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Waste management of discarded cell phones and proposal of material recovery techniques

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Abstract

The treatment of end-of-Life products is one of the key steps for progression towards a circular economy. In this context, mobile phone recycling has attracted considerable attention in recent years due to the high metal concentrations in printed circuit assemblies (PCAs). Contrary to precious metals, the recycling technologies for tantalum and rare earths are less well established despite their high importance for future sustainable technologies. In this study, mobile phone samples were manually dismantled into their main constituents and the qualitative distribution of elements in the PCA was determined. Tantalum capacitors can be visually distinguished from other components, whereas neodymium was detected in easily separable non-magnetic components sized between 0.5 and 1.5 mm. From tantalum capacitors, a tantalum rich-powder with a grade of 50% was obtained via a leaching process followed by an oxidizing heat treatment, while neodymium-rich fractions (4 to 14%) were obtained by sieving, magnetic and density separations.

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1. Introduction

Waste from electrical and electronic equipment (WEEE) is one of the fastest growing waste streams in the world. This trend resulted from technological advances and the rapid obsolescence of electronic devices. For example, the actual service life of mobile phones is less than 2 years and this short life time has resulted in about 700 million of these equipments that are discarded every year worldwide [1].

Traditional methods for managing solid wastes including landfills or incineration are inappropriate to treat electronic wastes, as they contain significant amounts of hazardous heavy metals and brominated flame retardants.

In response to this, regulations have been enacted in many regions of the world to ensure the proper collection and recycling of electric and electronic wastes [1]. The European WEEE directive, implemented by the European Union in

2002 and updated in 2012, is one of these initiatives. This directive classifies WEEE into 10 categories and establishes targets for recycling of each category. Mobile phones belong to category 3 and the minimum recycling target for this category is 70% during 2015-2018 [2].

Nomenclature

PCA	Printed Circuit Assembly
PCB	Printed Circuit Board
WEEE	Waste Electrical and Electronic Equipment
SEM	Scanning Electron Microscope
EDX (EDS)	Energy Dispersive X-ray Spectrometer
REEs	Rare Earth Elements
ICP	Inductively Coupled Plasma

A typical mobile phone device consists of several parts: the plastic frame, the battery, the printed circuit assembly (PCA), the liquid crystal display and the keyboard. The PCA is the most important part as it contains all the electronic components (transistors, diodes, integrated circuits, capacitors, resistors) required for the proper operation of the device. It accounts for about 20 – 30% of the total weight of the mobile phone depending on the phone model and manufacturing year and it is composed of two main components: the substrate (Printed Circuit Board-PCB) and the surface-mounted components [3, 4]. The metal content in the PCA is greater than that of some natural ores. Indeed, the PCA can contain more than 40 elements of the periodic table including base metals (Fe, Al, Cu, Ni, Sn etc.), precious metals (Au, Ag, Pt, Pd), strategic metals (Ta, Nd, Pr, Y etc.) and hazardous elements (As, Pb, Br, Cr) [4, 5]. Therefore, waste mobile phones can be considered as a secondary resource for the recovery of important metals and their recycling is required for both resource conservation and environmental protection.

Many studies have been conducted to recover copper and precious metals from waste mobile phones and advanced recycling processes for these metals already exist [1]. However, despite their high importance for future sustainable technologies and their relatively short life expectancies, the recycling rate of special metals such as rare earth elements (REEs) and tantalum from end-of-life products is currently less than 1% [6]. This poor recycling efficiency is probably due to the absence of a supply chain structure intended for the pre-processing of electronic wastes, with a particular focus on strategic metals [7].

Indeed, the dominant approach in most of the current recycling plants is to shred the entire electronic product leading to important losses of the components that contain these strategic metals [3, 7]. Owing to the complex structure of electronic equipments, manual dismantling seems to be the best option to remove these components. For example, Bachér et al. prove that the robust shredding of cell phones resulted to at least 50 % loss of these special metals compared to manual dismantling [3]. Since the total recycling efficiency depends on the effectiveness of each single step, an effective disassembly process is of great importance to enhance the recovery rates of these metals.

