CHARACTERIZATION AND PROCESSING, BY MECHANICAL PROCESSING, OF PRINTED CIRCUIT BOARDS OF POST-CONSUMER SMARTPHONES

CARACTERIZAÇÃO E PROCESSAMENTO, POR PROCESSAMENTO MECÂNICO, DE PLACAS DE CIRCUITO IMPRESSO DE SMARTPHONES PÓS-CONSUMO

CARACTERIZACIÓN Y PROCESAMIENTO, MEDIANTE PROCESAMIENTO MECÁNICO, DE PLACAS DE CIRCUITO IMPRESO PARA SMARTPHONES POST-CONSUMO

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ABSTRACT
Smartphones are electronic devices increasingly used by Brazilian society in recent years. With the advancement of technology, the consequence is an increase in the disposal of this equipment in a shorter period of time. On the other hand, there are elements such as copper, gold and others can be recycled. Thus, the aim of the study was to employ mechanical processing techniques on smartphone printed circuit boards (PCB) in order to concentrate metals, especially copper, and identify the main elements concentrated in each process. For this purpose, 87 smartphones were collected and disassembled, after a mass balance, the PCB were comminuted and separated into 4 particle size ranges. Through chemical characterization, it is possible to verify a variation in the composition, according to the granulometry. With magnetic separation, iron contents of 9% to 34% were obtained, increasing the concentration proportionally to the increase in granulometry. The non-magnetic fraction went through the electrostatic separation process, in which conductive fractions of up to 52% copper were generated. It can be concluded that mechanical processing techniques are efficient for concentration of metals and alloys, especially copper, and, therefore, can bring economic and environmental benefits.

KEYWORDS
Smartphones; Characterization; Electronic waste; Urban mining.

RESUMO
Os aparelhos smartphones são os eletroeletrônicos cada vez mais utilizados pelos brasileiros nos últimos anos. Com o avanço da tecnologia têm-se como consequência o crescimento no descarte destes equipamentos em um menor intervalo de tempo. Por outro lado, há elementos como cobre, ouro, entre outros possíveis de serem recuperados. Assim, o objetivo do trabalho foi empregar técnicas de processamento mecânico, em placas de circuito impresso (PCI) de smartphones, a fim de concentrar metais de interesse, especialmente o cobre, e identificar os principais elementos concentrados em cada processo. Para isso, 87 smartphones foram coletados e desmontados, após um balanço de massa, as PCI foram fragmentadas e separadas em 4 faixas de tamanho de partícula. Através da caracterização

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química, foi possível verificar uma variação na composição, de acordo com a granulometria. Com a separação magnética, obteve-se teores de ferro de 9% a 34%, aumentando a concentração proporcionalmente ao aumento da granulometria. A fração não magnética passou pelo processo de separação eletrostática, no qual foram geradas frações condutoras de até 52% de cobre. Pode-se concluir que as técnicas de processamento mecânico são eficientes para a concentração de metais e ligas, especialmente o cobre, e, portanto, podem trazer benefícios econômicos e ambientais.

PALAVRAS-CHAVE

Smartphones; Caracterização; Resíduos eletroeletrônicos; Mineração urbana.

RESUMEN

Los smartphones son los dispositivos electrónicos cada vez más utilizados por los brasileños en los últimos años. Con el avance de la tecnología, la consecuencia es un aumento en la eliminación de estos equipos en un menor período de tiempo. Sin embargo, existen elementos como el cobre, oro, entre otros que se pueden recuperar. Así pues, el objetivo del trabajo fue emplear técnicas de procesamiento mecánico en placas de circuito impreso (PCI) de smartphones con el fin de concentrar metales de interés, especialmente cobre, e identificar los principales elementos concentrados en cada proceso. Para ello se recolectaron y desmontaron 87 smartphones, luego de un balance de masa, los PCB fueron fragmentados y separados en 4 rangos de tamaño de partículas. A través de la caracterización química se pudo verificar una variación en la composición, según granulometría. Con la separación magnética se obtuvieron contenidos de hierro del 9% al 34%, aumentando la concentración proporcionalmente al aumento de la granulometría. La fracción no magnética pasó por el proceso de separación electrostática, en el que se generaron fracciones conductoras de hasta un 52% de cobre. Se puede concluir que las técnicas de procesamiento mecánico son eficientes para la concentración de metales y aleaciones, especialmente cobre, y, por tanto, pueden traer beneficios económicos y ambientales.

PALABRAS CLAVE

1. INTRODUCTION

The smartphone industry has been growing constantly, including the consumer market, development of new models, and suppliers. (Laricchia, 2024a). In 2022, smartphone sales reached 1.39 billion units all the word and must reach 1.34 billion units sold in 2023 (Laricchia, 2023b).

Consequently, this portion of post-consumer equipment significantly adds to the amount of electronic waste (WEEE) being produced and has the potential to cause negative impacts on the availability of natural resources, on human health, and on the environment, because, just like other WEEE, smartphones contain toxic, rare, and precious materials (Mejame et al., 2016; Bookhagen et al., 2020; Kastanaki; Giannis, 2022).

In order to meet requirements for size, lightness, and functionality, a smartphone can use up to 50 different types of metals, which can range from basic ones such as copper and tin, up to precious metals such as silver, gold, and palladium (United Nations Environment Programme (UNEP), 2009). Many of these metals are considered critical, present industrial and technological importance, as well as supply risk (Işildar et al., 2019; European Commission, 2020; Kastanaki; Giannis, 2022).

Mobile phones and smartphones are packed with high-tech minerals, which are strategically important for emerging Chinese industries, such as cobalt, palladium, antimony, beryllium, neodymium, praseodymium, and platinum (He, 2020).

In general, a cell phone device can be divided into larger parts: printed circuit board (PCB), a display unit (screen), a battery, and a box/cover; in addition, antenna and accessories such as headphones, for example, are also included (Internacional Precious Metals Institute (IPMI), 2003; Gu; Summers; Hall, 2019).

