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ORIGINAL ARTICLE

Recovery of metallic gold, silver and copper from end-of-life mobile phones by hydrometallurgy

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Abstract

The global amount of end-of-life mobile phones is increasing over the years and their proper valorization is nowadays of strategic importance. Mobile phones can be considered as an important source of valuable materials, such as metals, plastics, and glass, which can be recovered and reintroduced into new production cycles. In this paper, a recovery process based on selective materials' separation and hydrometallurgy was proposed. After manual dismantling followed by the separation of the different fractions (plastic, metallic fraction, printed circuit boards, batteries, displays and glass), a hydrometallurgical process based on leaching and precipitation/reduction was applied on printed circuit boards with the aim of recovering the metals of interests. Tin was precipitated from an aqua regia leachate with gaseous ammonia and gold was afterward recovered as metallic gold by reduction with sodium borohydride; silver was first precipitated as silver chloride from a nitric acid leachate and then reduced to metallic silver. Copper was selectively precipitated with oxalic acid from the solution coming from silver recovery and then recovered as metallic copper by means of a mild thermal treatment, without chemicals addition. The developed process allowed the recovery of gold, silver and copper in metallic form with yield and purity grade ≥ 98%.

Keywords Electronic waste · Metal recovery · Hydrometallurgy · Circular economy

Introduction

In Italy, over 80 million mobile phones (MPs) are estimated to be present over a population of around 60 million inhabitants, where the so-called "smart phones" are progressively replacing "classic" devices. In particular, in 2021, there were almost 15 billion mobile devices worldwide, and this number is expected to reach 18.22 billion by 2025 [1]. In 2023, the Italian collective systems collected more than 340,000 tons of Waste Electrical and Electronic Equipment (WEEE, or e-waste), 22% of which belong to the Italian "R4 category", including IT & consumer electronics, lighting appliances, and photovoltaic panels [2]. Although there has been an overall growth of collected WEEE belonging to R4 category, it is estimated that MPs collection rate is less than 1%, according to data collected by the treatment plants. At European level, it is estimated that there are more than 700 million MPs stored at home [3]. Hypothetically, from MPs collection and recycling, several tons of Au, Ag, Cu, Pd, Co, and Li could be recovered, for an economic value of over 1 billion Euros. The total value of the materials contained in a ton of MPs can exceed € 15,000 due to the high content of Au, Ag, Pt, Pd, Cu, and other metals. In particular, it is estimated that 1 ton of EoL MPs contains up to 53 kg Cu, 141 g Au, 270 g Ag, 10 g Pt, 18 g Pd, and 3.3 kg REEs [4]. According to a recently published study, Cu, Al, Au, Pd, Co, and Li are the most relevant elements to be recovered: they account for the 99% of the total economic potential as well as for the 90% and 97% of the calculated scarcity and environmental burden parameters, respectively [5].

Tan et al. (2017) group MPs into feature phones (without operating system and with keyboard), multimedia phones (with operating system, touchscreen, and/or keyboard), and smartphones (with responsive touchscreen, fast operating system, and third-party applications) [6]. MPs are complex and heterogeneous matrices as they are made up of a considerable quantity of elements and compounds present in different quantities, such as metals, plastic, glass fiber, etc. [7]. Furthermore, these devices have different characteristics

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depending on the manufacturer, model, and production year, and therefore, an effective sampling strategy must be identified to ensure that the process of separation and valorization of the materials can be exported to the entire category of waste and that the selected samples represent the variety currently present at the end of their life.

In the global effort to promote sustainable waste management and reduce environmental pollution, the recycling of e-waste has gained significant attention among researchers. End-of-life (EoL) devices, such as MPs, are a critical focus due to their increasing accumulation and the valuable metals they contain, including Au, Ag, and Cu. Recent studies have demonstrated the potential of innovative recycling methods to enhance material recovery while minimizing environmental impact [8]. These approaches align with global sustainability goals adopted by the United Nations Member States, which aim at integrating environmental sustainability with economic growth and welfare and emphasize the transition toward a circular economy by converting waste into valuable resources [9].

