

Review of Research on Li-Ion Batteries Waste Management

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Abstract

Li-ion batteries (Libs) are a mature technology widely used for energy storage in various electronic devices. Nowadays, this technology has become a leading candidate for the portable electronics market and for electric vehicles due to its good performance. As a result, the demand for Libs containing critical metals, rare earth elements and precious metals is increasing day by day with the accelerated upgrades of consumer electronics, which promotes the supply risk of many mining resources. In addition, the problems associated with the production of end-of-life Lib are increasing on a global scale. Used Libs are e-waste containing significant levels of critical raw materials (such as Co, Li, Mn and Ni) along with harmful substances. Without proper management of Lib waste, these precious metals and toxic substances may end up in nature and cause environmental and public health problems. In order to preserve nature, ensure sustainable resource management and stimulate the circular economy, it has become crucial to properly manage and recycle end-of-life Li-ion batteries. By the way, conventional methods focusing on pyrometallurgical treatments combined with hydrometallurgical treatment are widely studied to recover design metals from Libs waste. It is in this context that we have conducted this bibliographic synthesis, focusing on the efficiency of the solvents employed and their competitiveness for a more environmentally friendly economic management. In this manuscript, recent leaching, solvent extraction, electrodeposition and precipitation strategies to recover precious metals from end-of-life Li-ion battery designs are reviewed and the evolution of these processes is discussed.

Keywords

Libs Waste, Leaching, Extraction Rate, Affinity, Partition Coefficient

1. Introduction

In recent decades, making certain aspects of life much easier, electronic devices that are increasingly small, manageable, portable and economical have invaded our daily lives. In fact, there has been a meteoric rise in the number of cell phones, laptops, digital devices, electric and hybrid vehicles, etc. [1] [2]. These technologies work through electrochemical systems, batteries, storing chemical energy that they reversibly restore in electrical energy during their operation. For performance reasons, improvement studies are widely carried out, leading to the development of various types of batteries such as lead acid, nickel-cadmium (Ni-Cd), nickel-metal hydride (Ni-MH), Li-ion (Libs), lithium air, etc. [3] [4] [5]. After efficiency tests, Li-ion batteries have been shown to perform better due to higher energy densities, lower self-discharge rates, higher cyclabilities and no memory effect, high capacity, etc., hence the large-scale production of Li-ion batteries for consumer electronics. According to a study, the production of Li-ion batteries increased from 500 to 700 million tons from 2000 to 2004, while the number of users exceeded 6.8 billion in 2013 [6]. Due to the accelerated upgrading of consumer electronics, we are witnessing a surge in the production of used Li-ion batteries. Based on studies of the cyclability of Li-ion batteries [7] [8] [9], results had predicted the production of waste Libs in China to be around 500,000 tons in 2020 [8].

On the other hand, a Li-ion battery is essentially composed of a negative electrode, made of LiC_6 graphite, a positive electrode, a separator and an electrolyte, made of a mixture of solvents such as DMC, DEC, EC and a lithium salt (LiPF_6) [10] [11] [12]. The positive electrodes, which have a crucial role to play on the performance of the systems, often made of metal oxide (LiMO_2), are usually composed of heavy metals (Co, Mn, Ni, etc.) in relatively high concentration [3]. Therefore, data from a statistical study showed that 4000 tons of Libs waste could contain 1100 tons of heavy metals and more than 200 tons of electrolyte constituents, which can pollute the atmosphere [6]. Therefore, improper management of used Li-ion batteries can pose a threat to the environment and public health through pollution of nature, contamination of groundwater, etc. [13].

Then, used Li-ion batteries have an added economic value due to the significant amounts of precious metals (Co, Li, Ni, Mn, Al, etc.) they contain. In addition, the critical state of their natural ores, the cost and environmental impacts of their extraction process, etc., motivate the revaluation of these technologies. Therefore, the development of an environmentally friendly and cost-effective method of processing these wastes has become a major challenge for the sustainable development of portable electronics. Cost-effective and environmentally friendly management of Libs waste, has two major advantages that are the protection of nature and public health and the development of secondary sources of Libs design raw materials. According to the literature, many processes for Li-ion battery waste recovery are developed and classified into two main families such as pyrometallurgy and hydrometallurgy [13] [14] [15]. The pyrometallurgical

technique is more generally used on an industrial scale, and allows the efficient recovery of certain metals such as Ni, Co, Cu, etc. However, this method does not allow the recovery of metals such as Al and Li, which are generally found in slag [16]. Moreover, this technique consumes large amounts of energy and leads to the release of hydrogen fluoride (HF) which can pollute the atmosphere. Hydrometallurgical processes, on the other hand, are more studied in the laboratory and are more advantageous due to their low energy consumption, reduced environmental footprint, selectivity and, above all, the possibility of recovering all the design metals from a given Libs. Advances in improvement studies have led to industrial scale with the development of relevant processes [16] [17].

