed using these two types of batteries and experimental examples from the literature.

# 3. Results and Discussions

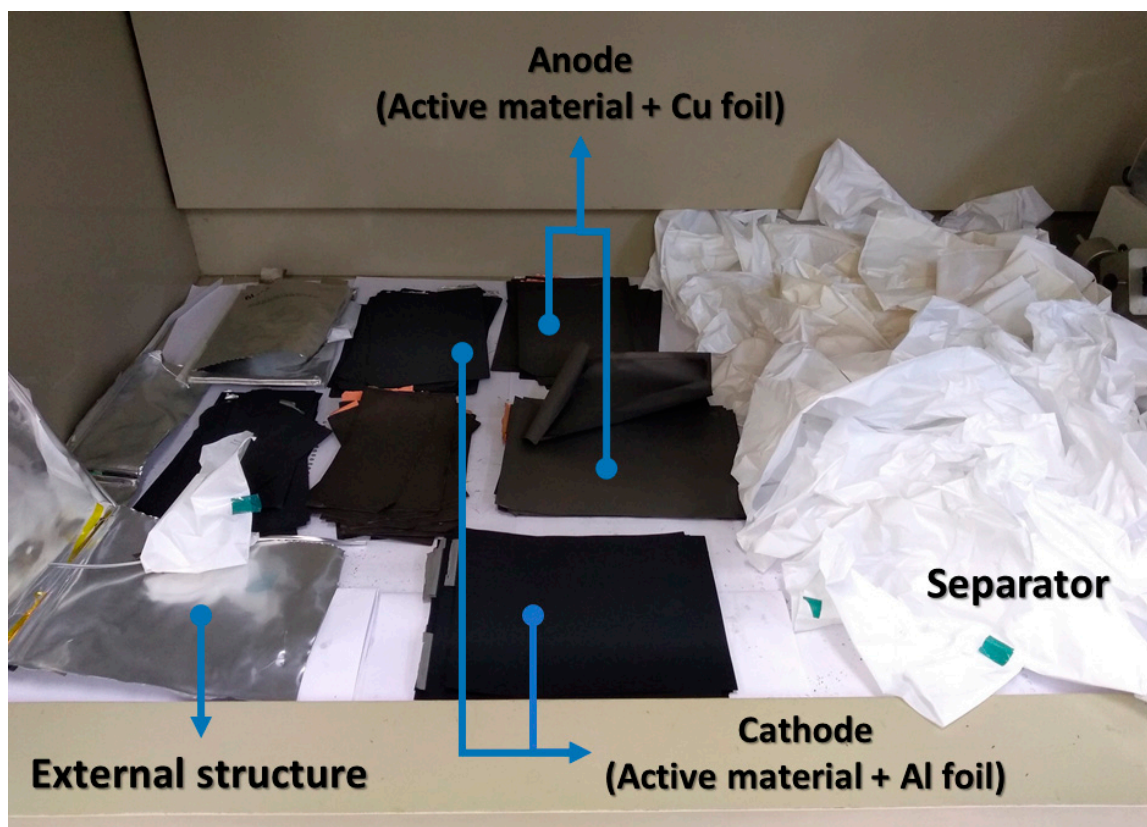
## 3.1. Discharging and Dismantling the Batteries

Spent batteries can have residual energy stored. If the batteries are incorrectly handled, this energy can be released quickly causing explosions due to the presence of energy and organic compounds (electrolytes) [26]. Batteries can be discharged by promoting a controlled short circuit using saline solutions or resistors. In the case of saline solutions such as NaCl, it is a low-energy, high-discharge efficiency method. However, saline impurities and undesired ions can be generated during this discharging process, which can contaminate the inner materials and complicate the subsequent characterization stage [23–25].

In addition, this discharging method can cause galvanic corrosions on the surface of the battery, allowing for organic electrolyte loss from the battery into the saline solution, which can also generate toxic gases such as HF [23,24]. Therefore, in the present study, the



discharging method using resistors was chosen to avoid those problems. Figure 5 shows one pouch battery with its electrode's extremities connected with a Ni-Cr wire that operates as a resistor with  $4\ \Omega$  resistance. Therefore, the circuit was closed, and the battery was discharged. This same type of resistor was used to discharge the other two pouch batteries. The voltage reached after 12 h of discharging was 0 V.

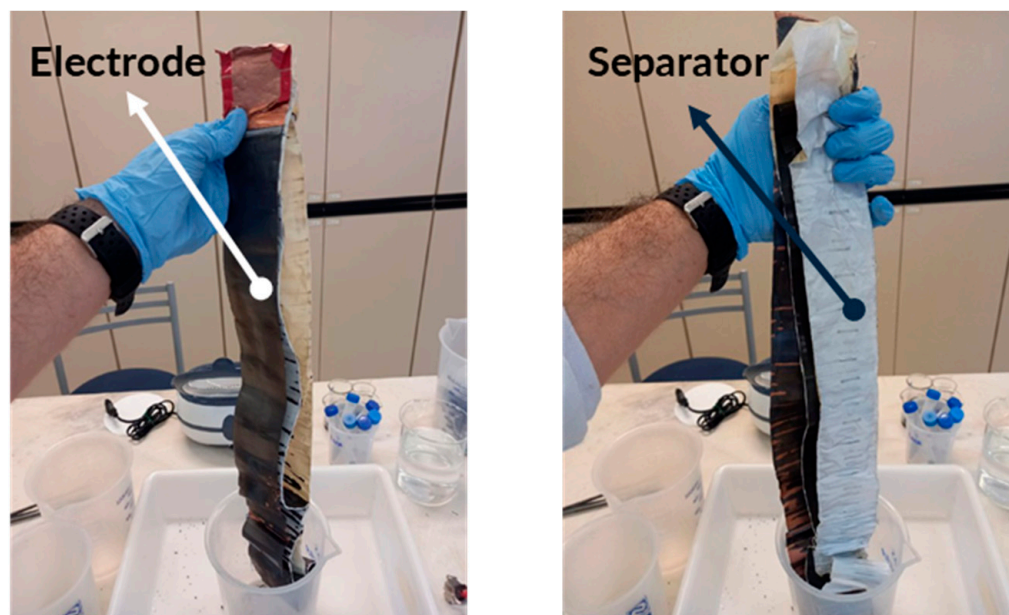


**Figure 5.** Dismantled pouch batteries and their components separated manually.

The pouch-type batteries used in the present study have planar electrodes. Each battery consists of an external structure that holds together the cathode, the anode, the electrolyte, and the separator (Figure 5). Their structures and internal configurations are the same: both the anode and the cathode have several electrode sheets, and a single separator sheet separates the cathode from the anode in each pouch battery (Figure 3).

The internal arrangement between the electrodes and the separator in a cylindrical LIB, however, is different. Each electrode has only one long sheet, and the separator (also a single long sheet) remains between the anode and cathode sheets. Figure 6 shows the internal arrangement of cylindrical LIB, as found in the literature [7]. The percentages of each component of the batteries were determined and compared with the literature (Table 1). The weight of the electrolyte was calculated after removing all the organic solvents by the drying process.

The results obtained in this study were compared with the literature data. Cylindrical batteries exhibited similar values to pouch batteries, such as the percentage values for the separators, electrolytes, the cathodes, and anodes (Table 1). The main difference can be observed in the values for the external structure. Table 2 shows the masses for the batteries studied in this work and the comparison to the literature. Despite the fact that the cylindrical batteries present lighter external structures compared to the pouch batteries, their total mass is significantly smaller than the total mass of the pouch batteries. This difference can explain the considerable variation in the weight percentages of external structures across different works.



**Figure 6.** Internal arrangement of an open cylindrical LIB (adapted from [7]).

**Table 1.** Percentage by weight of pouch battery components evaluated in the present work and comparison with those reported in the literature [7,13,27].

	Percentages (wt%)							
	Present Study (Pouch Batteries)			He et al. (2015) [27]	Velázquez-Martínez et al. (2019) [13]	Guimarães et al. (2023) [7]		
	P1	P2	P3	LCO		C1	C2	C3
Cathode	44.2	45.9	46.5	32.4	35.5	42.4	41.5	39.9
Anode	36.7	35.3	35.3	22.6	25.5	26.9	26.5	30.1
Separator	3.9	4.2	4.5	3.6	4.0	3.0	1.1	5.0
External structure	4.4	5.0	2.7	30.3	25.0	17.5	15.1	13.3
Electrolyte	10.8	9.7	11.0	11.2	10.0	10.2	15.8	11.7

**Table 2.** Mass of each battery component (g).

Components	Mass (g)					
	P1	P2	P3	C1	C2	C3
Cathode	189.0	222.4	142.6	19.9	16.7	29.5
Anode	157.2	171.1	108.3	12.6	10.6	22.2
Separator	16.5	20.3	13.8	1.4	1.0	3.7
External structure	18.8	24.0	8.4	8.2	6.0	9.8
Electrolyte	46.4	46.8	33.8	4.8	6.2	8.7
Battery cell (total)	427.9	484.6	306.9	46.9	40.5	73.9

The literature did not show whether the battery they studied was cylindrical, pouch, or prismatic [23–25]. Considering the most notable difference between the percentage values for their external structure and the present work's values for that same component, it is assumed that they have obtained those values from a cylindrical or a prismatic LIB. The cathode and anode weight percentages were close, about 45 wt% and 35 wt%, respectively. Even though the amounts of cathode and anode might be considered similar, the weight percentages for the electrodes reported in the literature are consequently lower than those determined in the present study, since the external structures of their batteries appear to be heavier.