Box 01 presents a review of the materials used in the main parts of a smartphone. According to the table, the diversity of critical metals that are used in a single smartphone can be seen, as well as how each metal is important due to its properties, and, consequently, assigns important functions to the equipment.

Part/ component	Elements
Electronics	Gallium, nickel, and tantalum, which are used in connections and various devices; silicon dioxide doped with phosphorus, antimony, arsenic, boron, indium, or gallium are used to produce the chips. Copper, gold, and silver make up the microelectronics and wiring. For welds that previously used tin and lead, these were replaced by mixtures of tin, silver, and gold after the 2003 RoHs.
Screen	They can be divided into three parts: - **A glass cover:** basically composed of aluminosilicate and may contain potassium ions to strengthen it. - **A capacitive layer (touchscreen)**: in which a thin, transparent, and conductive layer of mixed indium and tin oxides is used to produce the touch sensitivity property. In addition, silver, gold, and palladium can also be found. - **A screen:** in order to produce the colors of the screen, a varied composition of rare-earth elements is used, such as yttrium, lanthanum, terbium, praseodymium, europium, dysprosium, and gadolinium.
Microphones and speakers	Nickel is used in the diaphragms of the microphones; in addition, in these components there are usually magnets produced from neodymium-iron-boron alloys (NdFeB), and, in some cases, dysprosium, praseodymium, or gadolinium can be used in these alloys. These metals, in addition to terbium, can also be used in vibration units.
Cover	Covers can be produced from metal or polymers or a mixture of the two. The most commonly used polymers are ABS or PC/ABS; when made of metal, however, magnesium alloys are usually used, but aluminum, copper, and iron/steel alloys can also be used. In addition, brominated flame retardants can be found, although they have been minimized.
Battery	Nowadays, batteries have been made mostly of lithium ions. They consist of an aluminum housing, inside of which are a positive electrode, usually made of lithium-cobalt oxide, a negative electrode, which is usually graphite, and an organic solvent that acts as an electrolytic fluid.

Box 01: Composition of the main parts of a smartphone

Source: Brunning (2014); Cordella, Alfieri and Sanfelix Forner (2020); Venditti (2021); Konyalioğlu and Bereketli (2021); Santos (2016)

The technological advancement of this equipment is directly related to the increased use of metals, some of which are precious, increasing the economic potential of recycling this equipment (Gu; Summers; Hall, 2019).

According to Holgersson et al. (2018), the mass composition of smartphones is 20% printed circuit board (PCB), 33% polymer and metal casing, 26% screen, 13% battery, among others.

The main technologies used for recycling printed circuit boards involve physical, chemical, thermal and mechanical processes, often combining more than one process (Oliveira, 2012).

Disassembly is the first and most important process among all recycling processes, which is carried out mainly manually, valuing existing materials and concentrating metals (Knoth et al., 2002; Gouveia; Ferron; Kuno, 2014; Moraes; Espinosa; Lucena; 2014; Silveira; Santos; Moraes, 2019; Santos et al., 2022). The process of disassembling printed circuit boards can be classified into two different categories: selective and simultaneous disassembly. In selective disassembly, the board components are removed individually and simultaneously, all components are removed at the same time, being subsequently sorted (Layiding et al., 2005; Oliveira, 2012).

Subsequently, there is mechanical processing or physical processing, which is considered a pre-treatment with the objective of previously separating metals, polymers and ceramics, through the steps of comminution, classification (granulometric) and separation by different methods (Gerbase; Oleiveira, 2012). For the mechanical treatment of printed circuit boards, the steps involved can be grinding, particle size classification, magnetic separation, electrostatic separation, separation in a dense medium, among others (Moraes, 2011).

After this stage, there is the metal recovery process, which can be the hydrometallurgical and pyrometallurgical stages (Oliveira, 2012).

Although there are studies that grind entire devices for subsequent recycling of materials, the tendency towards the complete recycling of mobile phones and smartphones is the disassembly and separation by parts, each being treated by appropriate techniques (Bachér; Mrotzek; Wahlström, 2015; Gu; Summers; Hall, 2019; Flerus et al., 2019). Disassembly is one of the most important stages throughout the life cycle of electronic waste (Liu et al., 2022).

Having a disassembly step is very important for this type of waste, since each part presents value-added materials, which are easier to recover with disassembly (Santos et al., 2022). With this step, it is possible to better concentrate the metals of interest, considering that recovery techniques or parameters can vary according to the material, thus, it is possible to recycle the maximum of materials, including polymers (Santos et al., 2022). Manual disassembly is still widely used, as there are still challenges regarding automated disassembly of these devices (Gu; Summers; Hall, 2019).

Gold, silver, and copper are the most interesting metals of these devices, however, there are some studies that have been focusing on other metals, such as gallium, indium, germanium, nickel, palladium, strontium, among others (Gu; Summers; Hall, 2019; Flerus et al., 2019; Panda et al., 2020; Flerus; Friedrich, 2020; Silveira et al., 2020; Pourhossein et al., 2021).

Thus, the aim of this work was to employ mechanical processing techniques in order to concentrate metals of interest, especially copper, and identify the main elements concentrated in each process.

2. EXPERIMENTAL

This work can be divided into 2 stages. The first consisted of the collection, cataloging, disassembly, and mass balance of the equipment, while the second focused on the mechanical processing of printed circuit boards and chemical characterization.

2.1 Collection, Cataloging, Disassembly, and Mass Balance of the Equipment

Through campaigns carried out at UNISINOS by the Environmental Management System (EMS), as well as through personal donations in the years 2017 and 2018, 87 smartphone units were collected, with a total mass of 10kg. The cell phones were disassembled, weighed, and cataloged according to the disassembly order, registering the brand/manufacturer, color, model, and total mass of the device. Tools were also used to disassemble the cell phones (Philips screwdrivers and hex keys of different sizes). For mass balance, the intact cell phones and their parts were weighed in a BEL Engineering semi-analytical balance.

The devices were separated into printed circuit board, display, battery, housing, and others (parts that did not fall into any of the previous categories).