The hydrometallurgical treatment is quite simple and consists of a series of highly selective processes that can be divided into three main parts: leaching, extraction and purification [13] [14] [15] [16]. The hydrometallurgical treatment is usually preceded by a physical pretreatment of the system, which has as objective the separation of the electrode components and the separator for more selectivity with a lower solvent consumption.

2. Li-Ion Batteries

A Li-ion battery is a reversible electrochemical system which is able to store energy in chemical form that it restitutes in electrical energy during its operation. It is composed of two electrodes, negative and positive, insulated by an electrically insulating polymer and interconnected by a liquid electrolyte. All these components of the battery are kept in a steel and/or plastic case (Figure 1) [18].

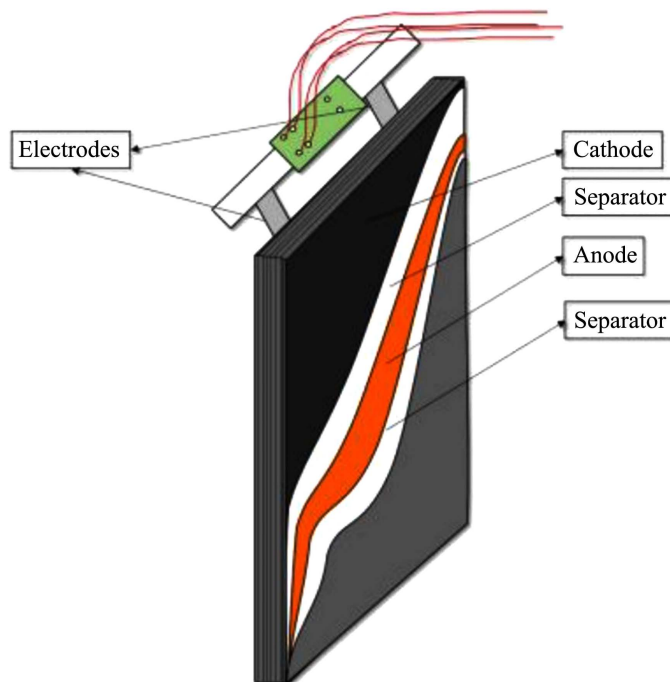


Figure 1. Structure of a Li-ion battery.

Most commercial Libs consist of graphite at the anode and a cathode generally composed of a layer of metal oxide and lithium (LiNiO_2 , LiCoO_2 , LiMn_2O_4) or iron phosphate and lithium (LiFePO_4), deposited on an aluminum foil using an adhesive binder such as PVDF [19]. These are interconnected by an electrolyte solution consisting of a binary or ternary mixture of organic solvents, typically dimethyl carbonate (DMC), ethylene carbonate (EC), diethyl carbonate (DEC), in which a lithium salt such as LiPF_6 , LiBF_4 or LiClO_4 is dissolved [10] [20]. In the design of a Lib, the cathode has a composition, often mixed, in relatively high concentrations of heavy metals and varying according to the manufacturer [21]. The cathode metal composition sometimes defines the name of the battery and is responsible for its performance (energy density, working potential, etc.). The electrolyte, which ensures the transfer of Li^+ cations from one electrode to another, plays an indispensable role in the safety of the system, facilitates operation and improves the life of the battery by its stability with the electrode components.

Despite the good use of Li-ion batteries, their heavy metal composition and the polluting nature of some of their electrolyte constituents, as well as the improper treatment of used batteries, can constitute sources of danger for public health and nature. Therefore, a cost-effective management of Li-ion battery waste, can allow the newly use of the design metals as a secondary source of metals. Proper management of Li-ion battery waste can play a vital role in the sustainable development of the electronics industry and avoid the depletion of mineral resources. Furthermore, most commercial electrolytes are composed of harmful, flammable and highly volatile components, so the disposal of used Libs or their open incineration can lead to adverse consequences [11] [22]. In view of all these threats, it is imperative for sustainable development of the electronics industry to develop a safe and economical method for the treatment of waste Libs.

