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Quantification of Recoverable Components of Spent Lithium-Ion Batteries

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ABSTRACT

Recovering spent lithium-ion batteries can help protect the environment and generate added value. The aim of this work is to characterize the various parts of these spent lithium-ion batteries for subsequent recovery of the precious metal elements. The batteries were collected, electrically discharged and dismantled, and the various components quantified. The cathode powder obtained after basic leaching was characterized by ICP and XRD. The batteries consist of steel (21.10%) and plastic shells, the anode (24.40%), the electrolyte-soaked separator and the cathode (35.86%). The anode consists of graphite deposited on a copper foil representing 15.15% of its weight, and the cathode of aluminum foil (3.93%) and lithium cobalt oxide. Physico-chemical characterization of the cathode powder yielded CoO (65.30%), Li_2O (5.39%), MnO (15.78%) and NiO (2.17%). At the end of this study, we note the presence of precious metals, on which our subsequent recovery work will focus.

Keywords: Lithium-ion battery, Anode, Cathode, Metals, Environment.

INTRODUCTION

The development of the electronics industry to improve the living conditions of man on earth. This development generates the waste known as waste electrical and electronic equipment (WEEE) which is a global problem[1]. In 2019, the world generated an impressive 53.6 Mt of electronic waste, an average of 7.3 kg per capita. This generated waste contains toxic or hazardous materials but also rare or precious metals[2]. The regions most affected by WEEE exports are Southeast Asia and West Africa. Togo, like the African countries, is affected by its second-hand WEEE products which end up in garbage cans, on landfills and sometimes even incinerated. Since there is no appropriate sector dedicated to the management of WEEE waste. Batteries, printed circuit boards (PCBs), liquid crystal displays (LCDs), cathode ray tubes (CRTs), hard disk drives (HDDs), refrigerators and mobile phones are an integral part of a typical WEEE[3].

Lithium-ion batteries (LIB), one of the electrical and electronic equipment, has been growing rapidly due to the prevailing implementation of new technologies in electronics, consumer attractive designs and demands of life daily. These batteries are widely used in mobile phones, laptops, video cameras due to their characteristics of light weight, high energy and good performance[4,5]. The lifetime of these batteries generally varies from 2 to 4 years[6,7]. The strong growth in LIB production due to the proliferation of portable equipment generates huge quantities of spent lithium-ion batteries, the management of which constitutes an environmental problem.

Global demand for metals is increasing. However, the exploitable reserve, which is only distributed in a few countries, is very limited and is running out[8]. In other words, the mismanagement of LIB leads to a waste of metal resources and environmental risks[9].

If spent LIBs are simply disposed of, a serious environmental problem would be caused by the leakage of organic electrolytes as well as a risk of explosion or fire due to the lithium it contains.

A lithium-ion battery consists of a cathode, a separator and an anode, all enclosed in a steel or plastic casing. The cathode has been made of lithium cobalt oxide ($LiCoO_2$), since its first synthesis by Goodenough in 1980[10] and its first commercialization by Sony in 1991[11]. Subsequently, various Li-containing materials, such as $LiFePO_4$[12], $LiMn_2O_4$[13], and $LiNixMnyCozO_2$[14], were developed and widely applied. However, $LiCoO_2$ is often used in mobile phone batteries due to its competitiveness and energy density[15]. The anode is a thin sheet of copper coated with graphite.

When lithium-ion battery waste is properly treated, precious metals such as cobalt and lithium can be recovered. From the point of view of preserving the environment and recovering valuable resources, the recycling of spent lithium-ion batteries is highly desirable. The current state of the recycling process has been examined in several studies[16,17,18].

The precious metal elements (Co, Li, Mn, Ni, Al, Cu, etc.) contained in the battery electrodes are essential. Lithium is an indispensable element in the manufacture of electrode materials for batteries and in other areas such as glass ceramics, enamels, adhesives, lubricating greases, metal alloys, air conditioning and dyeing[19]. Cobalt powders have been used in steel for cutting tools, in composites reinforced by abrasion, and in alkaline rechargeable batteries[20]. Additionally, recycling LIB batteries preserves primary resources for the future by recovering the precious metals associated with LIB batteries. In addition, LIB recycling is attractive for commercial production because it is free of siliceous matrix and its high concentration of metal ions compared to primary sources. It is therefore very important to recover these metallic elements for economic, health and environmental reasons.

It is in this logic that for decades, research on the recovery of precious metallic elements contained in LIB waste has been carried out. The main recycling technologies used are pyrometallurgy, biometallurgy and hydrometallurgy.

This work was undertaken on spent lithium-ion batteries from mobile phones. This involves collecting, discharging and dismantling spent lithium-ion batteries in order to recover and quantify the various components of the battery. Then the cathodic powder obtained after basic leaching with soda (NaOH) was characterized by ICP and XRD, thus revealing the chemical composition and the different phases present, with their content.