2.2 Physical processing of printed circuit boards

The next stages of the work were carried out only with the PCBs, while the other parts were stored for future studies. After weighing and separation, the printed circuit boards were comminuted whole in a cutter mill, model MGHS 2700, totaling approximately 1.5 kg of material.

The comminuted material went through two sieving phases. In a first moment, we used the classification methodology of the particle size according to the norm of Study Committee of Raw Materials (CEMP) 081 of the Brazilian Foundry Association (ABIFA) (2015), shaking for 30 minutes, using sieves with standardized openings - set of sieves with stainless steel frame 8" x 2" and Bertel shaker: sieves with the following openings: 3.35 mm; 2mm; 1.70mm; 1.0mm; 0.85mm; 0.60mm; 0.50mm; 0.425mm; 0.30mm; 0.21mm; 0.15mm; 0.106mm; 0.075mm; 0.053mm.

Then, the ground PCBs went through a second sieving, shaking for 15 minutes, after which the fractions presented in Box 2 were obtained.

Fraction	Particle size
F1	Smaller than 0.5 mm (F1<0.5 mm)
F2	Between 0.5 and 1 mm (0.5 mm<F2<1mm)
F3	Between 1 and 2 mm (1mm <F3< 2mm)
F4	Greater than 2 mm (F4>2mm)

Box 02: Fractions obtained in the second sieving.
Source: Authors.

The four fractions obtained were sent to a high intensity magnetic roller separator installed in the Laboratory of Corrosion, Protection, and Recycling of Materials (LACOR), from the Department of Materials Engineering in the Federal University of Rio Grande do Sul (UFRGS). The equipment was operated using a magnetic field of 1300 Gauss, with a roller rotation of 80 rpm45. The rotation was determined visually, so that there was no overlap of particles. The feeding was performed manually and subsequently calculated at 67 g/min. In this step, it is possible to separate the materials into magnetic (MAG) and non-magnetic (NMAG).

The non-magnetic material fractions were subsequently subjected to electrostatic separation. In this work, an INBRAS electrostatic separator, model ESP-14/01S, was used in the Corrosion, Protection, and Recycling Laboratory (LACOR) of the Department of Materials Engineering of UFRGS. The equipment was regulated at 30 kV voltage and roller rotation of 50 rpm. In order to reduce the humidity of the air, which was of 44% at the time of operation, a dehumidifier was attached to the separator. From this process, three different outputs of materials were obtained: a conductive (CON), an intermediate or mixed (material conducts momentarily) (INT) and a non-conductive (NCON).

2.3 Chemical characterization

Chemical analyzes were carried out at different moments of the process. The first was carried out after grinding and separating the circuit boards into different particle sizes, with the aim of identifying the metals present in the raw PCBs, that is, before the magnetic and electrostatic separations.

The second sequence of chemical analyzes was carried out after the magnetic separation process and the third occurred after the electrostatic separation process. In both, metals from the magnetic and non-magnetic, and conductive and non-conductive fractions were identified and quantified.

For the characterization of the materials, PANanalytical X-ray Fluorescence Spectrometer (XRF) by Dispersive Energy were used.

3. RESULTS AND DISCUSSION

The results found in the study will be presented and explained next.

3.1 Collection, Cataloging, Disassembly, and Mass Balance of the Equipment

87 smartphone devices were disassembled and identified, whose release years ranged from 2009 to 2015, belonging to 11 different manufacturers: Apple, Asus, Booster, LG, Motorola, Nokia, Positivo, Samsung, Sony, Wei, and ZTC. The total mass of the devices was 10 kg. From the initial characterization, it was possible to identify that the housing is the part with the highest mass (31%), followed by the display (30%), battery (20%), PCB (16%), and others (3%).

The higher mass of the housing occurs due to the fact that, in many mobile phones, it is made of polycarbonate (PC), or acrylonitrile butadiene styrene (ABS), or a blend of both, as well as a heavier internal housing, made of metallic or composite material. The value found by Holgersson et al. (2018) when quantifying the whole housing (plastic exterior plus interior) of smartphones was 33%, very close to the one found in this work.

Singh et al. (2018) separated the plastic housing from the metal one. The plastic one presented an average of 35.6% of the total mass of smartphones, while the metal one presented 27.72%, totaling 63.32% of the mass of smartphones in housings. However, the authors considered screws and other metal parts in the sum of the metal housing. Figure 1 shows the parts of the disassembled smartphones, in this work, and highlights the polymer and metal housings.

Figure 2: Outer housing parts (in red) with an inner housing part (yellow).
Source: Authors.

The displays of the devices also showed a high mass (30%). This result was already expected, as smartphones have touch displays with large dimensions. In addition, with the advancement of technology, there have been wider screen sizes than the ones from the previous generation cell phones. The same behavior was observed by Dorneles et al. (2016) when characterizing the devices of two generations (feature phones and smartphones), in which smartphone displays are equivalent to 18% of the mass of the cell phone, against 8% in conventional devices. Singh et al. (2018), when doing this same analysis, found screens of 9.13% and 14.79% of the mass of conventional cell phones and smartphones, respectively.

The printed circuit boards make up 16% of a smartphone and had an average mass of 18 g. The PCBs vary in size according to manufacturer and model. There is no standard size even between devices of the same manufacturer. In total, the studied PCBs corresponded to 1.50 kg of the 10 kg of cell phones. This means that for every 10 kg of smartphone waste, approximately 1.50 kg is of electronic circuits with metals that are available for recovery and insertion back into the production chain.

3.2 Physical processing of printed circuit boards

The printed circuit boards subjected to grinding suffered a 9% loss in the process. According to Tuncuk et al. (2012), losses between 10% and 30% may occur due to an insufficient release of metals during the comminution process. Silvas (2014) reports that, during grinding, the segregation of fine and light particles such as fiberglass can occur; these particles are expelled.

According to Andrade et al. (2022), grinding is one of the most widely used pretreatment processes in WEEE recycling. It can improve both their recycling as well as precious material recovery rates.