With the accelerated upgrading of consumer electronics and their positive impact on our daily lives, the production of used batteries is becoming problem. In addition, considering the various threats related to the improper treatment of Libs waste, the development of a perfect management technique occupies an important place in the current research [13] [16] [23] [24].

At the present state of research, many processes, consisting of several steps, aimed at the recovery of Libs waste have been developed in various laboratories [16] [24].

3. Physical Pre-Treatment

The sample pre-treatment phase includes all the processes such as sorting, unloading, removal of crusts and heat treatment of electrodes [6] [25]. However, the most important steps are unloading, dismantling and heat treatment. After sampling, the batteries are emptied of their residual charge by means of an adapted external circuit or ionic solution such as NaCl to avoid any risk of fire or explosion. The latter are dismantled by cutting the cover and the electrodes

and electronic insulation are recovered and then separated. At this level, it is often used alcoholic solvent to recover the electrolyte before heat treating the separated electrodes. The purpose of the unloading is to reduce the risks of ignition and the extraction of the electrolyte could improve the polluting character of the process and improve the efficiency of the process

4. Hydrometallurgical Treatment

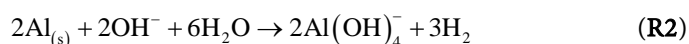
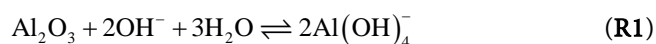
Hydrometallurgy is a method of chemical treatment which aims to recover the metals present in a given sample. The principle consists in using appropriate solvents to solubilize the metallic constituents. The general procedure is subdivided into three main parts which are leaching, liquid-liquid extraction and the purification or revalorisation phase [26].

4.1. Leaching

It consists of attacking a metal or a metallic constituent by oxidizing or reducing in the presence of strong ligands capable of forming soluble and stable complexes [3] [26]-[32]. After a bibliographical study of the Libs, two types of leaching such as oxidative or alkaline leaching and reductive or acid leaching, are studied in the treatment processes by hydrometallurgy [26] [28] [33].

4.1.1. Alkaline Leaching

The selective leaching of Libs cathodes aims at attacking the aluminium metal of the cathode collector in an alkaline medium in order to solubilize it [3] [26] [27] [28]. The originality of this step is to selectively recover the aluminum from the cathodic electrical connector and possibly other metals such as iron, copper, etc. qualified as impurities to the liquid-liquid extraction [34]. Since aluminum is generally in metallic form, the solubilization of the electronic connector can be explained by an oxidation to the Al^{3+} cation and then a complexation of the present cations. Therefore, the choice of the leachant can be based on its oxidizing character and/or the coordination strength of its constituents. It is in this context that Ferreira *et al.* (2009) tested soda ash in a process of selective solubilization of used Libs cathodes [26]. The aim of the work was to optimize operational variables such as concentration, contact time of the reactants, L/S ratio and reaction temperature. As a result, they showed that nearly 60% of the aluminum in the connector can be solubilized when the concentration is increased to about 4 M with an L/S ratio of 10 L/Kg for one hour at a temperature of about 60°C. Results from this work showed that the solubilization of aluminum metal is explained by its oxidation in the presence of oxygen from the air and then by the coordination of Al^{3+} cations by OH^- ligands in the medium (R1 and R2) [26].



Gaye *et al.* (2019) resumed the study at room temperature in an effort to im-

prove the fitting conditions [27]. In doing so, they evaluated and optimized the variables concentration, count time, and L/S ratio. Results of this work proved respective solubilization rates of about 55% and 10% for aluminum and lithium when the study parameters were set at 4 N, L/S = 10 L/Kg for 5 h.

Other solubilization tests have been performed with soda ash and results have confirmed its good performance as a leachant [33] [34] [35] [36], which may favor its choice as a reference solvent in the selective leaching of Libs cathodes.