Nowadays, there is a growing interest from the scientific community and several research papers focused on MPs recycling are being published over the last decades [10–19]. These investigated recycling processes at laboratory scale typically consist of a pretreatment step, including disassembly and size reduction, a separation step, based on particles size, density, magnetic and electrostatic differences, and the actual recovery step, which is normally performed by pyrometallurgical [12], hydrometallurgical [13–16], ionometallurgical techniques [18, 19], or a combination of them [17]. Moreover, novel approaches such as the use of supercritical water technology are being investigated for the removal of the organic fraction with simultaneous concentration of the target metals in the solid residue [11, 13].

The main gap to be overcome is the scalability of those processes from laboratory scale to industrial scale, by developing cost-efficient and sustainable recovery processes [10]. This study explores hydrometallurgical techniques as a cost-effective and eco-friendly method for recovering high-purity metals from e-waste, contributing to the ongoing global research on sustainable recycling technologies. The recovery of high-value metals from waste MPs allows both economic and environmental advantages, because it implies the reduction of primary raw materials depletion and the possibility of reintroducing the recovered metals in new production cycles. The objective of this paper is the selective separation and recovery of the materials contained in MPs with a focus on the recovery of high-purity metallic Au, Ag, and Cu. The novelty of the work mainly lies in the fact that the target elements (Au, Ag, and Cu) are recovered with high-purity grade directly in their metallic form, thus reducing the number of further unit operations before their use.

Materials and methods

Disassembling and material identification

Feature phones, multimedia phones and smartphones, produced by major suppliers from early 2000s to 2018, representative of the three generations of MPs, were used in the study: 115 devices were selected from the most popular manufacturing brands, including 55% smart phones and 45% old generation MPs. This selection allowed to obtain a bulk of samples which represents the variety of characteristics currently present on the market and guarantees that the recovery/valorization process of the constituent materials is applicable to all types of MPs. The presence of various materials in different quantities and types in every selected device makes product characterization necessary. Identifying the materials and grouping them by type allows to better select the potential supply chains which guarantee their best valorization routes.

Each device was subjected to the following stages, as shown in Fig. 1: (1) classification (brand and production year); (2) disassembly; (3) separation and chemical–physical analysis; (4) metal recovery process development; (5) process validation. The dismantling operation was carried out

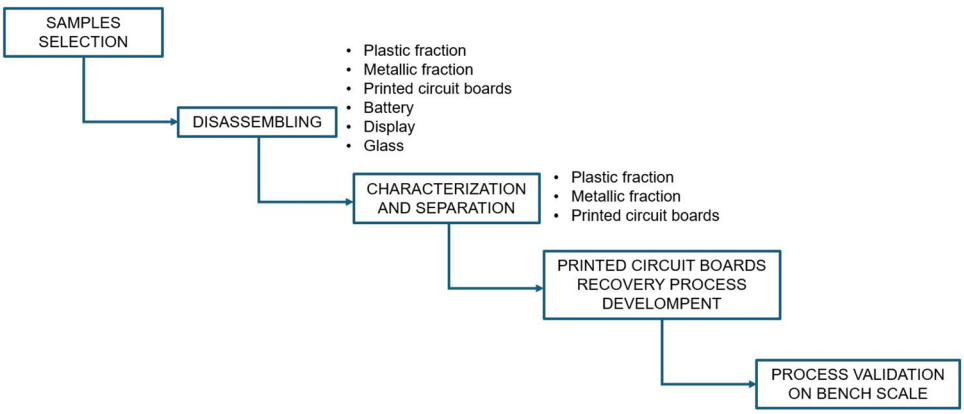

Fig. 1 Methodology used in this study for MPs valorization

manually and the weight of devices and fractions collected from the dismantling was determined by an EU-C LCD Gibertini balance. It was found that longer time was required for smartphones with respect to older featured phones (mainly 40% more time), mainly because the joints of smartphones are made partly with screws and partly with glue, making the separation of the different materials more difficult. Manual dismantling is a critical step, since it is expensive both in terms of time and work [20]. This operation is difficult to automate as each device has different characteristics and could only be simplified if EoL is considered in the design phase (eco-design); nevertheless, it is essential for the purposes of selective separation devoted to the valorization of materials.