Many researches involving metal recovery in PCBs of mobile phones and smartphones perform a grinding step as a pretreatment, employing various techniques and obtaining various particle sizes (Jeon et al., 2018; Hossain et al., 2018; Díaz-Martínez et al., 2019; Liu et al., 2020; Wang et al., 2021a).

After comminution in the cutter mill, the particle size of the printed circuit boards was identified through the particle size distribution, in which there was a large variation. However, the largest amounts of material were concentrated in the particle sizes between 3.35 and 1.7 mm (44%) and between 1.7 mm and 0.85 mm (39%), as shown in Figure 2.

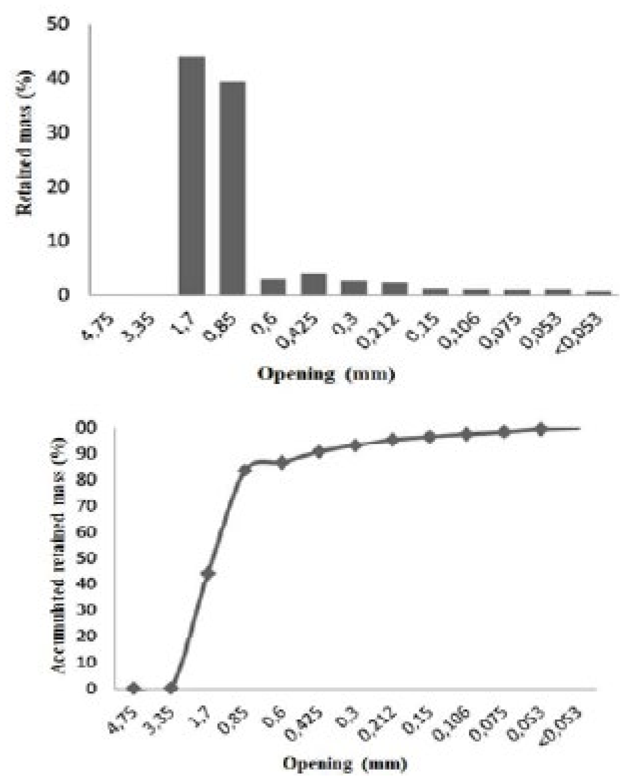

Figure 2: Particle size characterization of PCBs after comminution.
Source: Authors.

This variation occurred because PCBs are composed of materials that have different hardnesses. The particle sizes of 1.7 mm and 0.85 mm – 83% of the grinded PCBs – presented, predominantly, parts with copper tracks and other metals adhered to the glass fiber. In smaller particle sizes – from 0.6 mm – it was possible to visually some free metals, such as iron.

Subsequently, the separation of the material into the three particle sizes studied confirmed that the highest amounts of PCBs are concentrated in fractions of higher particle size, in a directly proportional relationship. That is, the higher the particle size, the greater the amount of PCBs retained. Most of the comminuted material (43%) was concentrated in the particle size above 2 mm (F4). The rest was retained in other sizes, being: F3 (33%), F2 (14%) and F1 (10%).

Moraes (2011) also verified that, after grinding, the PCBs from mobile phones have the tendency to accumulate in larger particle sizes. Copper and iron, for example, because they are ductile metals, are more difficult to grind than polymers. Therefore, they require a longer grinding time to achieve the same particle size of plastics, which are released more easily and in less time. Jesus and Casqueira (2015) state that "PCBs are materials that are difficult to grind, due to their high metallic content, which makes it difficult to obtain greater quantities of metals in the smallest fractions", this conclusion of the authors corroborates with the behavior obtained in particle size classification in the present work.

3.3 Chemical characterization

The samples before the separation processes (magnetic and electrostatic), called raw samples, which only underwent comminution and particle size separation, were chemically characterized by detecting the presence of many elements. In Figure 3 it is possible to verify the presence of 35% copper, 11% silicon, 5% aluminum, 3% iron, 3% nickel and 3% tin.

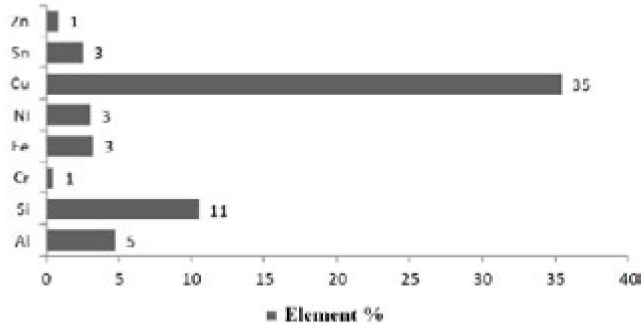

Figure 3: Elemental (average) analysis of gross printed circuit boards.
Source: Authors.

As expected, copper was the element found in the greatest quantity. This is the main metal used in the manufacture of PCBs. Its presence is mainly associated with the conductive tracks of the boards, connection pins, and also its presence in some electronic devices (Oliveira, 2012; Magalini; Kuehr; Baldé, 2015).

The presence of silicon and aluminum is related to the PCB armor. In addition to the glass fiber that is used to reinforce the board, silica and aluminum hydroxide are the main materials used to provide thermal expansion of the material. Aluminum is also used in some electronic devices as capacitors. The ceramic materials of PCBs are mainly represented by alumina and silica (Sanapala et al., 2009; Oliveira, 2012).

Iron is employed in some magnetic components and in PCBs links to the outside. Zinc is mainly found in resistors and also in Sn-Zn welds. Chromium and nickel are applied in metal alloys of electronic components, such as wires and conductive films of resistors. Nickel also has antioxidant function in circuits. Gold, despite having only an indication of its presence in the PCBs studied (< 1%), for its good electrical conductivity and solubility in tin welds, as well as its malleable and ductile nature, is used in the coatings of microchip connection pins and integrated circuits (Oliveira, 2012; Magalini; Kuehr; Baldé, 2015; Ribeiro, 2017).