However, it is noted that soda solutions do not completely solubilize cathodic aluminum, which can be explained by passivation and/or precipitation of aluminum oxide in basic media [37]. Data from the literature have shown that the precipitation of aluminium oxide is favoured by the presence of CO₂, the level of OH⁻ ions, temperature, time, etc. [38] [39] [40]. In an effort to improve the design aluminum recovery rates of Libs cathodes, researchers have investigated and tested other alkaline solvents [3] [41]. As a result, Nayl *et al.* (2017) studied the selective leaching of cathodic and anodic Libs materials with ammonia [3]. This work allowed them to access recoveries of about 98% for Al and 65% for Cu when the experimental conditions were set to NH₄OH 4 N with L/S = 15 mL/g for one hour under 60°C. This solubilization efficiency of Al metal can be explained by the coordination strength of the NH₃ ligand. The smaller recoveries for copper may be due to the redox potential of the Cu²⁺/Cu couple being closer to that of the oxidant, which may make the couples reactive.

4.1.2. Acid Leaching

All the metals present in Li-ion batteries are solubilized by acids. Thus, acidic solvents are used to solubilize the metals Co, Ni, Mn, Li, Al, etc. used in cathode compositions. Until today, many studies have been conducted and different organic and inorganic acids have been tested as acid leachers of Libs cathodes [13] [15] [42]. Inorganic acids such as H₂SO₄ [3] [25] [26] [36] [43] [44], HNO₃ [45], HCl [46] [47] [48] [49] [50], and H₃PO₄ [51] [52] have been widely studied and have shown great promise with solubilization rates of Li, Co, Mn, and Ni metals above 90%. Joulié *et al.* (2014) conducted a comparative study of the performance of HCl, H₂SO₄, and HNO₃ acids in a process to solubilize Li, Ni, Co, and Al metals from Libs cathodes [47]. Results have shown that HCl presents the best performances compared to H₂SO₄ and HNO₃, which can be explained by the presence of Cl⁻, very strong reductant towards Co(III) and Ni(III) cations (R3) contrary to NO₃⁻ and SO₄²⁻ anions, that have weak reducing powers.



Despite the good results with HCl, the reduction of metal cations leads to the production and release of chlorine (Cl₂), which is very harmful to humans [53]. Since most of the metals (Mn, Co and Ni) of Libs design are more soluble with their reduced form, reducing agents are therefore studied and tested in order to improve the performance of some acids [53] [54].

According to the literature, hydrogen peroxide (H₂O₂) is widely tested with many acids in the leaching of Co, Ni and Mn metals, which are generally used in

commercial Libs cathodes [3] [27] [33] [43] [55] [56]. Despite the good performance of inorganic acids associated with reducing agents, most of them, lead to the release of gaseous compounds of a polluting nature. Therefore, research is focused on other types of acids in order to improve the pollutant character of waste Libs treatment.

In this respect, many acids such as citric acid [57] [58] [59], oxalic acid [36] [60], ascorbic acid [61], DL-malic acid [62] [63], succinic acid [64], L-aspartic acid [55], L-tartaric acid [65], acetic acid [66], iminodiacetic acid [67] have been tested as cathode leachers of Libs. These types of acids have the advantage of being more environmentally friendly and are capable of solubilizing, at high levels, the design metals of Libs cathodes. Similarly, organic compounds including glucose [29] [68], ethanol [69], etc. are being studied and tested with organic acids as a reducing agent [54] [70] [71] [72]. The use of organic acids has made a significant contribution to safety in the management of waste electrical and electronic equipment in general, and of Libs waste in particular [39]. However, the use of these organic solvents often requires high concentrations, which can have economic consequences [73]. Thus, research continues in various laboratories with the objective of finding a cheap, ecological acid capable of completely solubilizing the design metals of Li-ion batteries.