In general, plastic is present in high percentages in older generation devices and is also present in considerable quantities in smartphones [21]; various MPs components such as the body, the internal supports and the keyboard are made of plastic with different properties (color, hardness, plasticity, elasticity, etc.), indicating the presence of different types of plastic materials. Plastic fractions coming from dismantling were separated and chemically characterized through Fourier-Transform infrared spectroscopy (FTIR) (SHIMADZU IR-Affinity-1 spectrophotometer equipped with ATR MIRACLE 10 device) and then classified according to the plastic polymer type; samples were analyzed without any specific treatment by recording the Fourier-transform infrared spectrum between 5000 and 600 cm^{-1}, performing 45 scans with 8 cm^{-1} resolution. The metallic fraction coming from dismantling is mainly made up of screws, nuts, supports, and pins, and its composition is highly variable from device to device; it was subjected to magnetic separation to separate ferrous fragments from the other non-ferrous. The components were also examined through a stereo microscope (Moticam 3.0 MP) to observe the surface of the various components to verify for example the presence of plastic films. These components were also analyzed via X-ray fluorescence (XRF) which indicated that they are made up of common non-ferromagnetic (brass, bronze) and ferromagnetic (iron, steel) alloys; this composition suggests that therefore they can be directly sent for recycling according to existing supply chains.

The electronic fraction is almost entirely made up of printed circuit board (PCB) and in a small part of connectors. PCBs are extremely heterogeneous matrices: many elements and compounds are present in different quantities, such as Ag, Au, Cu, Sn, and Pb; analysis of the non-metallic fraction in MPs shows the presence of bisphenol-A epoxy resins and unsaturated polyester resins in PBCs [4]. The abovementioned metals are tightly bonded to the board as a coated film or as welding, and this implies the need to extract, separate, and purify the elements of interest as if they came from a mineral: an effective valorization which includes most of the PCBs elements is possible only developing a specific process dedicated to this matrix to separate and purify the different metals. The quantitative MPs' electronic fraction characterization was carried out through two main steps: the first one was a grinding step by means of a cutting mill (Retch SM300) for 15 min at 2000 rpm and a homogenization with a planetary mill (Retch PM100) until dimensions ≤ 0.5 mm were obtained; the second step was a leaching through three consecutive steps: the first using 30% nitric acid (HNO_3) with solid-to-liquid (S/L) ratio 1:4 and the other two using aqua regia (HNO_3:HCl 1:3 v/v) with the same S/L ratio. At the end of each leaching operation, the solid residue was separated from the solution by centrifugation at 11,000 rpm, and then, it was washed with ultrapure water to reach neutral pH and to eliminate nitrate, chloride, sulfate, and phosphate anions at least below 1 ppm, dried, and weighted. Anions were detected with a Metrohm 883 Basic IC Plus ion chromatograph (IC). Analysis of the solutions obtained from the three consecutive leaching steps was performed by Microwave Plasma-Atomic Emission Spectrometer (4100 MP-AES, Agilent Technologies) and Atomic Absorption Spectrophotometer (AAS 6300, Shimadzu). Additionally, IC was used to verify the content of alkaline and alkaline earth metals, such as Na, Ca, Ba, etc. The obtained results were verified through material balances and inter-technical comparisons.

All reagents were purchased from Sigma-Aldrich and used as received without further treatments; the solutions were prepared with analytical grade reagents and distilled water. All experiments were performed at least in triplicate as a check on the experimental technique and precision, and the experimental results generally agreed within $\pm 5\%$ as a percentage error; the experimental error was lower than 5%.

Metal recovery from printed circuit boards

The recovery of metals was studied by a series of hydrometallurgical operations which allow the selective separation of the elements of interest (Au, Ag, Cu, Sn, etc.) with a defined chemical speciation and purity grade. The process was tested both on the grinded PCBs as such and on the non-ferromagnetic PCBs fraction.

The presence of different elements in different quantities closely linked in PCBs makes it necessary to adopt appropriate chemical separation and purification techniques to recover the elements of interest. The process for the recovery and purification of high added-value elements contained in PCBs here proposed involves two steps: a grinding pretreatment followed by a hydrometallurgical treatment. The grinding guarantees greater effectiveness of the subsequent hydrometallurgical process: the increasing of the contact surface between the leaching agent and the matrix allows the maximum solubilization of metals; it also exposes to the

leaching solution the metals of the PCB innermost layers, such as Cu, which otherwise could not be easily dissolved. After grinding, PCBs were subjected to the hydrometallurgical treatment.