The external structure of the pouch battery is a flexible bag of Al foil and plastic film. The plastic film is composed of thin layers of two polymers (3–14 mils) bonded by a binder. Polymers are a polyolefin, generally low-density polyethylene, and a polar polymer, such as polyethylene vinyl alcohol copolymer (EVOH), polyamide, or polyurethane. In the middle of the polymeric layer, a thin aluminum film is added. The external structure is closed by a heat-sealing process, forming a fusion bond between the polyolefin layers. These external structures protect the battery from moisture and have the advantage of being cheaper, lighter, and more flexible compared to cylindrical and prismatic batteries. For the cylindrical batteries studied, the external structures are all made of stainless steel.

### 3.2. Characterization of the Separator

The separator is a polymeric sheet that is located between the electrodes, allowing ionic conductivity and avoiding short circuits. The most widely used types of separators in LIBs are polyethylene (PE) and polypropylene (PP), which can also be found as a combination of PE and PP (bilayer PE/PP or multilayer PP/PE/PP), which is called polymer blends [28]. The characterization of the separators was carried out by FTIR (Figure 7). The separators, named SP1, SP2, and SP3, correspond to the batteries P1, P2, and P3, respectively. Similarly, SC1, SC2, and SC3 correspond to the separators of batteries C1, C2, and C3, respectively.

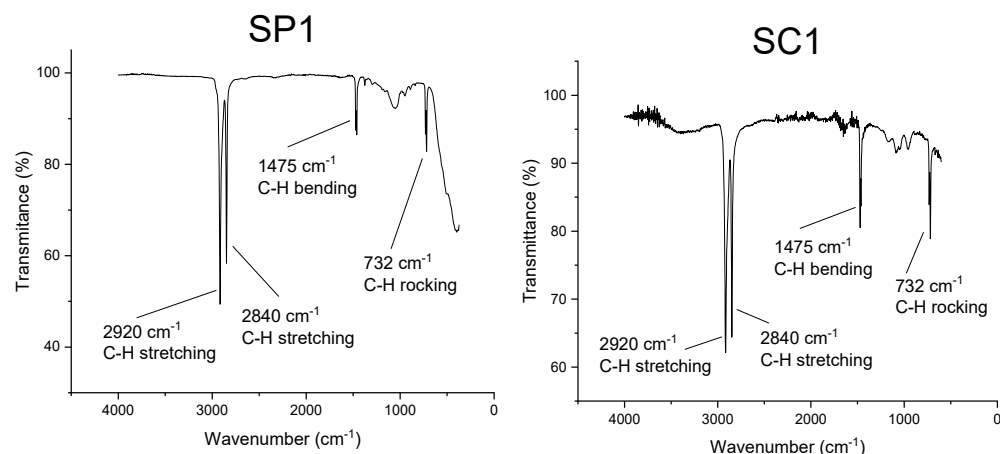
The FTIR analyses for all the batteries showed the main bands at 2840 and 2920  $\text{cm}^{-1}$  of C-H stretching, 1475  $\text{cm}^{-1}$  of C-H bending, and 732  $\text{cm}^{-1}$  of C-H rocking. For all the batteries studied, both pouch and cylindrical, the FTIR spectra of the separators in the batteries were characteristic of polyethylene (PE).

In addition, these polymers are produced from petroleum extraction, and losses by thermal reaction (pyrometallurgy) or landfilling result in  $\text{CO}_2$  emissions. Recycling them promotes the circularity of the carbon material and reduces the need for new separator production which, consequently, leads to a decline in  $\text{CO}_2$  emissions and mitigates climate change [29,30]. Furthermore, depending on the conditions the plastic is exposed to during the recycling process (such as heat), the plastic can be degraded and lose its properties. A polymer with a degraded chain makes mechanical recycling unfeasible, requiring chemical recycling [29]. Table 3 shows a comparison between the separator materials. The batteries studied here use PE separators, which have improved cyclic stability in comparison with PP separators.

**Table 3.** Comparison between the types of separators in pouch and cylindrical cells.

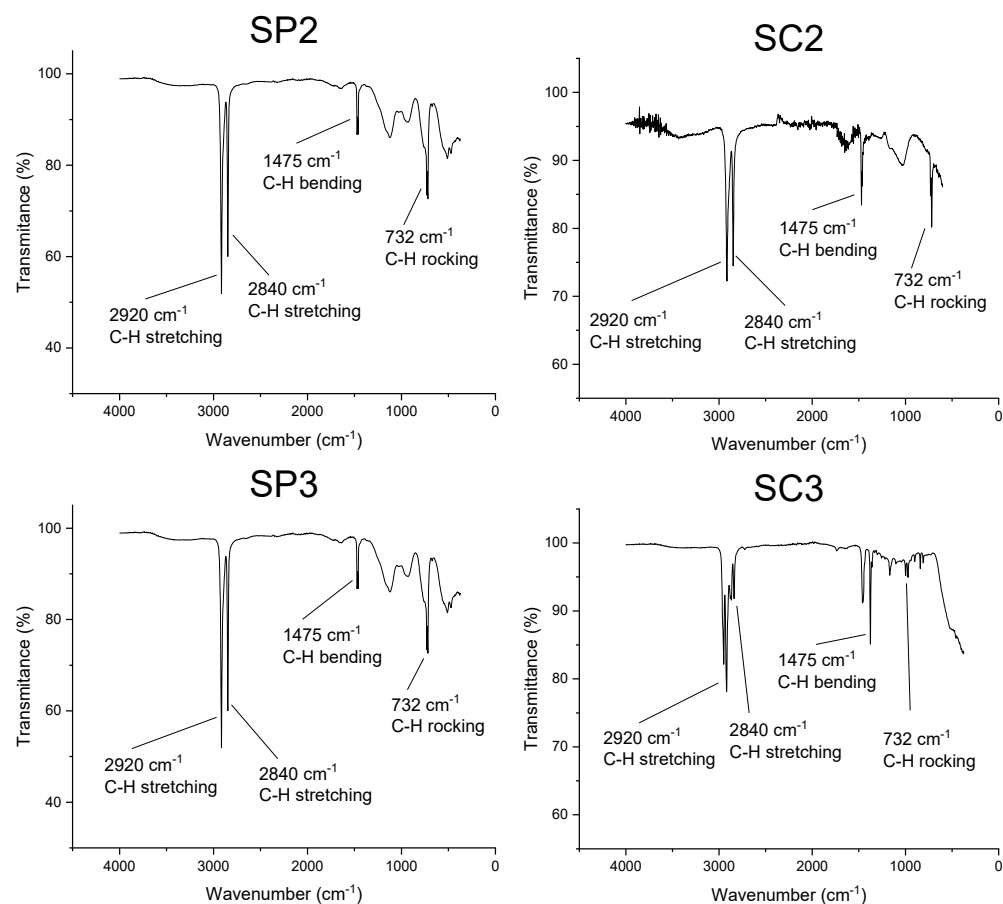
	P1	P2	P3	C1	C2	C3
Separator material	PE	PE	PE	PE	PE	PE