4.2. Liquid - Liquid Extraction

In hydrometallurgy, solvent extraction is a process of transferring soluble metals from the aqueous phase to the organic phase by means of an organic solvent. The principle is based on the use, in appropriate proportions, of organic solvent that is weakly miscible with the aqueous phase. When the final mixture is well stirred and the metals are loaded into the organic phase, a distribution equilibrium is established between the organic and aqueous phases, characterized by a partition coefficient (D) defined as follows [9] [10] [74] [75] [76]:

$$D = \frac{C_1V_1 - C_2V_2}{C_2V_2} \quad (1)$$

The extraction efficiency of the given solvent with respect to a metal is characterized by its extraction rate ($\% E$), which is determined from Equation (1) below [33] [74] [77]:

$$\% E = \frac{C_1V_1 - C_2V_2}{C_1V_1} * 100 \quad (2)$$

In the case of a mixture of several metals, the affinity of an extractant towards the metals or the extraction selectivity of a metal in a mixture is determined according to the separation factor of these metals. This separation factor of two metals for a given solvent is determined by Equation (3) below [76].

$$\beta = \frac{D_1}{D_2} \quad (3)$$

With D the partition coefficient, $\% E$ the extraction rate, C_1 and C_2 the initial

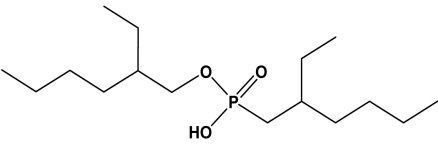
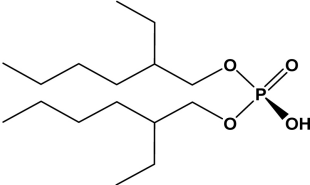
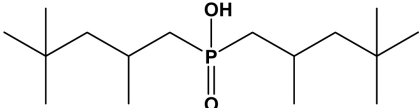
and final concentrations of a given metal in the aqueous phase, V_1 and V_2 the respective volumes of the aqueous phase before and after extraction, D_1 , D_2 the distribution factors of the metals M_1 and M_2 , and β the separation factor of the metal M_1 from M_2 .

From a literature review, we found that many organic solvents are studied and tested in the laboratory. Organophosphorus acids such as Cyanex 272, D2EHPA and PC 88A have shown very promising performances with high extraction rates for the recovery of Co, Mn, Ni with interesting selectivity [7] [33] [77] [78] [79] [80] [81]. **Table 1** below gives a presentation of chemical formulas of some well known organophosphorus solvents in the field of liquid-liquid extraction of Libs metals.

The characteristic chemical molecules of these solvents have the particularity of being able to form stable complexes that are completely insoluble in the aqueous phase, hence their good performance.

Cyanex 272 has been tested several times, for the extraction of precious metals, in the hydrometallurgical treatment of Libs wastes and the results showed an interesting affinity towards metals (Co, Mn, Ni, etc.) of cathodic design [25] [33] [35] [44] [79] [81] [82] [83]. In a process for the recovery of Co, Li, Cu and Al metals, Jha *et al.* (2013) conducted optimization tests of the operational variables that impact the performance of Co extraction by Cyanex 272 [83]. As a result, they obtained after 5 min, extraction rates of about 99.9% Co when the experimental conditions were increased to 15% Cyanex 272 and 3% isodecanol in kerosene and the O/A ratio was set to 1:1 (by volume) at pH equals 5. Vasilyev *et al.* (2019), conducted an optimization study of a process for extraction of Co, Ni, and Li metals by NH_4 -Cyanex 272 from Li-ion battery leachates [79]. Other optimization work has shown that an extraction solution consisting of 1 M Cyanex 272 neutralized to 48% with ammonia and 5% TOA, can achieve nearly 99.9%

Table 1. Structure and properties of widely used extractants.

Nom	Structure moléculaire	Identifiant	Réf.
PC-88A		$\text{C}_{16}\text{H}_{35}\text{O}_3\text{P}$ $M = 306.42 \text{ g}\cdot\text{mol}^{-1}$	[3]
D2EHPA		$\text{C}_{14}\text{H}_{31}\text{O}_4\text{P}$ $M = 294.3673 \text{ g}\cdot\text{mol}^{-1}$	[5] [7]
Cyanex 272		$\text{C}_{16}\text{H}_{35}\text{O}_2\text{P}$ $M = 290.42 \text{ g}\cdot\text{mol}^{-1}$	[7] [10]

extraction of these metals with high purity rates ($\leq 99\%$) for an A/O ratio equal to 1:1 for 5 min [81].