The present paper determines the location of strategic metals in a mobile phone device. This qualitative distribution of strategic metals is used to develop a process that ensures high concentrations of these metals. The paper is divided in two main sections. The first section summarizes the findings of the experimental study and the second section discusses the driving and limiting factors for the up-scaling of the recovery process.

2. Experiments and results

2.1. Disassembly

The study was realized with 5 identical mobile phones (Nokia 3310) that were obtained from stores specialized in the

repairing of mobile phones. Nokia 3310, launched in 2000, is one of the bestselling mobile phones of all time with 126 million units sold worldwide [8]. This mobile phone is representative of small regular phones from the production period 2000-2010. Given that less than 10% of spent mobile phones are currently returned for recycling [1], there is huge recycling potential for this generation of mobile phones. One sample was used to determine the qualitative distribution of elements in the printed circuit assembly and the rest was used for the material recovery process.

Mobile phones were manually dismantled with a Torx® screwdriver (T6) and the main constituting parts were weighted with an analytical balance. The average weight of the studied mobile phone is 81.23 g without taking into account the battery. Some small differences (0.3-5%) were noticed in the mass of the main constituting parts depending on the year of fabrication of the sample.

The largest component of the studied mobile phone is the plastic frame which is made of an ABS/PC blend. The display is made of glass and plastics and it may also contain indium. Two types of batteries were observed: Nickel-metal hydride battery (Ni-MH) and Lithium-ion (Li-Ion) battery. These types of batteries may contain nickel, cobalt, copper, lithium, lanthanum and cadmium [6]. The PCA accounts for only 22% of the total weight of the studied mobile phone but, as explained previously, it is the most interesting part to investigate from a recycling perspective. The recovery process in this study was thus focused on the PCA.

2.2. Qualitative distribution of elements in the PCA

2.2.1. Experiments

The qualitative distribution of elements in the PCA was performed with a scanning electron microscope (SEM) coupled to an energy dispersive X-ray spectrometer (EDX or EDS). One of the features of the equipment is the possibility to determine the global superficial composition of conductive samples over a surface of about 1 mm².

The analyses were performed in two stages: external analysis and cross sectional analysis. Firstly, each component of the PCA surface was scanned in order to determine its external chemical composition. Then, some components were selected for a cross sectional analysis in order to confirm the elements detected on the external surface and to determine the internal composition of the components. This procedure allowed to determine the distribution of metals on the surface and inside the components of the PCA.

2.2.2. Results

Fig. 1 summarizes the obtained results after mapping out an entire PCA. These results are essential to develop suitable concentration strategies in the subsequent recovery process.

The surface-mounted components are linked to the substrate (PCB) with solder joints which are made of a Pb-Sn-Ag alloy. The melting points of these solder joints vary between 176 and 228 °C depending on the alloy composition [9]. The substrate is composed of a large variety of materials: glass fiber, epoxy resin, brominated flame retardants components and metals. The most important metals detected

in the PCB were gold, copper and nickel. Gold provides a corrosion-resistant coating on top of the substrate, whereas nickel acts as an internal barrier to avoid the diffusion of copper through the gold layer.