As for rare-earth elements, the technique indicated the presence of Europium (Eu) as a trace element, in concentrations also below 1%. Rare-earth are used in the manufacture of PCBs electronic devices because they have the characteristic of, when added to a semiconductor, changing its electrical properties (Oliveira, 2012; Magalini; Kuehr; Baldé, 2015; Ayres; Peiró, 2013; Loureiro, 2013).

Tin and lead (currently prohibited in Europe by the RoHS directive) are used in the weld to fix the devices onto the plate substrate. However, a small amount of lead (1%) was identified in one of the fractions characterized (F3).

Comparing the characterization of the PCBs of the electronics from this study with others, the amount of copper is similar to that found by Yamane et al. (2011) and Holgersson et al. (2018) when chemically characterizing the PCBs of cell phones, presenting a low percentage variation among themselves when analyzing only the results of these authors. In all studies the major element was copper, as expected.

Wang et al. (2021b) analyzed, via ICP-OES, six points of PCBs of mobile phones (not specifying whether they were smartphones or not) that had a gold plating and, even so, for all points copper was the main metal.

Table 01 presents the concentration of the main metals in printed circuit boards obtained and comparison with other authors.

As can be seen, there is a difference in the concentration of other elements (iron, mainly) found by different studies. The PCBs of the cell phones studied by Holgersson et al. (2018) contained 34.27% copper, while those characterized by Bachér, Mrotzek, Wahlström (2015) had 21%. This difference was more significant for this element and for aluminum. For the other elements, the difference did not vary more than 1.5%. The variation of the concentration of the quantified metals may occur due to the different techniques used in the quantification (ICP-OES, ICP-MS, XRF, etc.), as well as the different types of devices (conventional cell phone, smartphone), manufacturing year, and PCB models (conventional cell phone, smartphone) (Hamerski, 2018).

Figure 4 shows the concentration of the main elements in raw PCBs with different particle size ranges.

The analysis by particle size fraction indicated that most of the elements are present in all particle size fractions, varying only their content. Aluminum varied between 6% and 4%, decreasing its concentration as the particle size increased. The same behavior was seen in silicon (18-8%), iron (6-1%), and tin (5-0.5%). Copper was the only element that remained predominant in all particle size ranges, having its lowest concentration in F1 (24%) and reaching the range of 40% in the other particle sizes studied (F2, F3 and F4.). Because it is ductile, copper is harder to grind than silicon and aluminum, for example. Due to this property, the amount in finer particle size ranges is smaller.

The copper concentration presented a directly proportional relation to the particle size, as 24% of Cu was identified in F1, with it remaining in the range of 39% in the largest particle sizes (F2, F3 and F4). It can be seen that, as the particle size of the PCB fraction increases, the amount of the element also increases. This is because copper is mainly used on the surface of the PCB as conductive tracks, making it difficult for it to be released. In larger particle sizes, copper is not free, but adheres to the tangles of fiberglass and resin, making it more difficult to separate due to its ductility (Oliveira, 2012; Silvas, 2014).

Economically important elements such as nickel and gold were more concentrated in the intermediate fractions – Ni with a content of 5% in F2 and <1% Au in F3, the only particle size in which the presence of gold was indicated. Aluminum had its highest concentration in the fraction with the smallest particle size, as well as in the PCBs of computers studied by Ribeiro (2013), who reports that this is probably "due to the fact that aluminum is in the form of oxide (alumina), a hard and fragile compound". According to the author, ceramic materials are pulverized, being concentrated on the smallest particle sizes. It should be noted that aluminum is also present in electronic devices as capacitors (Oliveira, 2012; Magalini; Kuehr; Baldé, 2015).

The contents of most metallic elements are generally lower in the finer fractions, according to Oliveira (2012), because scrap metals are hardly pulverized in grinding, as they have higher mechanical strength than the polymeric material. However, observing the PCBs before the magnetic and electrostatic separation processes, several quantified elements showed their highest concentration in the smallest particle size studied (Al – 6%, Si – 18%, Fe – 6%, and Sn – 5%), indicating that the cutter mill grinding was able to release these metals and/or their alloys or compounds.

Author	PCB Origin	Analytical technique	Metal (%)						
			Fe	Al	Cu	Sn	Pb	Zn	Ni
Moraes (2011)	Cell phone	ICP-OES	12.49	0.26	35.5	3.39	1.87	5.92	3.41
Yamane *et al.* (2011)	Cell phone	ICP-OES	10.57	0.26	34.49	3.39	1.87	5.92	2.63
Bizzo *et al.* (2014)	Cell phone	-	3.08	ND	14.2	4.79	2.5	0.18	0.41
Bachér, Mrotzek, Wahlström (2015)	Cell phone	XRF	1.5	4.8	21	2.4	0.42	0.49	1.6
Holgersson *et al.* (2018)	Cell phone	ICP-MS	0.68	1.9	34.27	1.93	0.37	0.55	1.16
	Smartphone	ICP-MS	0.88	1.78	39.5	3.22	0.03	0.67	1.54
Park and Kim (2019)	Mobile phone	ICP-OES	0.50	ND	47.90	2.00	ND	ND	0.8
Wang *et al.* (2021)	Mobile phone	ICP-OES	1.4	1.9	29.7	0.2	0.6	0.8	1.3
This study	Smartphone	XRF	3	5	35	3	0	1	3

Table 1: Concentration of the main metals obtained in printed circuit boards and comparison with other authors.
Source: Authors.