PE: polyethylene



**Figure 7.** Cont.





**Figure 7.** Analyses of the separators of the batteries identified by FTIR.

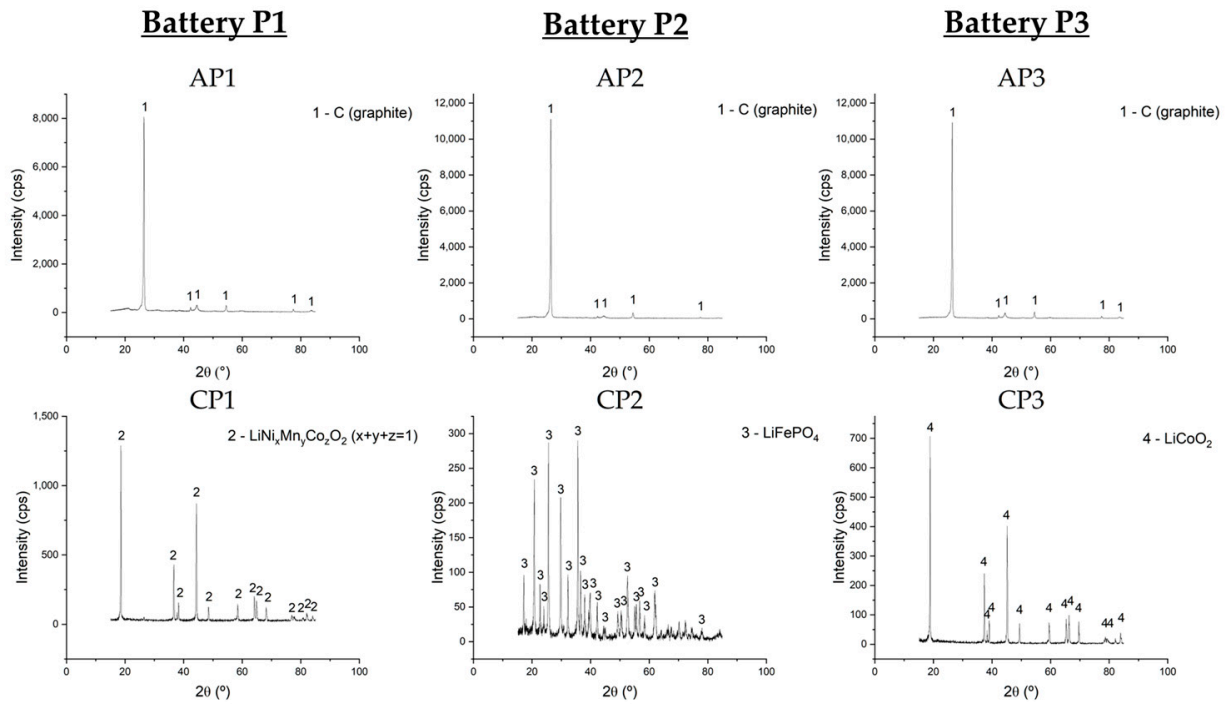
### 3.3. Characterization of the Electrodes—Cathode and Anode

The Al and Cu foils and the graphite were determined by gravimetric analysis, while the metallic content was determined by chemical analysis (Table 4).

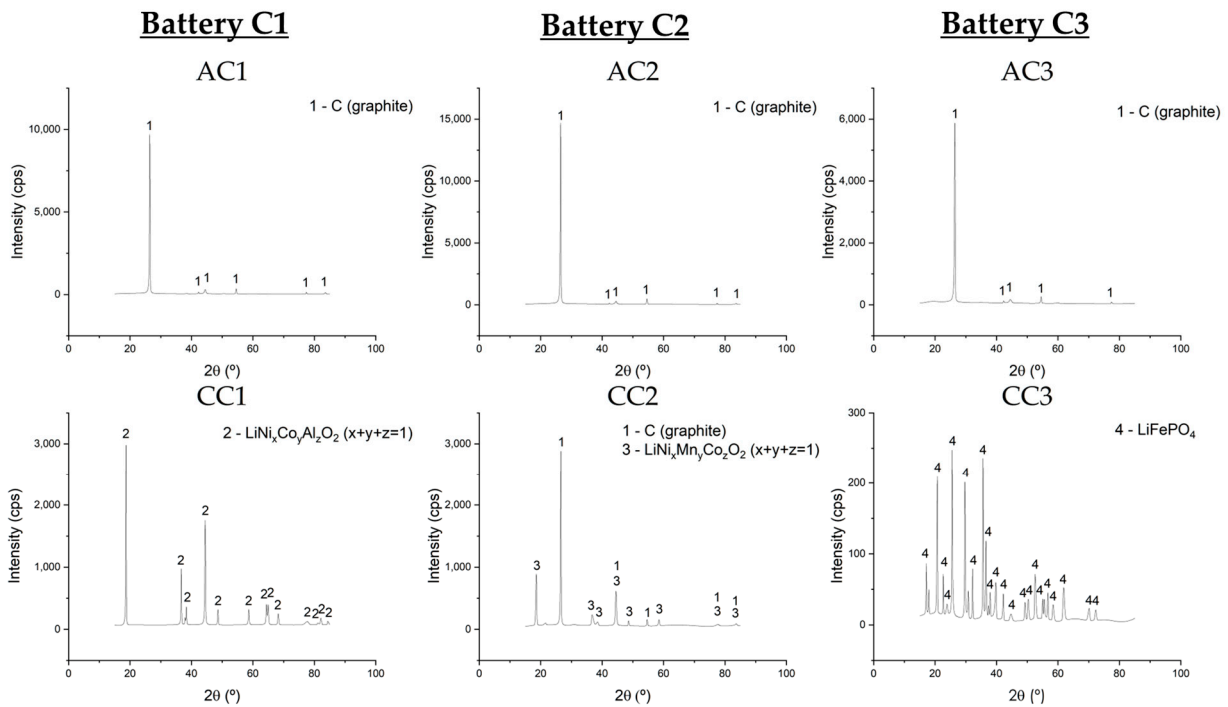
**Table 4.** Percentage of metals present in the cathode and in the anode of the samples.

Elements	Pouch Batteries						Cylindrical Batteries					
	Percentages in the Cathode (wt%)			Percentages in the Anode (wt%)			Percentages in the Cathode (wt%)			Percentages in the Anode (wt%)		
	CP1	CP2	CP3	AP1	AP2	AP3	CC1	CC2	CC3	AC1	AC2	AC3
Al	12.5	17.9	9.2	-	-	-	9.2	10.0	19.6	-	-	-
Cu	-	-	-	17.0	38.7	37.3	-	-	-	24.5	44.6	47.5
C	5.1	5.3	3.8	53.0	44.0	45.0	5.3	8.8	6.0	61.0	42.4	46.3
Co	7.4	-	45.5	-	-	-	6.5	6.4	-	-	-	-
Ni	17.6	-	-	-	-	-	45.7	35.3	-	-	-	-
Mn	20.1	-	-	-	-	-	-	6.8	-	-	-	-
Li	6.0	3.0	5.6	0.18	0.19	0.44	5.7	4.3	2.4	0.5	0.9	0.3
Fe	-	23.3	-	-	-	-	-	-	26.4	-	-	-
P	-	12.9	-	-	-	-	-	-	14.6	-	-	-

The active materials of the electrodes were also characterized by XRD determining their respective crystalline structures. As shown in Figure 8, CP1, CP2, and CP3 stand for the cathodes, and AP1, AP2, and AP3 represent the anodes for the batteries P1, P2, and P3, respectively. In Figure 9, the cathodes for the batteries C1, C2, and C3 are named CC1, CC2, and CC3, respectively; and the anodes are named AC1, AC2, and AC3, respectively.



**Figure 8.** XRD analyses showing crystalline structures of the active materials of the anodes and of the cathodes of pouch batteries P1, P2, and P3.



**Figure 9.** XRD analyses showing crystalline structures of the active materials of the anodes and of the cathodes of cylindrical batteries C1, C2, and C3.

According to Figures 8 and 9 and to Table 4, the anodes of these batteries were formed by carbon as graphite, which is the most common anode active material used in commercial LIBs. There are other materials also used as anode material, such as lithium titanate ( $\text{Li}_4\text{Ti}_5\text{O}_{12}$ ) and graphene. However, graphite stands out in LIB applications because of its high electrical conductivity, low cost, mature manufacturing method, and abundant supplies [3].

Cathode CP1 contains lithium-nickel-cobalt-manganese oxide ( $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$ ,  $x + y + z = 1$ ). Based on the chemical analysis and the XRD spectrum, it was determined that the cathode of the battery was composed of  $\text{LiNi}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$ , which classifies the P1 battery as an NMC 442 type. This type of battery combines the high specific energy of nickel, the good thermal stability of manganese, and the good energy density of cobalt. It is the one chosen for use in electric vehicles such as the Nissan Leaf, Chevy Volt, and BMW i3 [28,31].

Battery P2 contains a cathode composed of iron and lithium phosphate, being classified as an LFP battery ( $\text{LiFePO}_4$ ). This type is very safe due to high thermal stability, high durability, and high power/weight ratio, but has lower performance. The LFP batteries are used in motorhomes such as the Iridium E Mobil [31]. The analysis of battery C3 demonstrates that, similar to battery P2, there is a lithium-iron phosphate in the cathode ( $\text{LiFePO}_4$ ).

Battery P3 contains a cathode composed of lithium and cobalt oxide, classifying it as an LCO-type battery ( $\text{LiCoO}_2$ ). These batteries, used in the Tesla Roadster car and electronic equipment, have high energy density because of the presence of Co [28].

For battery C1, the cathode is composed of Li, Ni, Co, and Al in the form of a lithium-nickel-cobalt-aluminum oxide, classifying it as an NCA-type battery ( $\text{LiNiCoAlO}_2$ ). This type of battery has high power and energy densities, as well as good service life, making it a good option for EVs [28,31].

Battery C2 contains a cathode composed of lithium-nickel-manganese-cobalt oxide ( $\text{LiNiMnCoO}_2$ ). This indicates that the battery is classified as an NMC-type battery. Furthermore, it was determined that the molar ratio between Ni, Mn, and Co was 8:1:1 (Table 4), which indicates that the battery is NMC 811. The morphology of each active material carried out by SEM-EDS and the semiquantitative elementary analysis for the pouch batteries are shown in Figure 10, and those for the cylindrical batteries are shown in Figure 11.