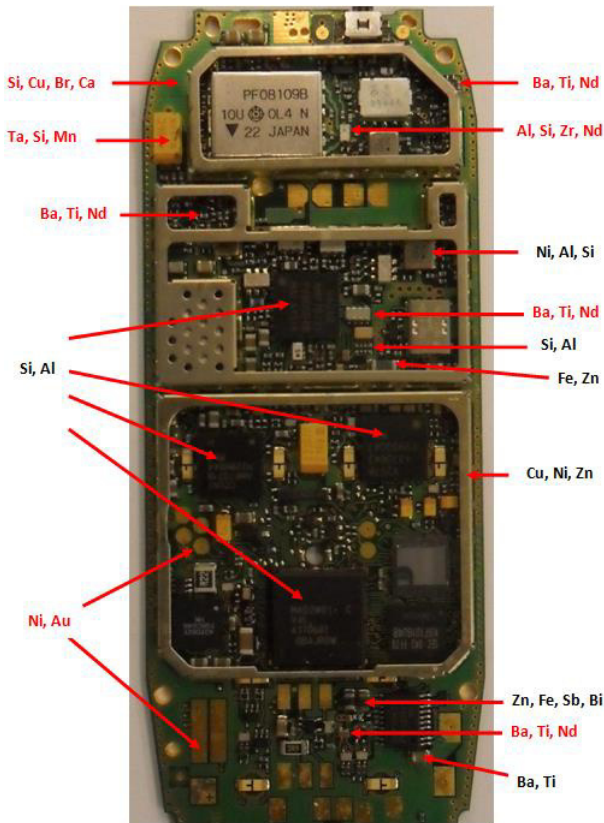


Fig. 1: Qualitative distribution of the main detected elements

Tantalum is concentrated in capacitors which can be visually distinguished from other electronic components by their box-shaped appearance and yellow color. Tantalum capacitors can be divided in 4 main parts [10]: (i) A terminal which is made of iron and nickel, (ii) a polymeric fraction in which silicon oxide is added to improve the thermal stability, (iii) a tantalum electrode which contains tantalum and other elements such as manganese and (iv) the dielectric which is made of tantalum pentoxide (Ta_2O_5).

Neodymium is found in the dielectric material of ceramic capacitors which are dispersed in different regions of the PCA. These ceramic capacitors have a size of less than 2 mm and they are mainly made of non-magnetic elements. They can be, therefore, separated from other surface-mounted components by physical separation techniques such as sieving, magnetic and density separations.

2.3. Mechanical pretreatment of surface-mounted components

2.3.1. Experiments

As discussed previously, the surface mounted parts are linked to the PCB with solder joints and the melting points of these solder joints vary between 176 and 228 °C. Thus, in

order to separate the surface-mounted parts from the substrate, the PCA was placed in an oven at 250 °C for 30 minutes to melt the solder and the surface mounted-parts were subsequently removed by scraping the heated PCA.

Following the liberation of the mounted parts from the PCA, tantalum capacitors were manually sorted by visual inspection. As illustrated in Fig. 2, the other mounted components were divided into nine fractions with 8 different-sized sieves 12 mm, 9.42 mm, 4.76 mm, 4 mm, 3.27 mm, 2 mm, 1 mm and 0.85 mm. Then, in all the produced fractions, a magnetic separation was manually performed by using a magnetic stirring bar and finally, in all the obtained fractions, a density separation was performed with tetrabromoethane which was chosen for its high density (2.97 g/cm^3). Experiments were conducted four times with different samples of the studied phone to verify the reproducibility of the separation process and all the produced fractions were weighed to determine the mass balance of the process.

Nd-rich fractions were shredded into a powder, which has an average particle size less than 100 μm , using a vibratory disc mill machine. Then, the elemental composition of the crushed powder was determined by EDS.

2.3.2. Results

Magnetic separation allowed to separate ferromagnetic fractions from non-ferromagnetic fractions and, as expected from the EDS analysis of ceramic capacitors, Nd-rich components are concentrated in non-ferromagnetic fractions. It can be seen from Fig. 2 that tetrabromoethane was efficient to separate the light components, which mainly consist of light elements such as silicon and aluminum, from the heavy components which contained other metals. The heavy components decanted while the light components floated on the surface of tetrabromoethane.

It can also be noticed that neodymium was detected in four fractions (2a, 7a, 8a, 9a). In fraction 2a, neodymium capacitors were integrated into an electronic package and it was not possible to shred this package with the disc mill machine. Thus, this fraction was not taken into account for the EDS analysis.

This EDS analysis revealed that the neodymium content varies between 4 and 14% and its concentration is higher in the smaller size fraction. By taking into account the mass of each fraction, it can be concluded that the studied mobile phone contains at least 18 mg of neodymium. This is an indicative value given that EDS only provides a semi-quantitative analysis. Moreover, it is estimated that the loudspeakers of mobile phones contain about 50 mg of neodymium [6], but they were not analyzed in the framework of this study.