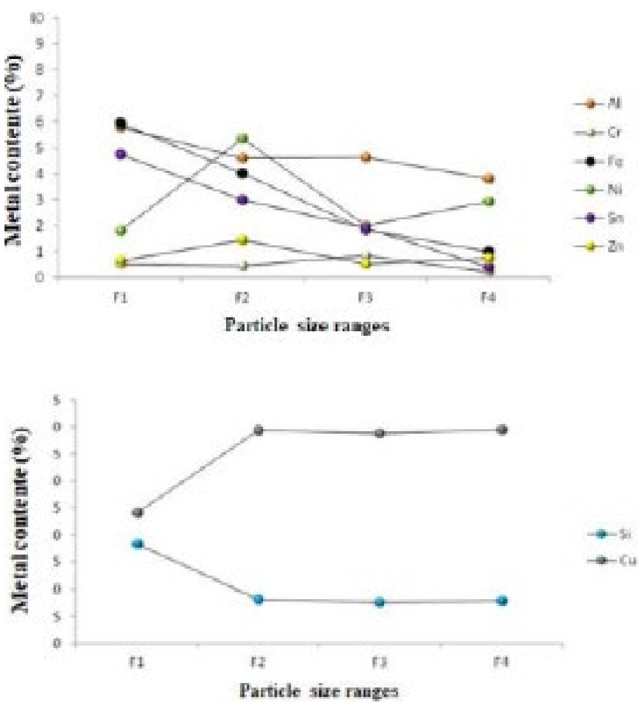

Figure 4: Concentration of the main elements in the gross PCBs with different particle size ranges
Source: Authors.

3.4 Magnetic and electrostatic separation

It was observed that 68% of the PCB of the devices are constituted by a fraction of non-magnetic material, while 29% of it are magnetic. This result was already expected, since the most abundant metal found in PCB is copper, considered a non-magnetic conductor. Checking the mass balance at the outlet of the electrostatic separation, it was proved that the conductive material (72%) predominates over the non-conductive (23%) and intermediate (5%). The same trend was observed by Yamane et al. (2011) when processing computers and cell phones; the authors obtained non-magnetic material in greater quantity and, after an electrostatic separation, predominantly conductive material. For conventional cell phones, the author obtained 82% of non-magnetic material. The amount of conductive material, on the other hand, was lower (50%) than the one found in this study.

Figure 5 shows the amount of magnetic and non-magnetic material according to the granulometric range.

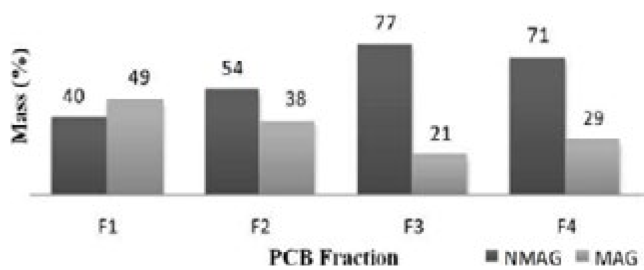

Figure 5: Portion of magnetic and non-magnetic material by PCB fraction
Source: Authors.

Thus, it is noted that, except in F1, non-magnetic material predominates over the magnetic one in all other fractions. That is, it is already inferred, from only the mass balance, that copper (non-magnetic element) is in greater quantity than iron and nickel, being concentrated mainly in the fractions F2, F3, and F4 than in the studied fraction of finer particle size. This is confirmed by the chemical analysis presented previously, in which copper, in the gross PCB analysis, had its lowest concentration in F1, with 24%.

Thus, it was already possible to identify, in the magnetic portion, the predominance of iron in all fractions analyzed. Although, in the F4 (>2 mm), only 29% of magnetic material was obtained, this was the particle size in which the release of this element was most easily noticed. In the non-magnetic portion, free copper was observed in fractions from lower to higher particle size, as well as polymers.

3.5 Chemical characterization

The results obtained from the quantitative chemical analysis by X-ray Fluorescence after the magnetic and electrostatic separations are presented in Table 02 in Appendix A and below is a discussion of these results.

In all samples, the concentration of copper is the most noticeable, except in the magnetic fraction of larger particle size (F4), in which the iron content is higher than that of copper. In F1 (<0.50 mm), the copper content reached 52% in the conductive portion, decreasing to 49%, 44%, and 40% as the particle size increases.

Analyzing the concentration of iron and nickel after magnetic separation, which aims precisely to separate these two metals from the rest, it is observed that the process allowed the concentration, in the magnetic fraction, of 9-34% of iron, 0-1.5% in the conductive fraction, and 0-0.5% in the non-conductive fraction. As for nickel, its highest contents were also concentrated in the magnetic portion when comparing the conductive and non-conductive ones. However, the amount of this element is much lower than that of iron, with contents between 3 and 11%.

Regarding the electrostatic separation, whose main objective is to concentrate copper, a content of 40-52% of the element was found in the conductive fractions. The portions of non-conductive material, however, also presented a considerable content of this element (22-37%). This means that the electrostatic separation process was not as effective, with high amounts of copper remaining in the portion of non-conductive material.

The presence of metals such as chromium (used in the manufacture of electronic components) and lead (used in welding, the most toxic of the elements and accumulative) was found. Lead-based welds are used for fixing electronic components in PCBs, and the tin-lead alloy is the most used one; this is due to its characteristics that give a good finish to the product, such as the degree of wetting (Almeida et al., 2013), low melting point, low cost, among other properties (Wehbie; Semetey, 2022).

According to Silvas (2014), the composition is still influenced by technology, since in more recently manufactured devices there is a tendency to replace the Sn/Pb alloy, used in older devices, with alloys containing Sn/Ag. and Sn/Bi, as well as other materials that have been replaced, such as Hg, Cd, Cr VI and flame retardants (polybrominated biphenyl-PBB and polybrominated diphenyl ether-PBDE).

This replacement is a consequence of Directive 2011/65/EU called Restriction of the use of certain hazardous substances (RoHS), which determines the restriction on the use of dangerous substances, such as lead, in electrical and electronic equipment (European Union, 2011; European Union, 2012; Silvas, 2014; Cheng; Huang; Pecht, 2017).

The results obtained from the PCB fractions after magnetic separation can be seen in Figure 6.