METHODOLOGY

Collection and sorting of spent batteries

Spent Lithium-ion cell phone batteries were collected from second-hand cell phone dealers in the Lomé port area and from repairers. The potential difference of all the batteries used in this study was 3.7 V. The batteries collected were of different brands, wattages, sizes and origins. They were sorted and grouped according to brand and power. They include brands such as Samsung, HTC, SonyEricson, Nokia.

Fig. 1. Spent lithium-ion batteries

Dismantling of lithium-ion batteries

For safety reasons, prior to disassembly, the spent lithium-ion batteries were soaked in a 10% NaCl solution for 24 h to discharge them, in order to avoid any danger of short-circuiting or auto-ignition.

The batteries were then dismantled manually, using pliers and saws. The steel casings were cut and removed. The internal components were unrolled, separating the anodes, cathodes and plastic separators.

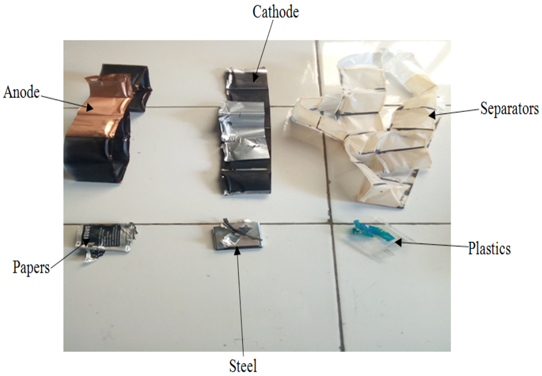

Fig. 2. Components of the spent lithium-ion battery

Quantification of the different components recovered from the lithium-ion batteries

The mass of each component (steel, paper, plastic, electrodes, etc.) of spent lithium-ion batteries was determined using a Nimbus brand balance.

Fig. 3. Nimbus Scale

The content of the various components obtained was evaluated using the formula:

$$\text{Percentage of components} = m_c/m_b \text{x}100 \quad (1)$$

m_c: mass of the component recovered from the battery
m_b: mass of the lithium-ion battery

Separation of copper at the anode

Once the anode had been dismantled, it was immersed in demineralized water in a beaker and stirred for 2 hours. The graphite detached from the copper foil, allowing recovery of the copper in its metallic form, which was dried and weighed. The mass thus obtained was used to calculate the copper content of the anode using the following formula:

$$\text{Copper percentage} = m_{cu}/m_a \text{x}100 \quad (2)$$

m_{cu}: mass of copper obtained
m_a: mass of the anode

Preparation and characterization of cathode powder

The cathode was treated with a known volume of 1M NaOH solution (1:5) for 2 h with magnetic stirring to dissolve the aluminum foil which serves as an electron collector. The cathode material separated from the aluminum was recovered after filtration and washing to constant pH, in order to eliminate the residual sodium hydroxide before being dried in an oven at 105°C for 24 h and ground to 75 μm. Then the powder is kept for leaching studies and analyses.

Aqua regia (HCl: HNO_3, ratio 3:1) was used for the complete digestion of the LIB cathode powder before the quantification of the metal contents.

The determination of the metallic elements contained in the cathode powder was carried out using an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES), brand (Thermo Fisher X Series II, Thermo Fisher Scientific , Bremen, Germany) after digestion with Lugol water (HNO_3: HCl=1:3, v/v).

Crystal phases of the raw cathode were determined by X-ray diffraction, using a Rigaku Miniflex 300 instrument operating in the 2θ 5-98° range, with CuKα radiation (λ= 0.15418 nm), a voltage of 45kv, and a current of 40 mA.

RESULTS AND DISCUSSION

Quantification of the different parts of the lithium-ion battery

The results of the quantification of the various parts of the lithium-ion batteries studied are recorded in Table 1. The average mass of the batteries studied is 25.5 g. These results reveal that the cathode and the anode represent an important part of the lithium-ion battery since they concentrate more than half (60%) of the mass of the batteries. This is in agreement with the work of[21,22]. These two components constitute the active part of lithium-ion batteries and contain recoverable metallic elements (aluminum, copper, cobalt, nickel, lithium and manganese, etc.). The steel boxes, representing 21% of the batteries by mass, protect the active components of the batteries. Other materials (plastics, paper and separators) account for 11% of battery mass[23].

Table 1: Proportion of the different components of the batteries studied

Battery weight (g)	%Anode	%Cathode	%Steel	%Others
25,49±7,41	24,40±7,84	35,86±7,08	21,10±1,36	11,67±3,31

The steel (21% of the battery) and the plastic material obtained following dismantling can directly return to the recycling circuit. The anode and the cathode can undergo treatments with a view to recovering the metallic elements which they contain.[24,25]

Pre-separation of battery components would be very cost effective for recycling, since the steel case, paper, plastic, separator, aluminum and copper foils, solvent are all directly recyclable after separation[26].