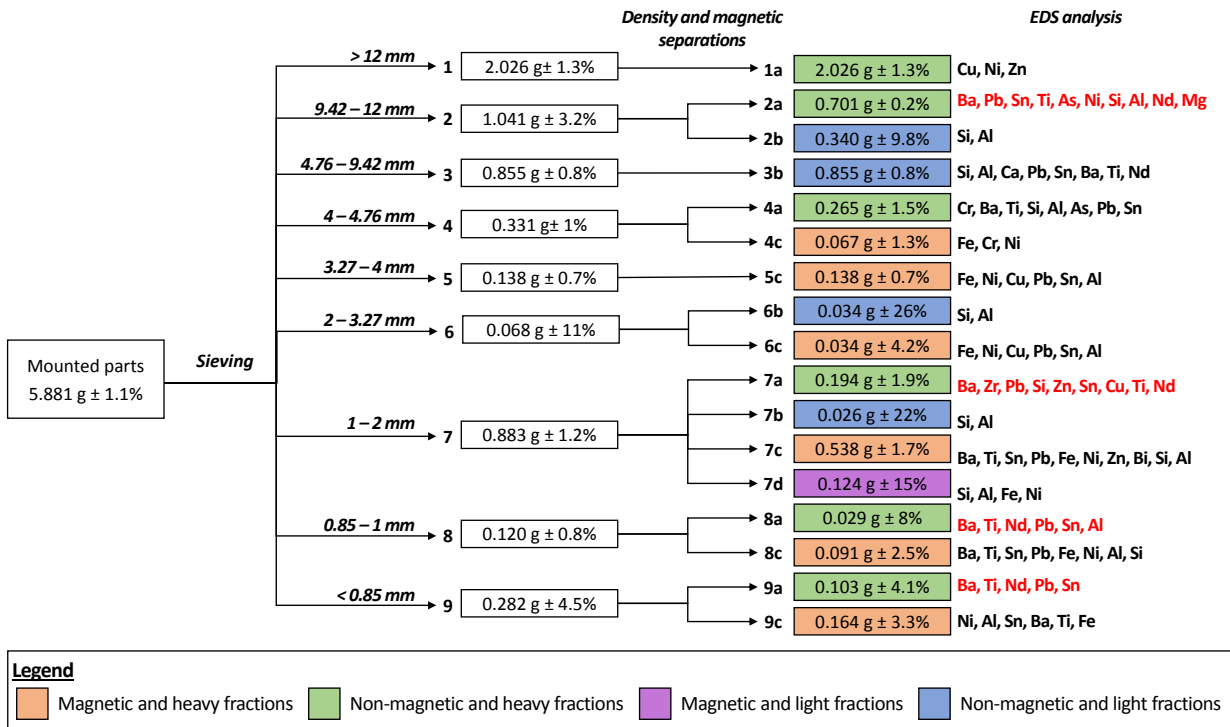


Fig. 2 : Mechanical pre-treatment of surface-mounted components after having excluded tantalum capacitors.

2.4. Tantalum recovery

2.4.1. Experiments

The EDS analysis of tantalum capacitors revealed the presence of Ta, Mn, Si, Pb, Sn, Fe, Ni and organic materials in tantalum capacitors. After an evaluation of the Pourbaix diagrams of these elements, it was decided to use a two-stage leaching procedure: NaOH leaching to eliminate Sn and Si and HNO₃ leaching to eliminate Fe, Mn, Ni and Pb. The leachates were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES).

The residual powder was heated in order to remove organic materials and analyzed by EDS. This combustion was followed by a differential scanning calorimeter. The sample was first subjected to a heating scan from 30 to 800 °C at a heating rate of 10 °C/min under oxygen and then, an isothermal heating at 800 °C for 1h.

Fig. 3 summarizes the tantalum recovery process with a mass balance at each step and the experimental conditions. It can be seen that significant material losses occurred in the crushing step and in the leaching steps (27% in total). Indeed, a small amount of the powder stays on the disc mill machine and on the filter paper. Moreover, the crushed powder is made of very small particle size (less than 100 μm) and they can be easily swept by ambient air during the processing.