Nayl *et al.* (2015) also employed Cyanex 272 to recover Mn(II), Co(II), and Ni(II) cations from spent Libs mixed cathode leachates [32]. Beforehand, Cu(II), Fe(II) and Al(III), considered as impurities, are removed with 20% Acorga M5640 diluted with kerosene after 5 minutes at 30°C. When Na-Cyanex 272 is applied after 5 minutes under 30°C, respective extraction rates of about 91.2%, 89.3%, 95.6% are obtained for Mn(II), Co(II) and Ni(II) cations at pH 3.5, 5 and 8 respectively [32].

D2EHPA has been tested as an extractant in cathode treatment processes of waste Libs [5] [60] [78] [84]-[89]. For example, Yang *et al.* (2017) tested the extraction and separation of Co, Mn, Ni, and Li metals from Libs leachates [89]. After optimization tests of pH, O/A ratio, contact time, and D2EHPA concentration, extraction efficiencies up to about 100% for Mn, 99% for Co, and 85% for Ni were achieved with 40% D2EHPA, O/A ratio = 1, at pH 3.5, for 6 minutes [89]. They thus showed that the extractant has a selective affinity towards these metals (with Mn > Co > Ni > Li) and that this selectivity depends on pH. In addition, Li *et al.*, (2018) developed a simple and effective method for manganese recovery from Libs leachates [74]. They investigated the optimization of pH, O/A ratio, contact time, and saponification rate of the extractant, and results proved high recovery rates for Mn [74]. This confirms the selective affinity of the extractant towards Mn.

Another organophosphorus such as 2-ethylhexyl phosphonic acid-mono-2-ethylhexyl ester (PC88A) has been studied and tested as an extractant of metal cations from Libs cathode leachates [85] [86] [87] [88] [89]. Yang *et al.* (2020) conducted an optimization study of the variables pH, O/A ratio, and concentration of PC88A in kerosene in a selective recovery study of Mn²⁺, Co²⁺, Ni²⁺, and Li⁺ cations from sulfuric leachate [89]. An extraction efficiency of 98% for Mn and 90% for Co can be obtained when the experimental conditions are set at pH = 5, O/A ratio = 2/1 for 10 minutes. Most importantly, they obtained high separation factors of Co and Mn compared to Li and Ni, found in the raffinate. These results prove an interesting extraction selectivity depending on the reaction pH value.

These works show the good performances of these extractants with selective affinities towards the metals Co, Mn, Ni and Li; D2EHPA has more affinity with Mn while Cyanex 272 and PC-88A have comparative affinities towards Co, Mn [90] [91]. These results prompted the study of their synergistic action in combinations such as Cyanex 272/PC-88A [75] [90] [92], Cyanex 272/D2EHPA [7], Cyanex 272/Cyanex 302/D2EHPA [91], Cyanex 272/Cyanex 301/D2EHPA [93], Cyanex 272/PC-88A/D2EHPA [94] [95]. Thus, results have shown that synergistic systems often have higher recovery rates with better separation for mixed leachates.

This is the case with Sarangi *et al.* (1999) who showed that the binary mixture Na-PC88A (0.005 M) and Na-Cyanex 272 (0.025 M) presents, under the same

conditions, a better separation of Co(II) and Ni(II) from a hydrochloric acid leachate compared to the solvents Na-Cyanex, Na-PC88A, Na-D2EHPA used alone, as well as the binary mixtures of these three solvents [94].

Despite the important performance of organophosphorus solvents, the search for an extractant system that can recover cathodic design metals in a single step and with different affinities towards these metals remains a challenge. It is in this context that the family of ionic liquids is being explored and very promising studies are underway [96] [97].

4.3. Electrodeposition

Electrodeposition is an electrochemical technique for metal cations recovery or removal from leachate or effluents. The principle is based on the application of a direct current between positive and negative electrodes immersed in the electrolyte solution. When an appropriate electrodeposition potential is applied, the targeted metal cation(s) is reduced (to its metallic or metal oxide form) and deposited on the cathode. The deposit is then solubilized in an acid medium (reduced pH) for characterization and purification tests [98]. This technique is widely applied in water treatment (removal of heavy metals), industrial effluent treatment, pyrometallurgical treatment of WEEE, etc. [15] [73]. According to the literature, electrodeposition is explored by various research teams and in various laboratories during cathode treatment of used Libs [99] [100] [101] [102] [103]. Thus, it has been shown that the deposition of a given metal depends on the working potential, the nature of the electrolyte, the temperature, the pH of the medium, etc. [99] [100] [101].