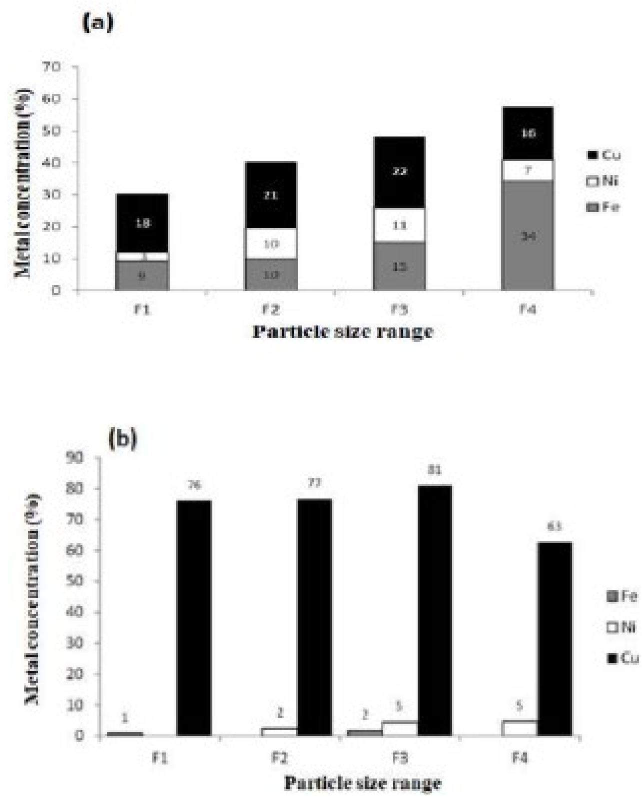

Figure 6: Amount of Fe, Ni, and Cu in magnetic particle size fractions (a) and Amount of Fe, Ni, and Cu in non-magnetic particle size fractions (b).
Source: Authors.

A high amount of iron is observed, which increases as the particle size of the material increases, as follows: 9%, 10%, 15% and 34%. According to Santana et al. (2013), in cell phones, iron is used in supports, shields, and screws, which is why the element is generally free, concentrating, then, in the thicker fractions.

Copper is the predominant element in PCBs and, despite not being magnetic, its presence in this portion

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of material is due to the drag it suffers with the iron and nickel particles attracted by the magnet (Veit, 2005).

The main objective of this stage of the process is precisely to separate iron, in addition to nickel, considered magnetic materials, to enable a greater concentration of copper in electrostatic separation. Therefore, it is more interesting that the highest concentrations of copper are found in the non-magnetic material portion, as can be seen in Figure 6 (b). These reasons determined the selection of these three elements for quantification in this topic: iron and nickel because they are the main magnetic elements, and copper because it is the element in greatest quantity and the one of greatest interest in PCBs.

In the portion of non-magnetic material (Figure 6 (b)), very high concentrations of copper were obtained for all particle size ranges studied: 76% in F1, 77% in F2, 81% in F3 and 63% in F4. Comparing with the amounts found in the magnetic portion (18%; 21%; 22% and 16%) the concentration of copper is evident, as well as the high amount of this element in all particle sizes. This result corroborates the study by Veit (2005), which found that metals tend to concentrate in intermediate particle sizes.

Iron was detected only in two particle size ranges (F1 and F3), and nickel in three (F2, F3 and F4), in low concentrations (1-5%), indicating that the objective of the magnetic separation process was achieved – to separate iron, mainly, and nickel, from the non-magnetic portion, keeping mostly copper (63-81%) for the next stage of the process (electrostatic separation).

After comminution of the PCBs, separation into different particle size ranges and magnetic separation, the portion of non-magnetic material was subjected to electrostatic separation, generating three different outputs of process materials: conductive, mixed and non-conductive.

From the mass balance (Figure 7), it is observed that, in all fractions, the predominance of conductive material over non-conductive material, leaving a smaller portion of mixed material, which corresponds to materials that momentarily conduct. In F4 (> 2mm) the largest amount of conductive material was observed, with 79% of mass.

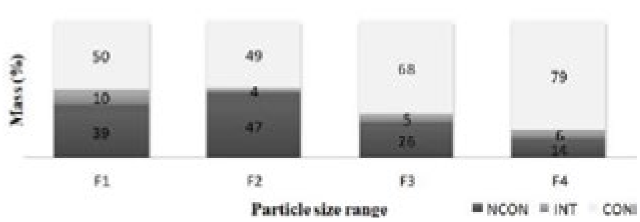

Figure 7: Amount of conductive, non-conductive, and intermediate PCB after electrostatic separation.
Source: Authors.

With the percentages shown in Figure 7, there is a predominance of copper, as it is a conductive metal, over other materials that make up the PCBs of smartphones.

In each particle size fraction, the elements for both the conductive and non-conductive fractions were quantified, disregarding the intermediate portion due to its low mass representation (4-10%) when compared to the two other portions. The results of the main elements found for each sample are presented in Figure 8.

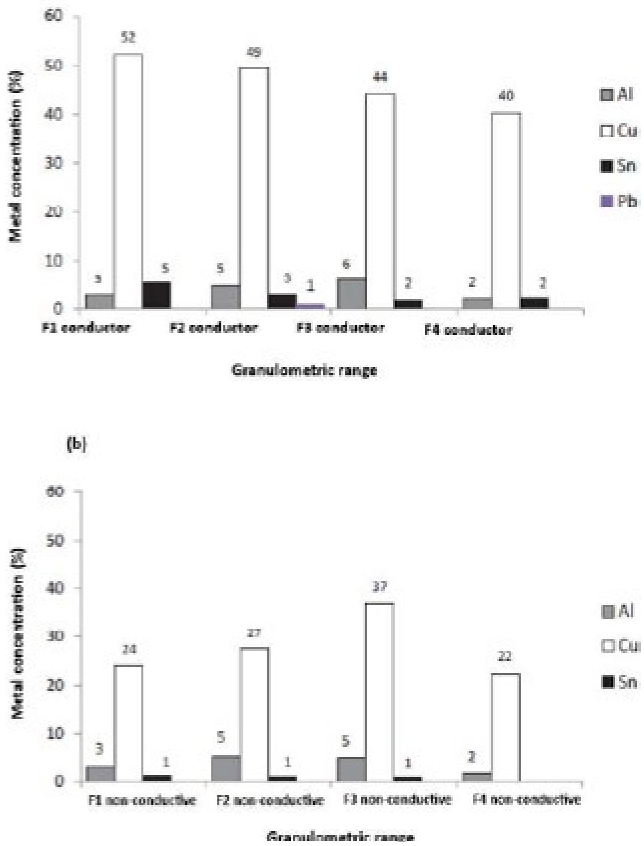

Figure 8: Amount of Cu, Al, Sn and Pb in conductive (a) and non-conductive (b) samples of different particle sizes.
Source: Authors.