2.4.2. Results

The ICP-OES analysis of the leachates demonstrates the feasibility of the hydrometallurgical recovery of tantalum

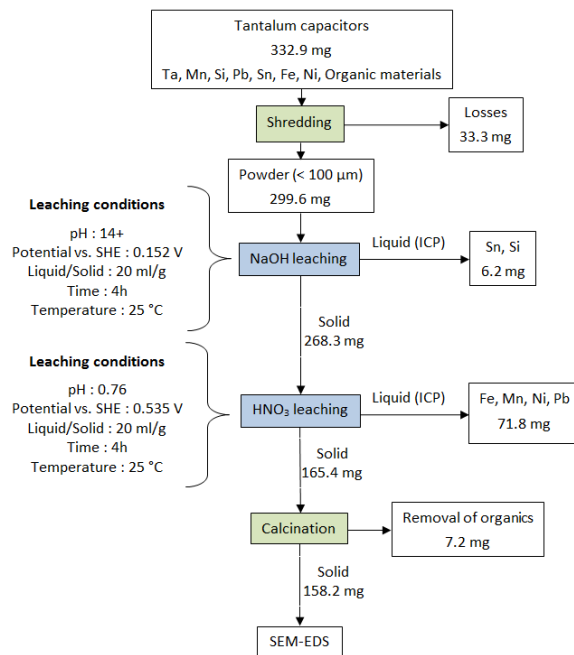


Fig. 3 : Schematic flow diagram of the recovery of tantalum at the lab scale

from tantalum capacitors of discarded mobile phones. Indeed, 80 mg of impurities were removed and it was found that Fe, Mn, Ni, Pb, Sn and Si are the main elements of the leachates as expected. However, a significant amount (about 16%) of Si is still present in the obtained powder after the refining

process and this is probably due to its presence in the oxide form. SiO_2 can only be solubilized by hot caustic alkaline solutions but in these conditions a significant amount of tantalum could also be dissolved. Another possibility would be to develop a process based on particle size difference in order to remove silica and to obtain a higher quality tantalum powder.

The EDS analysis of the powder at the end of the refining process revealed that the tantalum concentration in the residual powder is close to 50%. This means that the studied mobile phone contains at least 80 mg of tantalum. This value is in the same order of magnitude as that found in the literature [3, 6].

3. Towards an industrial process

3.1. A potential scale-up process

The proposed process allowed to obtain neodymium-rich fractions with neodymium concentrations varying from 4 to 14% and a high-quality tantalum powder. The metallurgical extraction of these metals is generally well known and this study demonstrates the need of a supply chain structure intended for the pre-processing of electronics wastes, with a particular focus on strategic metals. The recycling potential of neodymium and tantalum from end-of-life mobile phones is significant given the large amounts of mobile phones that are discarded every year worldwide.

The separation techniques used in this study are easily adaptable to an industrial scale. The mechanical pre-treatment of mobile phones could be done in a work integration social enterprise, in order to reduce the costs associated with the manual dismantling step. Mobile phones could be manually disassembled into their main constituting parts: plastic frames, metal frames, PCAs, batteries etc. Plastic frames which are made of engineering thermoplastic polymers could be used for purposes other than energy recovery. Given the high share of plastics in the total weight of mobile phones, their recycling would help reach the minimum recycling target imposed in the European WEEE directive which is 70% during 2015-2018. Tantalum capacitors could be retrieved from the PCA in the manual dismantling step or by using sensor-based separators, since they are easily distinguishable from other surface-mounted parts. Then, they could be sent to refining companies in order to recover tantalum by means of thermal or non-thermal processes.

The liberation of surface-mounted parts from the PCA could be performed in an oven with an automatic scraping system or with the help of flow of hot air. The substrate, which contains copper, gold and silver could be sent to refining companies. The mounted parts could be sieved with vibrating screens and the magnetic separation could be performed with magnetic drum separators. The obtained neodymium-rich-fractions could be sent to refining companies whereas the ferrous metal fractions could be fed into steel manufacturing processes.

3.2. Driving factors for an up-scaled process

The current recycling process focusses on the recovery of precious metals, while tantalum and REEs are currently lost in the slag despite their essential role in a large number of key technologies. Indeed, REEs are the most critical elements in terms of supply risks according to the criticality assessment of raw materials of the European commission [11], whereas tantalum is essential in electronic applications where miniaturization is of importance such as computers and mobile phones. These critical metals are at risk of supply shortage because of monopolistic supply conditions. For example, China currently produces about 90% of the world supply of REEs and the largest part of the estimated reserves are concentrated in this country [6, 11]. The recycling of strategic metals is therefore extremely important since it could help decrease this high supply risk.