These results confirm the composition of the electrodes, such as Al, C, Co, Ni, and Mn present in CP1 (NMC battery). CP2 contains Fe and P (LFP battery), while CP3 contains Co and C (LCO battery). The anodes AP1, AP2, and AP3 contain C in the form of graphite, as confirmed by previous XRD analysis. The presence of C in the cathode might be used to improve electrical conductivity [7].

The SEM-EDS analysis showed the existence of other elements, such as fluorine and oxygen. Fluorine was present due to the binder PVDF or residual electrolyte ( $\text{LiPF}_6$ ). The oxygen identified can be explained by the cathode oxide, while in the anode, it might originate from the formation of the SEI, which is an oxidation product of metallic lithium by the liquid electrolyte [32].

Based on the data previously shown, weight percentages of the battery materials are shown in Figure 12. Oxygen was calculated from the molecular formula of the crystalline structure of the cathode material and from the concentration obtained of other metals of the structure. The binder was calculated based by mass balance.

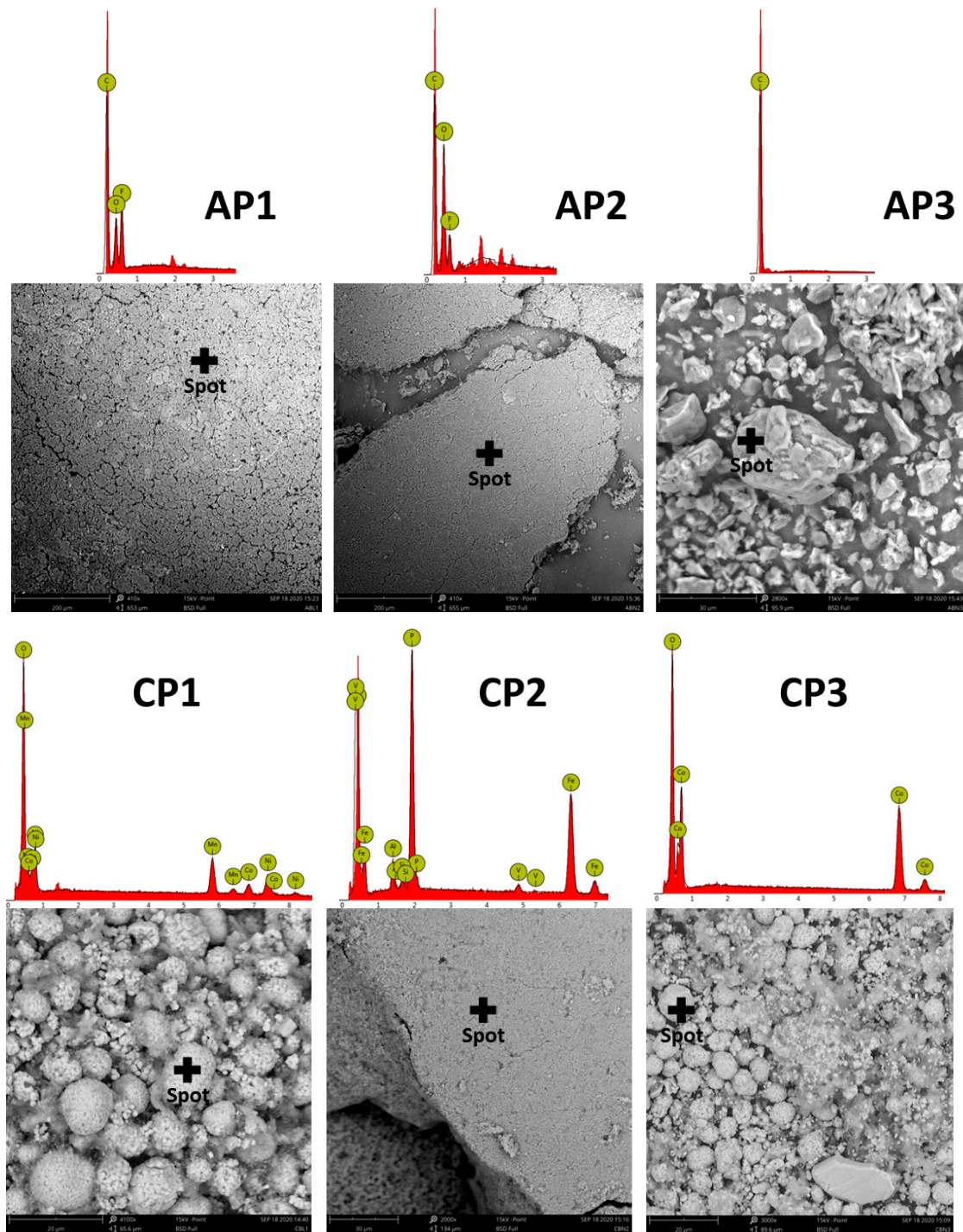
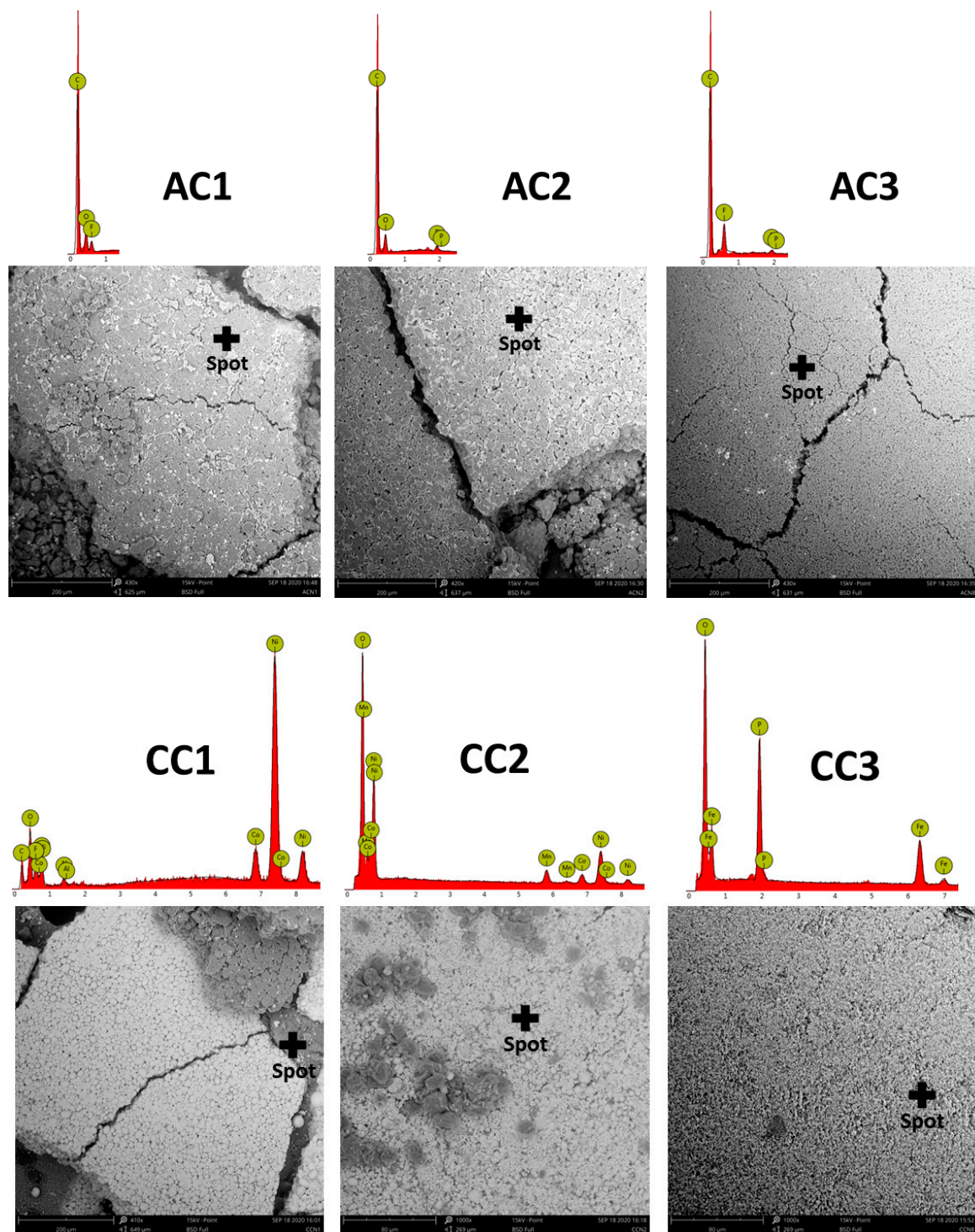


Figure 10. Morphology and semiquantitative analysis in SEM-EDS of the active materials of the three pouch batteries studied.