Tanong *et al.* (2017) studied the recovery of Zn(II), Mn(II), Cd(II), and Ni(II) cations from H₂SO₄ leachates by electrodeposition [104]. The metal cations in the leachates are selectively extracted by Cyanex 272 and D2EHPA, then stripped with H₂SO₄ solution and the electrolytes are tested for electrodeposition. When an energy density of about 370 A·m⁻² is applied at pH = 2, metallic layer deposits appear on the cathodes (made of steel) in all experiments. These deposits are subsequently solubilized by a 5% HNO₃ solution and subjected to characterization and precipitation tests.

Kim *et al.* (2021) studied the selective recovery of cobalt and nickel in a hydrometallurgical treatment process of Li-ion battery cathodes [98]. This work demonstrates the role played by the electrolyte on the efficiency and selectivity of metal deposition. In addition to the pH of the medium and the working potential, the use of LiCl as an electrolyte promotes the selective deposition of cobalt through the formation of the [CoCl₄]²⁻ complex by stabilizing the cationic complex [Ni(H₂O)₅Cl]⁺ [98]. Data from this experiment showed high recoveries with purities in the range of 96.4% and 94.1% for cobalt and nickel respectively.

Thus, electrodeposition is an advantageous purification process in that it reduces solvent consumption and limits the production of effluents and polluting gases [99] [100] [101]. However, this method has technical limitations in the treatment of mixtures of metals with very close redox potential [98] [104].

4.4. Chemical Precipitation

Precipitation is a process of forming insoluble end products from two or more reagents under specific conditions. After extraction or electrodeposition and subsequent stripping, the new aqueous phase is mixed with a precipitating reagent in precise proportions to precipitate metal hydroxides, sulfides or carbonates. Data from the literature have shown that this precipitation depends on many parameters, the most influential of which are the pH, contact time, nature and concentration of the reagents, temperature, etc. [33] [48] [84] [103]-[108].

Nayl *et al.* (2017) evaluated the impact of pH on the performance of selective precipitation of Co(II), Mn(II), Ni(II) and Li(I) cations from H₂SO₄ leachate [3]. When they employed a sodium carbonate solution, the carbonates MnCO₃, NiCO₃, CoCO₃, and Li₂CO₃ precipitated selectively when the pH was adjusted to 7.5; 9; 11 and 12, respectively. This pH-dependent selectivity of metal precipitation is confirmed by the work of Meshram *et al.* (2015) [109] and Natarajan *et al.* (2018) [110]. Furthermore, it is shown that this metal precipitation is also dependent on the reagent employed. Barik *et al.* (2017) demonstrated that under specific conditions, a NaClO solution results in the selective precipitation of a manganese oxide mixture from a leachate containing Mn(II), Co(II), Ni(II), Cu(II), Al(III) and Li(I) [48]. Cobalt and lithium precipitated using sodium carbonate solution while copper and aluminum precipitated using soda ash [48].

5. Conclusions

In the face of the rapid production of waste Libs and the growing demand in the portable electronics market, the economic and proper management of used Libs has become a major political and economic issue. These types of waste, if improperly treated, become a threat to public health and nature. In addition, the critical state of the ores and the economic value of the design metals make recycling a major asset for the sustainable development of consumer electronics. Nowadays, many hydrometallurgical treatment processes of Libs scrap are developed and promising results are obtained from literature data. After bibliographic synthesis, we retain that in hydrometallurgy;

Alkaline leaching can facilitate the acid leaching and liquid-liquid extraction steps by eliminating impurities such as Al, Cu, Fe, etc. and reduce the solvent consumption and minimize the number of steps. However, the solvents tested in this field lack efficiency, which calls for further studies.

Acid leaching, which is the essential step, consists in solubilizing all the metallic constituents of a given sample. Most of the inorganic acids tested release gases that can pollute the atmosphere and pose a threat to public health, while organic acids are less polluting but not too effective.

Liquid-liquid extraction and electroplating are complementary and are often confronted with a lack of selectivity in the treatment of mixed batteries. In addition, extraction can lead to the consumption of large quantities of solvent and the production of effluents.

For future studies, it would be interesting to test the use of specific oxidants of very strong complexing agents in order to improve the performance of selective leaching and the use of ionic liquids as extractants for solubilized metals.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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