Tantalum capacitors account for about 60% of tantalum use and given the dynamism of the electronic sector and the recent trends to decrease the size of electronic products, the global demand of tantalum is expected to increase in the coming years [6]. The primary production will no longer be enough to meet the increasing demand in the next few years. The recycling of tantalum capacitors could therefore help decrease the pressure on tantalum reserves.

Additionally, the average tantalum concentration of a tantalum capacitor is 36% whereas the content of tantalum in its natural ore varies between 0.015% and 0.025% [6, 10]. Moreover, tantalum is always associated with niobium in its ore and these two elements have very similar chemical properties. Therefore, large amounts of energy and chemicals are used during the concentration process of tantalum. Since tantalum capacitors do not contain any niobium, they can be considered as high quality ores of tantalum [10].

The recycling of REEs is essential not only for their supply risks and strategic economic importance, but also to provide a solution to the “material balance problem”. Indeed, the natural ores from which neodymium is currently produced, contain several REEs which are present in different amounts and the REEs content of these ores is not proportional to the market demand. This imbalance between the demand of individual REEs and their abundance in natural ores, leads to the production of some REEs in higher quantities than required by the market. The recycling of high-demand REEs such as neodymium could help get a perfect match between demand and production and to prevent the oversupply of REEs that are less in demand [12].

3.3. Limiting factors

The main limiting factor in the global recycling chain is not the efficiency of recycling technologies but the collection rate. Indeed, the current collection rate of end-of-life mobile phones is less than 10% at a global scale [1]. It is therefore essential to develop suitable collection systems and to enhance consumer awareness of the need and benefits of mobile phone recycling.

Additionally, an efficient recycling of discarded mobile phones is expensive due to the large number of steps

involved. The waste management of mobile phones is currently driven by the precious metal content in mobile phones and the recycling of tantalum and REEs is not economically viable at the current price levels.

The increasing complexity of mobile phones is another inhibiting factor for recycling. Indeed, current mobile phones are optimized for their functionality and cost and not for their reusability or recyclability. For example, most of the current mobile phones have an integrated lithium-ion battery and these batteries could cause explosion hazard in recycling plants [6]. This design trend means that high labor costs are needed to remove batteries before feeding them into a separate recycling process. A suitable design of future mobile phones is therefore essential to increase the recycling rates. This could be done by optimizing the number of screws in mobile phones, or by using only one type of screws in all mobile phone models. It could also be helpful to use the same shape, color and size for components which contain valuable metals, in such a way that they could be easily distinguished and separated from other components. This standardization would help automate the disassembly process.

4. Conclusions

The tremendous increase in the use of mobile phones together with their rapid obsolescence leads to the generation of large amounts of discarded mobile phones every year. Mobile phones contain many valuable resources and their recycling could help partly meet the increasing demand for key materials.

The aim of this paper was to determine the location of strategic metals in a mobile phone device and to develop a recovery process that will ensure high concentrations of these strategic metals. Mobile phone samples were manually dismantled into their main constituents and the qualitative distribution of strategic elements in the PCA was determined with a scanning electron microscope coupled to an energy dispersive X-ray spectrometer. This analysis revealed the presence of tantalum and neodymium in electronic components mounted on a substrate, which is composed of gold, copper and organic materials. Tantalum capacitors were manually sorted from mounted-parts and a tantalum rich-powder with a grade of 50% was obtained after applying a two-stage leaching process and a heat treatment under oxygen. Three neodymium-rich fractions with neodymium concentrations varying from 4 to 14% were obtained at the end of a mechanical process which includes sieving, magnetic and density separations. The proposed process could help increase the recycling rate of these strategic metals.

However, poor collection rates, complex product design and the lack of economic incentives are the main limiting factors for an effective waste management of end-of-life mobile phones. Therefore, combined efforts from manufacturers, recycling industries, consumers and governments are needed to develop an efficient recycling system. In this context and, given the current limitations of recycling, reuse of products or components is a better option. Indeed, the average service life of mobile phones is less than 2 years in developed countries, even though they can function

much longer. Reuse could extend the lifetime of mobile phones and thus reduce the amount of waste generated and the pressure on natural resources for manufacturing new products. It is therefore essential to enhance public awareness about our current utilization of mobile phones.

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