The first leaching step was carried out at room temperature with 30 wt% HNO_3. The choice of HNO_3 as leaching agent was based on its proven efficacy in dissolving Cu and Ag [22]. Furthermore, it allows the simultaneous precipitation of Sn, facilitating its separation. Moreover, if compared to aqua regia, it does not contain chloride ions, allowing process simplification in case of industrial scale-up, since the use of special alloys such as Hastelloy® in the treatment plant could be avoided.

Acid concentration was set at 30 wt.% and contact time at 4 h, as resulted from preliminary studies. The S/L ratio was set at 1:3, which proved to be the minimum one necessary to guarantee the correct wettability of the sample and the effectiveness of the solubilization while ensuring minimization of wastewater production and thus environmental compliance. The system was kept under constant stirring to ensure full contact between the sample and the leaching agent.

Two fractions are obtained from the leaching step: a leachate containing the solubilized metals and a solid residue containing Au, Sn, glass fiber, and plastic material.

The solid residue was separated from the leachate by filtration and subjected to two consecutive leaching steps with aqua regia (S/L ratio 1:3, $t = 4$ h), with the aim of maximizing metal dissolution. After separation of solid residue, the leachate was treated with NH_4OH produced by bubbling NH_3 gas in the solution at pH 3.6 to precipitate Sn.

After Sn precipitation and concentration of the solution by evaporation, Au was recovered by reduction with $NaBH_4$. The Au solid metallic residue was washed first with a diluted solution of 5 wt.% HNO_3 then with ultrapure water, and finally it was melted in a muffle at 1150 °C.

Ag and Cu were recovered from the leachate obtained from the first leaching step with HNO_3 by fractional precipitation and cation reduction to metallic state as a powder. In particular, NaCl was added in a small molar excess to the solution to precipitate Ag. The obtained Ag° powder was washed with ultrapure water, dried, and melted.

Finally, Cu was precipitated with oxalic acid from the solution coming from Ag recovery and reduced to metallic Cu by means of a mild thermal treatment.

The experimental tests were carried out with the aim of reducing the use of reagents to the minimum amount necessary to ensure the effectiveness of the individual unit operations.

Once optimized, the developed recovery process was applied on about 10 kg of PCBs to verify it on a bench scale.

Each experimental test was performed at least three times. Each analysis was repeated three times. The associated error was always lower than 5%.

Results and discussion

Disassembling and material identification

From MPs dismantling operations, six material fractions were collected: plastics, glass, metals, electronic parts, batteries, and displays. The weighted average, which considers each of the fractions, is shown in Table 1. The effective recovery of the materials from these fractions is closely linked to an effective dismantling and selective separation system. Plastics is the most abundant fraction which constitutes on average 30 wt.% of the MP devices. This material fraction is made up of different types as indicated in Table 2; plastics identification by IR can be carried out in situ allowing their separation/recycling especially for the plastic materials present in greater quantities, such as PC which has been detected in 61% of the samples. Batteries and LCD displays, which, respectively, constitute 25% and 9% wt.% of MPs, once separated from other materials, can be headed toward the corresponding treatment and recovery chains. The same approach could be applied to the glass fraction, being present in minimal quantities (5 wt.%). Metals are present in two different fractions: the scrap fraction (screws, nuts, supports, and pins), which constitutes on average 12 wt.%, and the PCBs fraction with 19 wt.% (as reported in Table 1). The metals present in the scrap fraction analyzed by XRF showed the presence of a few elements typical of the most common metal alloys (Mn, Fe, Ni, Cu, Zn, and Mg); these fragments, once adequately collected, could be sent as such to existing supply chains to be recycled.

In Table 3, the results of the PCBs quantitative characterization are reported. These results allow the identification of the elements present in higher quantities and/or of greater value, in particular Cu, Sn, Ag, and Au, which were, therefore, selected as target elements of the recovery process. The large amount of iron (> 8%) suggested a magnetic treatment of the matrix to remove it mechanically, to save reagents and facilitate separation operations during the hydrometallurgical processes. Through this magnetic treatment, the sample was divided into a ferromagnetic fraction (FF) and a non-ferromagnetic fraction (NFF). The characterization of the two fractions is showed in Table 4.