**Figure 11.** Morphology and semiquantitative analysis in SEM-EDS of the active materials of the three cylindrical batteries studied.

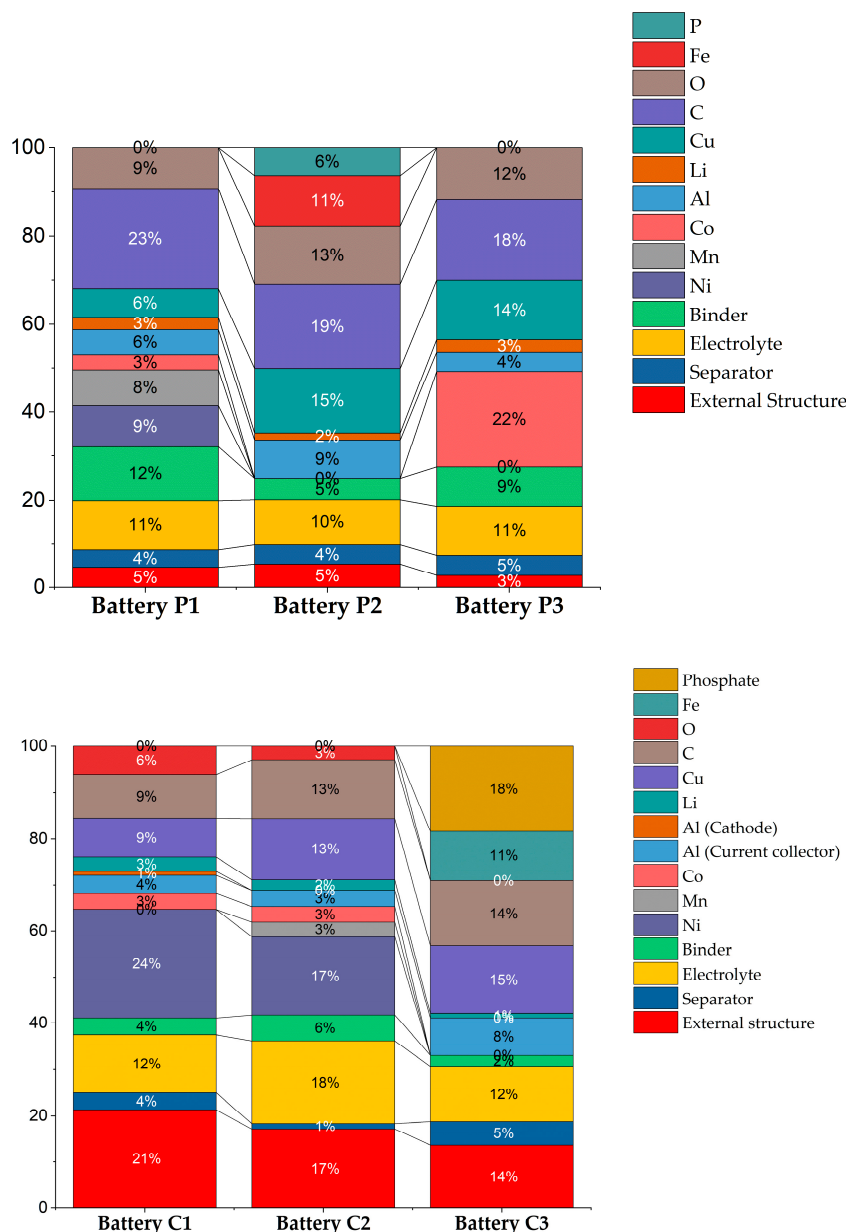


Figure 12. Weight percentage (wt%) of the components and elements in each of the pouch batteries P1, P2, and P3, and in each of the cylindrical batteries C1, C2, and C3.

### 3.4. Discussions and Preliminary Economic Evaluation

#### 3.4.1. Availability of LIBs and Composition Variation

In this work, three pouch LIBs, P1, P2, and P3, of different compositions, were characterized as NMC 442, LFP, and LCO, respectively. In addition, three cylindrical LIBs, C1, C2, and C3, were characterized as NCA, NMC 811, and LFP, respectively. Batteries with LCO cathodes emerged in the 1980s as one of the first cathodes developed, dominating more than 55% of the LIB total market until 2015. This battery’s market share has declined due to the introduction of newer cathodes and may come to an end in the next few years, since it offers low thermal stability and is thus becoming unsuitable for energy-intense applications such as EVs and energy storage devices [2]. According to the data provided by Statista, LCO batteries are no longer in the market for EVs, being only used in some electronic equipment [33].

NMC-type batteries are constantly being developed to reduce the cobalt content. There are several compositions, including 333, 422, 532, 622, and 811 (proportion of Ni,

Mn, and Co, respectively). This battery has begun to dominate the LIBs market since 2020, and it is estimated that the NMC battery market will continue to grow in the coming years, especially for variants with a lower amount of cobalt. In addition, there are currently studies in the literature on the synthesis and use of LIB cathodes with  $\text{LiNi}_{0.9}\text{Mn}_{0.05}\text{Co}_{0.05}\text{O}_2$  chemistry (which would be considered a NMC 9½½-type battery) in devices with optimal electrochemical properties [34].

The LFP battery was the most recently developed cathode that attracted attention due to the aforementioned properties, including low cost and high stability. In addition, the increasing scarcity of Co in the world's reserves has also led to the increase in the use of batteries without this metal in their composition, such as LFP. In 2022, China had the largest electric car market, which led to a substantial waste generation from LFP used batteries alone, confirming that this type of LIB is still a trend [35].

NCA batteries account for 8% of the market. This battery type is utilized for particular applications because it has a high specific energy, good specific power, and a long service life when compared to other types of LIBs. However, the downsides are connected to safety and production costs. NCA cathodes are an appealing alternative for EV and HEV due to their high energy and power densities, as well as their long service life. Nevertheless, the literature lacks recycling research and pathways to a cost-effective recycling procedure for this type of battery [6].

The interest in recycling LIBs is valid, owing to the number of batteries produced and disposed of every year, as well as the growth of the electric car market. In 2021, 6.61 million battery-powered and plug-in hybrid electric vehicles were sold worldwide. In 2022, the number rose to 10.25 million, representing an increase of 55% in just one year. It is predicted to have 17.07 million sales of these vehicles around the world in 2028 [33,36].

LIBs have a lifetime of approximately 10 years, meaning that LIB waste has already started to be produced. It is estimated that approximately 11 million LIBs will reach the end of their life by 2030, and the production of this waste will continue to grow, as will the use of electric vehicles, which is expected to reach 140 million by 2030 [8,33].

#### 3.4.2. Urban Mining Throughout Battery Recycling

The percentage of metals present in the battery electrodes studied shows the feasibility of using them as a secondary source of mineral resources, as the percentage of certain metals is higher than in primary sources, as in the case of cobalt, which is present in 45.5% of the P3 cathode (LCO batteries). This represents a much higher percentage than many common minerals that contain cobalt, such as cobaltite (35.52%) [34]. The nickel present in the NMC 442, in the NMC 811, and in the NCA batteries studied has a content of 8.9%, 16.9%, and 23.7%, respectively.

Comparing the amounts of Li present in the batteries in question with those in the mineral sources containing this element, the NCA, NMC 442, NMC 811 and LCO batteries studied contain amounts equal to or greater than the amounts of Li found in minerals such as lepidolite (3%–4%), petalite (3%–4.5%), spodumene (1.5%–7%), and zinnwalite (2%–5%). The mineral jacobsonite, which contains around 24% Mn, has only slightly more of this element compared to the cathode of the NMC 442 battery studied, which contains 20.1% Mn [35].

Therefore, this demonstrates how the recycling of LIBs can be significantly important in order to represent another source of critical metals and avoid obtaining them directly from the global reserves.