Characterization of the electrodes

Table 2, shown below, summarizes the results of the contents of metallic elements (Cu and Al) and of the cathodic and anodic materials. The anode, having an average mass of 6.22 g, consists of a copper film whose proportion is evaluated at 15.15%. The cathode with an average mass of 9.14 g consists of an aluminum film (3.93%). This aluminum film is connected to an active cathodic powder thanks to a binder, the whole of which has a content of 96.07% and contains metal oxides. These results differ from those found by[27,28,23], who respectively found for copper a content of 9%; 5.5% for aluminum and 8.84% for copper and aluminum together. As pointed out Georgi-Maschler *et al.,* (2012)[28], battery producers produce their own specific types of LIBs, so it is difficult to provide precise general values. It is therefore difficult to provide precise general values for the masses of the electrode components of a lithium-ion battery, since the composition varies depending on the manufacturing process[29].

Table 2: Proportion of copper and aluminum in lithium-ion battery electrodes

Anode	% Components	Cathode	%Components
Copper	15,15	aluminum	3,93
Graphite+binders	84,85	Cathodic powder+binders	96,07

Copper and aluminum can be recovered in the metallic state by hydrometallurgical or pyrometallurgical and biometallurgical means.

Characterization of the cathode powder

Elementary chemical analysis at the ICP and mineralogical analysis by X-ray diffraction made it possible to characterize the cathode powder. The results of its analyzes are recorded in Table 3 and Fig. 4. The ICP analysis showed that the cathodes of the batteries studied are essentially made up of Cobalt (CoO, 65.30%), Manganese (MnO 15. 78%), lithium (Li2O 5.39%) and traces of nickel (NiO 2.17%). These results show that the cathode powder is rich in cobalt oxide, and a minor phase of manganese oxide, which is used in some cathodes to reduce battery production costs, since cobalt is expensive[30,31,32,33,34,35,36]. Consequently, given the high content of Mn in the sample, we will think about its extraction, as well as for Co and Li.The nickel content is thought to be due to doping and surface modification used to increase battery capacity.[37,38]

Table 3: Content of metallic ingredients in the cathode of Lib

	%CoO	%MnO	%Li_2O	%NiO
Cathode	65,30 ±1,09	15,78±1,15	5,39±0,42	2,17±0,50

The X-ray diffraction analysis, carried out to identify the phases present in the cathode powder, showed the presence of three crystalline phases. Lithium cobalt oxide $LiCoO_2$ (18.9°, 37.32°, 39°, 45°, 49°, 59°, 65°, 69°) constitutes the major phase of the material with a proportion of 68.4%. The manganese and lithium oxide $LiMn_2O_4$ (36.32°, 44°, 58.29°, 63.99°) is also present with a proportion of 31.6%.

We note a third minority phase whose peak is detectable at 2θ=26.6° corresponds to graphite.

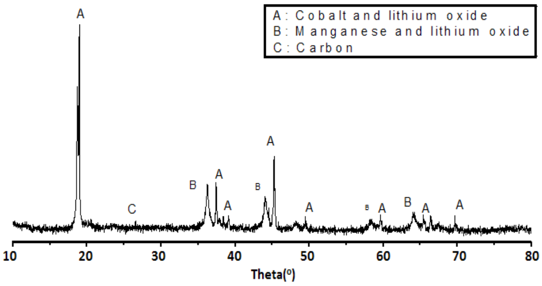

Fig. 4. X-ray diffraction analysis of the cathode electrode of the lithium-ion battery

The presence of this third phase shows that the cathode contains some graphite.[37,39,40]

The cathode powder obtained is a mixture of cathodes from batteries of different brands and origins. It is therefore possible to have cathodes composed of $LiCoO_2$, of $LiMn_2O_4$ or a combination of the two oxides. But we can remember that the cathodes are mainly composed of $LiCoO_2$.

The presence of nickel in the cathode was not detected by X-ray diffraction, although chemical analysis revealed a NiO content of 2%. This can be explained by the fact that nickel was used to dope $LiCoO_2$ and $LiMnO_2$.[37,38].

From the cathodes of spent lithium-ion batteries of mobile phones, lithium, cobalt, manganese and nickel can be recovered. The recovery of its useful metals from spent lithium-ion batteries is an asset for the environment since the extraction of these metals has enormous impacts on the environment.

To recover these metals from the cathode, the techniques of leaching and sequential precipitation[41,42,43], electrolysis[44,45,46] or solvent extraction.[47,48,49]

CONCLUSION

At the end of this work, we note that spent lithium-ion batteries contain recoverable components. The characterization of these batteries shows that steel, plastic, aluminum and copper can be recovered using simple techniques. The constitution of cathode powder, the active material of lithium-ion batteries, reveals that it consists of lithium and cobalt oxide, lithium manganese oxide and nickel oxide. These metals can be recovered after leaching by precipitation, electrolysis or solvent extraction.

In order to minimize the environmental impacts of the recovery of these useful metals, it is important to develop environmentally friendly leaching and recovery techniques.

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Conflict of interest

The authors declare that there is no conflictof interest to be reported.

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