Table 1 Material fractions collected by manual dismantling operations (weighted average)

Fractions	wt.%
Battery	25
Metallic fraction	12
LCD display	9
Plastic fraction	30
PCB	19
Glass	5

Table 2 Average weight percentages of each plastic type compared to the total wt.% of plastic materials present in MPs

Plastic material	Average wt.% (with reference to the tot. plastic materials)	Plastic material	Average wt.% (with reference to the tot. plastic materials)
PC	61	PC-ABS	10
Polyamide	14	PET	1
Epoxy resin	2	PMMA	3
PU/Silicon	3	Polyacetylene	1
Acrylonitrile	1	PC-PBT	1
Others (composite plastics)	3		

Table 3 Metal composition of PCBs

Element	wt.%	Element	wt.%	Element	wt.%
Cu	36.3	Al	1	Ti, Mn	0.2
Fe	8.3	Ba	0.7	Co, Pb	0.03
Zn, Ni	2.7	Ca	0.5	Au	0.07
Sn	1.9	Pb	0.4	V, Pd, Cd, Dy, Si, Na, Pr	< 0.03
Cr	1.1	Ag	0.3	Nd	0.04

Table 4 Metal content in NFF and FF expressed as wt.% compared to the total amount in MPs

Element	NFF wt.%	FF wt.%
Cu	74	26
Zn	58	42
Ni	34	66
Ag	67	33
Sn	45	55
Fe	1	99
Pb	50	50
Au	50	50
Others (Na, Si, V, Mn, Al, Ti, Cr, Ba, Na, Ca, Co)	50	50
Residue	50	50

It can be observed that from the magnetic separation, Cu mainly enriches the NFF (Cu is present for 74%) corresponding to 87% by weight of the total, while, as expected, Fe is present for 99% in the FF one. This finding considerably facilitates the subsequent separation and purification process of Cu in the NFF fraction, given the absence of Fe. The FF mainly consists of Fe (35%) and Cu (33%). Au is equally distributed between the two fractions and an eventual concentration through mechanical/physical separation is not possible as they are intimately connected.

Metal recovery from printed circuit boards

The recovery process was tested both on the grinded PCBs as such and on the non-ferromagnetic PCBs fraction.

Regarding the leaching step with 30 wt% HNO_3, as explained above, it was found that the S/L ratio 1:3 is the minimum one necessary to guarantee the correct wettability of the sample and the effectiveness of the solubilization.

Sn recovery

The solid residue obtained after the first leaching steps with HNO_3 was subjected to two consecutive leaching steps with aqua regia; this treatment resulted in the solubilization of Sn and Au and other PCB metals present in small quantities (< 1 wt%) which were not solubilized during the first HNO_3 leaching. The sequential leaching approach demonstrated superior efficiency due to the increased surface area achieved through grinding, facilitating complete metal solubilization.

The obtained solid residue represents 45 wt.% of the matrix and is constituted by glass fiber, plastic etc.); the leachate was treated with bubbling NH_3 which allowed quantitative precipitation of Sn (> 98%), as well as Ti (97%) and, if present, Fe (98%). Ti is present in PCBs in small quantities (0.2%); therefore, it does not significantly affect Sn purity. Fe is present in quantities greater than 8%; therefore, if the solid residue comes from the NFF, which does not contain Fe, the Sn precipitate has a purity > 94%, since the separation from this element occurred in the magnetic pretreatment phase. If, however, the solid residue comes from the treatment of the PCBs as they are, it is necessary to treat the Sn precipitate with diluted HNO_3; this allows part of the Fe present to be partially eliminated. The purity of the recovered Sn compound is in this case about 70%.

Au recovery

After the selective precipitation of Sn, Au remains in the liquid phase. Mn, Al, Co, and Ni are present in the solution in small quantities, but they can interfere with Au recovery; therefore, the solution pH was adjusted to 6 to

precipitate them. As explained above, after solid/liquid separation and concentration of the solution by evaporation, Au was recovered by reduction with $NaBH_4$. The yield of the reduction and purification operations was 98% and the purity grade of the obtained product (metallic Au) was > 98%.