#### 3.4.3. Preliminary Potential Economic Evaluation

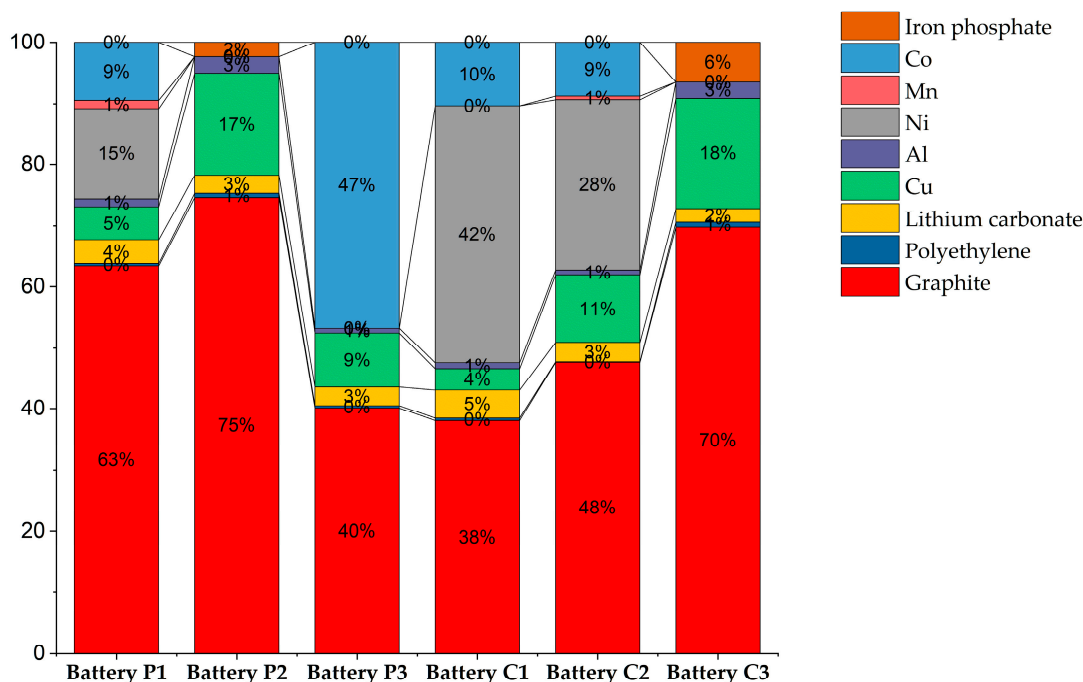
From the values shown in Figure 12 and the market prices, it was possible to calculate an approximate potential economic value to be recovered from the batteries (Table 5 and Figure 13). Here, our goal was the analysis of the material present in a ton of each Li-ion battery cell we characterized. We did not consider recycling costs, only the values of the cells themselves according to physical and chemical composition.

**Table 5.** Prices of the products and the final value of each battery, in USD/t.

Products	Prices (USD/t)
Li <sub>2</sub> CO <sub>3</sub>	14,093.70
FePO <sub>4</sub>	1510.72
Al	2313.00
Cu	8528.00
Ni	16,400.00
Mn	1752.03
Co	28,205.00
Graphite	28,800.00
Polyethylene	1109.66

Batteries	Final price (USD/t)
P1 (NMC 442)	9856.38
P2 (LFP)	1708.43
P3 (LCO)	12,718.57
C1 (NCA)	7541.06
C2 (NMC 811)	9113.47
C3 (LFP)	6719.10



**Figure 13.** Percentages of the price of each component and compound relative to the total price of the correspondent battery.

The prices (USD/t) for Al, Cu, Co, and Ni were obtained from the London Metal Exchange website; the prices of FePO<sub>4</sub> and Mn were extracted from the Shanghai Metals Market (SMM) website; the price of Li<sub>2</sub>CO<sub>3</sub> and polyethylene were obtained from the Trading Economics website; and the price of graphite was obtained from the Lab Alley website.

Table 5 shows the prices of the metals and products, as well as the final total value of each battery studied, in USD/t. The value of lithium was expressed in the form of lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>), as it is the commercialized form. Furthermore, the price of FePO<sub>4</sub> was considered instead of the separate prices of Fe and P. Ni, Co, Mn, Cu, and Al prices were obtained considering their metallic form.

Polyethylene has a lower economic value (USD 1109.66/t as of December 2023, by Trading Economics) compared to the critical metals in the batteries. For this reason, many



companies do not properly separate them during battery recycling in addition to low battery content (about 4%). However, in line with a circular economy, recycling this polymer is also important [13].

In the case of the P1 battery, which has a cathode of NMC 442, its value is USD 37,836.5/t of cells. Approximately 69% of the value of the battery is in the cathode, more specifically in Co (14.9%), Ni (17.3%) and  $\text{Li}_2\text{CO}_3$  (37.2%). The value of Cu (the anode current collector) is also quite significant in the composition of the value of this battery (22.5%). It is important to mention that the final value of the battery was determined considering the adjustments on the prices of Ni, Mn, and Co according to the ratio among them (4:4:2).

The P2 battery—LFP cathode—has a value of USD 26,445.4 /t. Over 59% of the value is in the cathode, specifically in  $\text{Li}_2\text{CO}_3$  (53.3%) and in  $\text{FePO}_4$  (5.7%), and 32.2% is concentrated in the Cu present in the anode. The P3 battery (LCO cathode) has a value of USD 53,139.7/t. Almost 80% of the value is in the cathode, being 53.1% Co and 26.5%  $\text{Li}_2\text{CO}_3$ . The Cu in the anode represents 16% of the value of the battery.

In all the batteries, the active material from the cathode is primarily responsible for the value of the batteries, being the focus for recycling processes. Battery P2 has the lowest cathode value among the three pouch batteries. This can be due to the low value of iron phosphate in the market (1510.72 USD/t). Battery P3 is the most valuable pouch battery evaluated in this work, due to the large amount of cobalt. It is the most valuable element due to its unstable supply, as its reserves are concentrated in the Democratic Republic of Congo, a country with great political instability. In addition, among the metals present, Co is the one with the biggest global scarcity [3,36].

Among the cylindrical batteries studied, battery C2 (with an NMC 811 cathode) had the highest price, making it the most valuable and in the highest market demand in December 2023. This might be due to the high amount of Ni in the battery (15.6% in the whole battery cell), which is one of the critical metals, with a high price in the market (16,400 USD/t). Cylindrical battery C3 (LFP cathode) was the one with the lowest value among the cylindrical LIBs studied. Again, this can be due to the low value of  $\text{FePO}_4$ .

#### 3.4.4. Recycling Technologies towards Circular Economy

There are some widely used technologies for urban waste recycling and metal recovery, which are also consolidated LIB recycling techniques. Focusing on the extraction of the metals found in the cathode, the most common technologies are pyrometallurgy and hydrometallurgy. Pyrometallurgical procedures are widely used to treat high-grade ores, such as iron ore, whereas hydrometallurgical processes are generally used to extract low-grade ores, such as copper [37]. The oldest one, pyrometallurgy, is a thermal extraction technique, in which the use of extreme temperatures is responsible for the extraction of metals [26,38]. Some of the popular pyrometallurgical processes include smelting, incineration, pyrolysis, and molten salt processing [39].

However, even though the energy usage and carbon footprint are lower in pyrometallurgy than those used in the primary production of the metals of interest in the recovery process, the high energy demand and the production and discharge of hazardous gases (such as  $\text{CO}_2$ ) into the environment are the key restrictions related to this technology. Furthermore, only Ni and Co from the cathode's active material and Cu from the current collector are recovered in pyrometallurgical routes. On the other hand, Li, Al, and other materials, such as polymers, graphite are not recovered [39].

Hydrometallurgy is a technology used for performing the extraction of the metals from aqueous media. It involves processes such as leaching, dissolving the metallic fraction for further separation, and recovery. This technology has become a promising because of its favorable recovery rate and excellent purity of the product. In addition, even though it involves a long recovery process and has a high chemical reagent consumption, it features low energy usage, high metal recovery rates, and low emissions [3,38]. Table 6 summarizes and highlights some comparisons between those two metallurgical processes.

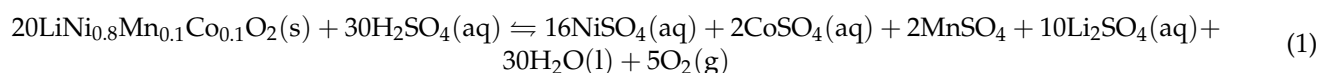
**Table 6.** Some comparisons between pyrometallurgy and hydrometallurgy processes [3].

	Pyrometallurgy	Hydrometallurgy
Initial Capital Investments	High	Low
Primary Waste Product	Air and solids	Liquids and solids
Energy Consumption	High	Low
Level of Separations	Low	High
Rate of Chemical Reaction	High	Low
Production Costs	Low	Intermediate

A LIB cell also contains other components that should not be ignored during a recycling process. Since the separator (polymer), the current collector foils (metallics Al and Cu), the electrolyte, and the external structure are made of materials that are common in all LIBs, their recovery can be performed during the pretreatment step of the LIB recycling process, prior to the leaching step, if a hydrometallurgical route is considered. This can involve dismantling, separation, surface modification or a combination of at least two of those methods. This physical separation treatment is thus important because it allows the recovery of these typical materials in a single pretreatment process [40].

Following the pretreatment in a hydrometallurgy recycling route, the leaching process is responsible for dissolving the cathode materials in chemicals (also called leaching agents), such as HCl, HNO<sub>3</sub>, and H<sub>2</sub>SO<sub>4</sub> (leaching with inorganic acids), citric, malic, and oxalic acids (leaching with organic acids), and even alkaline chemicals such as NaOH and ammonia (alkaline leaching). Later, during the same process of leaching, the cathode materials are separated and extracted. The literature also reports the addition of reducing agents such as H<sub>2</sub>O<sub>2</sub> to the solution in order to optimize the efficiency of extraction of the metals of interest [41,42].