Copper was the element found in the greatest quantity, both in conductive samples (F1 had the highest value at 52%) and in non-conductive samples. However, the variation in the content of this element between the particle size ranges studied was between 40-52%, decreasing as the particle size increased. In non-conductive samples, the variation identified was between 22% and 37%.

Some metals, such as copper, are found in both the magnetic and non-magnetic fractions, as it is a metal with a high quantity in WEEE PCBs, this element ends up being dragged along with the attracted iron and nickel particles (Veit, 2005). This is also a result of the agglomeration of particles which results in the attraction of some non-ferrous being carried by the ferrous fraction.

In addition to copper, metals with conductive characteristics such as aluminum (2-6%), tin (1-5%) and lead (1% in conductive F2) were identified in low concentration.

Iron and nickel, magnetic elements, were also quantified and their concentration can be observed in Figure 9 (a) and (b).

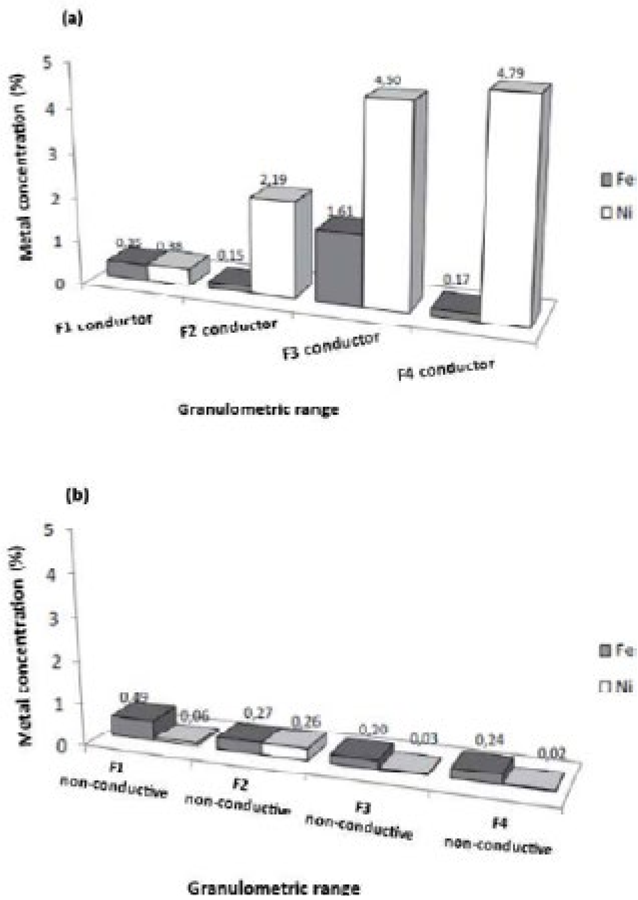

Figure 9: Amount of Fe and Ni in conductive (a) and nonconductive (b) samples.
Source: Authors.

Iron (0-2%) and nickel (0-5%), despite being magnetic, are also present in the conductive portion, but as trace elements (<5%). In non-conducting samples, this quantity is insignificant, with these elements in concentrations of 0.02 and 0.49%. This indicates that the magnetic process was efficient to concentrate both elements, mostly, in the magnetic fraction.

4. CONCLUSION

From the results obtained in this work, it is possible to conclude that:

- Smartphones are composed of housing (31%), display (30%), battery (20%), PCB (16%), and others (3%);

- The grinding of PCB in a cutter mill causes a 9% loss of the material, generating PCB fragments with particle sizes ranging from over 2 mm (44%) to 1-2 mm (33%), 0.5-1 mm (14%), and <0.5 mm (10%);

- Smartphone printed circuit boards are mainly composed of, on average, copper (35%), silicon (11%), aluminum (5%), iron (3%), nickel (3%), tin (3%), zinc (1%), and chromium (1%), as well as polymers and ceramics;

- From the chemical analysis by particle size fraction, it was verified that these elements are present in all particle size fractions, especially copper, which reached a 39% content in fractions F2, F3, and F4 of the PCBs;

- Copper is the element in greater quantity in smartphone PCBs, with contents of 81% in F3 and 77% in F2 in the portion of non-magnetic material. The variation in the portion of conductive material was between 52-40%, from the lowest to the highest particle size fraction studied. This indicates the efficiency of electrostatic separation, for concentrating mostly this element, as well as the grinding of PCB, for providing the release of this metal;

- In order to obtain pure copper it would be necessary to adopt complementary refining techniques, which could be applied in the conductive fractions, especially in F4. Hydrometallurgy or biotechnology techniques could be used, for example;

- Regarding the influence of particle size on the concentration of metals in the gross/before the separation processes PCBs, copper is concentrated in the intermediate particle sizes. The lowest concentration (24%) was in the smallest particle size (F1). Aluminum and Iron increase the amount as the particle size decreases. On the other hand, the relationship between particle size and metal concentration after the separation processes was as follows: in the conductive portion, the higher the particle size of the PCB, the lower the amount of copper. In the magnetic fraction, the larger the particle size fraction of the PCB, the greater the amount of iron;

- The use of magnetic separation proved to be efficient, as it allowed the accumulation of iron in the magnetic fractions (9-34%), retaining little of this element in the non-magnetic fractions (<5%);

- The presence of metals in high concentrations, especially copper, according to the characterization carried out, makes smartphones an interesting source of materials.

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