The leaching experiments are conducted by studying parameters such as the leaching and reducing agents' concentrations, temperature, reaction time, and solid/liquid ratio, which is a ratio between the mass of the solid sample and the volume of the leaching agent. After the leaching experiment, the solution is filtered, and a leaching liquor is generated. Equation (1) shows an example of the chemical reaction between the cathode's active material of an NMC 811 battery and H<sub>2</sub>SO<sub>4</sub> [41]. In contrary, in Equation (2), the chemical reaction occurs between the cathode's active material of a LCO battery and citric acid. Neither equation includes H<sub>2</sub>O<sub>2</sub> in the reaction.



The leaching process is followed by a purification step, in which techniques such as selective precipitation, solvent extraction, and ion exchange resins are used. This step is necessary and responsible for obtaining high-purity products that already have great commercial value for the industries. For the precipitation, the objective is to add some reagent (also called precipitating agent) into the leaching liquor in order to selectively extract the desired compound, with the metal of interest in the solid form [43]. The solution's composition determines the precipitating agent that will be used.

Solvent extraction is a liquid–liquid separation process in which the target metal is transported from the aqueous phase (the solution studied) to an organic phase, which is an organic extractant. This extractant is diluted in kerosene or related organic substances, and its concentration is one parameter studied, along with temperature, time, the pH of the solution, and the aqueous/organic ratio [44]. The use of an organic extractant will depend on what metal is targeted to be extracted.

The ion exchange technique uses resins with specific functional groups, responsible for selectively adsorbing the metal of interest and extracting it. Similarly to the precipitation and the solvent extraction techniques, the choice of the resin used is related to the

composition of the solution and to what metal will be extracted. In addition, this technique can be performed by batch or continuously, in columns [45–50].

All these technologies and techniques used in hydrometallurgical processes are fundamental to developing a secondary source of critical materials present in LIBs, since they can recover over 95% of these materials. In other words, they contribute to the development of a circular economy, since it is possible to reuse the materials and compounds in the production of new LIBs, preserving global reserves and avoiding e-waste disposal. Moreover, hydrometallurgy, applied from a circular economy perspective, allows low energy consumption and low emissions of CO<sub>2</sub>.

#### 4. Conclusions

The complete characterization and economic evaluation of lithium-ion batteries with different compositions are crucial for assessing the economic viability of recycling, and they are fundamental for establishing strategies for their treatment from a circular economy perspective. This approach transforms battery waste into a secondary source of (critical) raw materials.

This work has characterized three pouch LIBs, including NMC 442, LFP, and LCO, and three cylindrical LIBs, including NCA, NMC 811, and LFP. The characteristics between pouch and cylindrical batteries have been punctuated with regard to their different external structures (Al with plastic for pouch cells and stainless steel for cylindrical cells), the different internal arrangement of their components, and their similarities, such as polyethylene and graphite, which are used as materials in all their separators and anodes, respectively. In addition, a preliminary economic evaluation has been performed to determine the materials with the highest market trend (December/2023) based on their compositions: LCO and NMC 811. It has also been determined that different compositions of the cathodes change the value of the battery.

Therefore, the following conclusions have been drawn based on theoretical and experimental studies:

- The characterization step of the LIBs shows fundamental importance during the treatment of this e-waste since it helps to develop the next recovery strategies during the recycling process. Therefore, this step should be detailed more in the literature.
- The batteries discharged with a resistor do not generate toxic gases, losses of materials, or subproducts, and a resistor must be used in industrial processes to improve sustainability.
- An electrolyte is a toxic component and must be recovered by evaporation in recycling processes.
- The cathode is the most valuable component in the LIBs depending on the battery type, which is different for LFPs.
- Cobalt is the most valuable metal present in some of the LIBs studied.
- Metals, such as Co and Ni, are more concentrated in battery waste compared to their primary sources. For this reason, the recycling process represents a valuable and green approach.
- A recycling process based on a circular economic perspective brings benefits to the environment, such as the preservation of the global reserves of critical raw materials used in LIBs, as well as low emissions of hazardous gases such as CO<sub>2</sub> into the environment.

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## References

1. Zheng, X.; Zhu, Z.; Lin, X.; Zhang, Y.; He, Y.; Cao, H.; Sun, Z. A Mini-Review on Metal Recycling from Spent Lithium Ion Batteries. *Engineering* **2018**, *4*, 361–370. [CrossRef]
2. Chakraborty, S.C.; Qamruzzaman, M.; Zaman, M.W.U.; Alam, M.M.; Hossain, M.D.; Pramanik, B.K.; Nguyen, L.N.; Nghiem, L.D.; Ahmed, M.F.; Zhou, J.L.; et al. Metals in E-Waste: Occurrence, Fate, Impacts and Remediation Technologies. *Process Saf. Environ. Prot.* **2022**, *162*, 230–252. [CrossRef]
3. Harper, G.; Sommerville, R.; Kendrick, E.; Driscoll, L.; Slater, P.; Stolkin, R.; Walton, A.; Christensen, P.; Heidrich, O.; Lambert, S.; et al. Recycling Lithium-Ion Batteries from Electric Vehicles. *Nature* **2019**, *575*, 75–86. [CrossRef] [PubMed]
4. Lyu, Y.; Wu, X.; Wang, K.; Feng, Z.; Cheng, T.; Liu, Y.; Wang, M.; Chen, R.; Xu, L.; Zhou, J.; et al. An Overview on the Advances of LiCoO<sub>2</sub> Cathodes for Lithium-Ion Batteries. *Adv. Energy Mater.* **2021**, *11*, 2000982. [CrossRef]
5. Ferg, E.E.; Schuldt, F.; Schmidt, J. The Challenges of a Li-Ion Starter Lighting and Ignition Battery: A Review from Cradle to Grave. *J. Power Sources* **2019**, *423*, 380–403. [CrossRef]
6. de Castro, R.H.; Romano Espinosa, D.C.; Gobo, L.A.; Kumoto, E.A.; Botelho Junior, A.B.; Tenorio, J.A.S. Design of Recycling Processes for NCA-Type Li-Ion Batteries from Electric Vehicles toward the Circular Economy. *Energy Fuels* **2024**, *38*, 5545–5557. [CrossRef]
7. Guimarães, L.F.; Botelho Junior, A.B.; Espinosa, D.C.R. The Characterization of Li-Ion Batteries and the Importance of the Recycling Processes. *JOM* **2023**, *75*, 3622–3631. [CrossRef]
8. Xia, X.; Li, P.; Xia, Z.; Wu, R.; Cheng, Y. Life Cycle Carbon Footprint of Electric Vehicles in Different Countries: A Review. *Sep. Purif. Technol.* **2022**, *301*, 122063. [CrossRef]
9. International Energy Agency. Global Critical Minerals Outlook. 2024. Available online: [https://www.iea.org/reports/global-critical-minerals-outlook-2024?utm\\_content=buffer3599f&utm\\_medium=social&utm\\_source=linkedin.com&utm\\_campaign=buffer](https://www.iea.org/reports/global-critical-minerals-outlook-2024?utm_content=buffer3599f&utm_medium=social&utm_source=linkedin.com&utm_campaign=buffer) (accessed on 24 July 2024).
10. USGS. Reserves & Production of Various Elements. *USGS Miner. Resour. Program* **2018**, *1*, 2016–2017. [CrossRef]
11. Bobba, S.; Carrara, S.; Huisman, J.; Mathieux, F.; Pavel, C. *Critical Raw Materials for Strategic Technologies and Sectors in the EU—A Foresight Study*; European Union: Brussels, Belgium, 2020; ISBN 9789276153375.
12. Sonoc, A.; Jeswiet, J.; Soo, V.K. Opportunities to Improve Recycling of Automotive Lithium Ion Batteries. *Procedia CIRP* **2015**, *29*, 752–757. [CrossRef]
13. Velázquez-Martínez, O.; Valio, J.; Santasalo-Aarnio, A.; Reuter, M.; Serna-Guerrero, R. A Critical Review of Lithium-Ion Battery Recycling Processes from a Circular Economy Perspective. *Batteries* **2019**, *5*, 68. [CrossRef]
14. Lv, L.; Hor, M.P.; Su, F.; Zhao, X.S. Competitive Adsorption of Pb<sup>2+</sup>, Cu<sup>2+</sup>, and Cd<sup>2+</sup> Ions on Microporous Titanosilicate ETS-10. *J. Colloid Interface Sci.* **2005**, *287*, 178–184. [CrossRef] [PubMed]
15. Pagliaro, M.; Meneguzzo, F. Lithium Battery Reusing and Recycling: A Circular Economy Insight. *Heliyon* **2019**, *5*, e01866. [CrossRef] [PubMed]
16. Martins, L.S.; Rovani, S.; Botelho Junior, A.B.; Romano Espinosa, D.C. Sustainable Approach for Critical Metals Recovery through Hydrometallurgical Processing of Spent Batteries Using Organic Acids. *Ind. Eng. Chem. Res.* **2023**, *62*, 18672–18682. [CrossRef]
17. Guimarães, L.F.; Botelho Junior, A.B.; Espinosa, D.C.R. Sulfuric Acid Leaching of Metals from Waste Li-Ion Batteries without Using Reducing Agent. *Min. Eng.* **2022**, *183*, 107597. [CrossRef]
18. Zachmann, N.; Petranikova, M.; Ebin, B. Electrolyte Recovery from Spent Lithium-Ion Batteries Using a Low Temperature Thermal Treatment Process. *J. Ind. Eng. Chem.* **2023**, *118*, 351–361. [CrossRef]
19. Mao, Z.; Song, Y.; Zhen, A.G.; Sun, W. Recycling of Electrolyte from Spent Lithium-Ion Batteries. *Next Sustain.* **2024**, *3*, 100015. [CrossRef]
20. Zou, H.; Gratz, E.; Apelian, D.; Wang, Y. A Novel Method to Recycle Mixed Cathode Materials for Lithium Ion Batteries. *Green Chem.* **2013**, *15*, 1183–1191. [CrossRef]
21. Chen, M.; Zheng, Z.; Wang, Q.; Zhang, Y.; Ma, X.; Shen, C.; Xu, D.; Liu, J.; Liu, Y.; Gionet, P.; et al. Closed Loop Recycling of Electric Vehicle Batteries to Enable Ultra-High Quality Cathode Powder. *Sci. Rep.* **2019**, *9*, 1654. [CrossRef]
22. Xiao, J.; Guo, J.; Zhan, L.; Xu, Z. A Cleaner Approach to the Discharge Process of Spent Lithium Ion Batteries in Different Solutions. *J. Clean. Prod.* **2020**, *255*, 120064. [CrossRef]
23. Amalia, D.; Singh, P.; Zhang, W.; Nikoloski, A.N. Discharging of Spent Cylindrical Lithium-Ion Batteries in Sodium Hydroxide and Sodium Chloride for a Safe Recycling Process. *JOM* **2023**, *75*, 4946–4957. [CrossRef]
24. Mossali, E.; Picone, N.; Gentilini, L.; Rodríguez, O.; Pérez, J.M.; Colledani, M. Lithium-Ion Batteries towards Circular Economy: A Literature Review of Opportunities and Issues of Recycling Treatments. *J. Environ. Manag.* **2020**, *264*, 110500. [CrossRef] [PubMed]
25. Xu, J.; Thomas, H.R.; Francis, R.W.; Lum, K.R.; Wang, J.; Liang, B. A Review of Processes and Technologies for the Recycling of Lithium-Ion Secondary Batteries. *J. Power Sources* **2008**, *177*, 512–527. [CrossRef]



26. Huang, J.; Boles, S.T.; Tarascon, J.M. Sensing as the Key to Battery Lifetime and Sustainability. *Nat. Sustain.* **2022**, *5*, 194–204. [CrossRef]
27. He, L.P.; Sun, S.Y.; Song, X.F.; Yu, J.G. Recovery of Cathode Materials and Al from Spent Lithium-Ion Batteries by Ultrasonic Cleaning. *Waste Manag.* **2015**, *46*, 523–528. [CrossRef]
28. Miao, Y.; Hynan, P.; von Jouanne, A.; Yokochi, A. Current Li-Ion Battery Technologies in Electric Vehicles and Opportunities for Advancements. *Energies* **2019**, *12*, 1074. [CrossRef]
29. Zhang, L.; Xu, Z. A Review of Current Progress of Recycling Technologies for Metals from Waste Electrical and Electronic Equipment. *J. Clean. Prod.* **2016**, *127*, 19–36. [CrossRef]
30. Georgi-Maschler, T.; Friedrich, B.; Weyhe, R.; Heegn, H.; Rutz, M. Development of a Recycling Process for Li-Ion Batteries. *J. Power Sources* **2012**, *207*, 173–182. [CrossRef]
31. Hannan, M.A.; Hoque, M.M.; Hussain, A.; Yusof, Y.; Ker, P.J. State-of-the-Art and Energy Management System of Lithium-Ion Batteries in Electric Vehicle Applications: Issues and Recommendations. *IEEE Access* **2018**, *6*, 19362–19378. [CrossRef]
32. Kabir, M.M.; Demirocak, D.E. Degradation Mechanisms in Li-Ion Batteries: A State-of-the-Art Review. *Int. J. Energy Res.* **2017**, *41*, 1963–1986. [CrossRef]
33. Statista Electric Cars an Overview of the Electric Car Industry and Associated Technology. 2019. Available online: <https://www.statista.com/study/62346/statista-dossierplus-on-the-electric-car-industry/> (accessed on 24 July 2024).
34. van den Brink, S.; Kleijn, R.; Sprecher, B.; Tukker, A. Identifying Supply Risks by Mapping the Cobalt Supply Chain. *Resour. Conserv. Recycl.* **2020**, *156*, 104743. [CrossRef]
35. Wesselkämper, J.; Dahrendorf, L.; Mauler, L.; Lux, S.; von Delft, S. A Battery Value Chain Independent of Primary Raw Materials: Towards Circularity in China, Europe and the US. *Resour. Conserv. Recycl.* **2024**, *201*, 107218. [CrossRef]
36. Statista Electric Vehicles Worldwide. 2020. Available online: <https://www.statista.com/study/11578/electric-vehicles-statista-dossier/> (accessed on 24 July 2024).
37. Shittu, O.S.; Williams, I.D.; Shaw, P.J. Global E-Waste Management: Can WEEE Make a Difference? A Review of e-Waste Trends, Legislation, Contemporary Issues and Future Challenges. *Waste Manag.* **2021**, *120*, 549–563. [CrossRef]
38. Alves Dias, P.; Blagoeva, D.; Pavel, C.; Arvanitidis, N. *Cobalt: Demand-Supply Balances in the Transition to Electric Mobility*; Publications Office of the European Union: Luxembourg, 2018; ISBN 9789279943119.
39. Reddy, R.G. *Applications of Process Engineering Principles in Materials Processing, Energy and Environmental Technologies*; Springer International Publishing: New York, NY, USA, 2017; ISBN 978-3-319-51090-3.
40. Or, T.; Gourley, S.W.D.; Kaliyappan, K.; Yu, A.; Chen, Z. Recycling of Mixed Cathode Lithium-Ion Batteries for Electric Vehicles: Current Status and Future Outlook. *Carbon Energy* **2020**, *2*, 6–43. [CrossRef]
41. Crundwell, F.K.; Moats, M.S.; Ramachandran, V.; Robinson, T.G.; Davenport, W.G. *Extractive Metallurgy of Nickel, Cobalt and Platinum-Group Metals, 1st, ed.*; Elsevier: Oxford, UK, 2011; ISBN 978-0-08-096809-4.
42. Träger, T.; Friedrich, B.; Weyhe, R. Recovery Concept of Value Metals from Automotive Lithium-Ion Batteries. *Chem. Ing. Tech.* **2015**, *87*, 1550–1557. [CrossRef]
43. Ebin, B.; Isik, M.I. *Pyrometallurgical Processes for the Recovery of Metals from WEEE*; Elsevier Inc.: Amsterdam, The Netherlands, 2016; ISBN 9780128033647.
44. Premathilake, D.S.; Botelho Junior, A.B.; Tenório, J.A.S.; Espinosa, D.C.R.; Vaccari, M. Designing of a Decentralized Pretreatment Line for EOL-LIBs Based on Recent Literature of LIB Recycling for Black Mass. *Metals* **2023**, *13*, 374. [CrossRef]
45. Benjamasutin, P.; Promphan, R. Recycling of Lithium-Ion Batteries—Determination of Optimal Parameters for the Application of Hydrogen Peroxide as Reducing Agent in the Leaching Process. Master’s Thesis, Chalmers University of Technology, Gothenburg, Sweden, 2020.
46. Nayl, A.A.; Hamed, M.M.; Rizk, S.E. Selective Extraction and Separation of Metal Values from Leach Liquor of Mixed Spent Li-Ion Batteries. *J. Taiwan Inst. Chem. Eng.* **2015**, *55*, 119–125. [CrossRef]
47. Lv, W.; Wang, Z.; Cao, H.; Sun, Y.; Zhang, Y.; Sun, Z. A Critical Review and Analysis on the Recycling of Spent Lithium-Ion Batteries. *ACS Sustain. Chem. Eng.* **2018**, *6*, 1504–1521. [CrossRef]
48. Kislik, V.S. *Solvent Extraction: Classical and Novel Approaches*; Elsevier: Amsterdam, The Netherlands, 2014; ISBN 978-0-444-53778-2.
49. Ahamed, M.I.; Asiri, A.M. *Applications of Ion Exchange Materials in the Environment*; Springer International Publishing: New York, NY, USA, 2019; ISBN 9783030104290.
50. Inamuddin, M.L. (Ed.) *Ion Exchange Technology I*; Springer: Dordrecht, The Netherlands, 2012; ISBN 978-94-007-1699-5.

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