Au recovery yield was found to be similar to other reported studies: Au recovery yield was over 98.4% when using dimethylformamide–$CuCl_2$–NaCl as leaching media followed by precipitation with water [11], while the use of microwave pyrolysis as pretreatment followed by leaching and oxidative precipitation with perchloric acid allowed to recover approximately 93% Au [14].

Ag recovery

To recover Ag from the leachate, it was necessary to remove Pb and Ba by adding concentrated H_2SO_4 in a small molar excess; this step allowed quantitative precipitation of both elements (Pb > 99%, Ba > 98%). Subsequently, NaCl was added in a small molar excess to this solution to achieve the quantitative precipitation of Ag (> 98%). The obtained salt (AgCl), with has a purity grade > 98%, was solubilized in a basic environment and the Ag^+ was reduced to metallic Ag° with a small stoichiometric excess of solid glucose or sucrose. Both reducing agents were selected, because they are considered as green chemicals, since they come from renewable sources. The reduction and purification yield was 98% with a purity > 98%.

Cu recovery

Cu was quantitatively precipitated with oxalic acid from the solution coming from Ag recovery and this is due to the low solubility of copper oxalate in water [23, 24].

Oxalic acid was here selected as precipitating agent for Cu, because Fe is present in relatively high concentration in the leachate as well (cfr. Tables 3, 4), and oxalic acid is selective for Cu against Fe: for this reason, the obtained precipitate has a high-purity grade.

Furthermore, copper oxalate is a valid precursor to obtain Cu in metallic state by a mild thermal treatment without adding further reduction agents. The solid residue was heated ensuring an inert environment on the surface (nitrogen). The thermal treatment causes the degradation of oxalate with the production of metallic Cu. The two-step products (Cu oxalate and metallic Cu) were analyzed to determine their purity which was found to be > 98%. The recovery yield and purity here achieved were found to be comparable to those reported using leaching with ionic liquids followed by electrowinning [19].

Overall material recovery

As reported above, the optimized recovery process was applied on about 10 kg of PCBs to verify it on a bench scale; it was found that the results are aligned with the laboratory scale tests previously performed.

The developed hydrometallurgical recovery process is shown in Fig. 2.

In conclusion, it can be stated that the selective MPs separation obtained through dismantling and the additional hydrometallurgical process carried out on the PCBs allow the recovery of several materials and their valorization and reintroduction into the most appropriate production/supply chains. In particular, Table 5 shows the quantity of each product which can be obtained from the treatment of one ton of MPs. It can be observed that from the treatment of one ton of MP devices it is possible to: (1) separate 250 kg of batteries and 90 kg of LCD displays to be sent to the proper treatment plants; (2) reintroduce into existing recovery chains approximately 120 kg of metal scrap, 180 kg polycarbonate and 50 kg glass; (3) obtain secondary raw materials such as about 70 kg Cu, 600 g Ag, and 140 g Au (as metallic Cu, Ag, and Au) with a purity grade > 98%. Finally, with reference to PCBs treatment, Table 6 shows the chemical speciation, the purity grade, and the recovery yield of the elements of interest obtained from the hydrometallurgical recovery process here proposed, according to the different unit operations.

The novelty of the proposed process lies in the fact that the elements of interest (Au, Ag, and Cu) are recovered with high-purity grade directly in their metallic form, thus reducing the number of unit operations required before they can be used in new production cycles. The innovation lies also in the fact that oxalic acid allowed the selective precipitation of Cu against Fe; the obtained copper oxalate is already a product, but it can be also directly reduced in metallic form by means of a mild thermal treatment, without the addition of further chemicals.

By considering the current market value of the target elements, it can be assumed that from the treatment of 1 ton of MP devices, a value of about 15,000 € can be reclaimed [25].

It must be pointed out that attention was here paid to the reduction of the reagent's dosage to the minimum amount necessary to ensure the effectiveness of the individual unit operations; consequently, the production of wastewater was minimized as well. Precipitating/reducing agents were always added in solid form, except for the NH_3; however, the required NH_3 volume was significantly lower compared to the volume of the leachate to be treated. Such wastewater is a hypersaline solution which would need to be subjected to further treatment before safe disposal; alternatively, after neutralization, it could be used for other treatment cycles.

Compared to pyrometallurgical methods, the proposed hydrometallurgical process offers significant advantages in

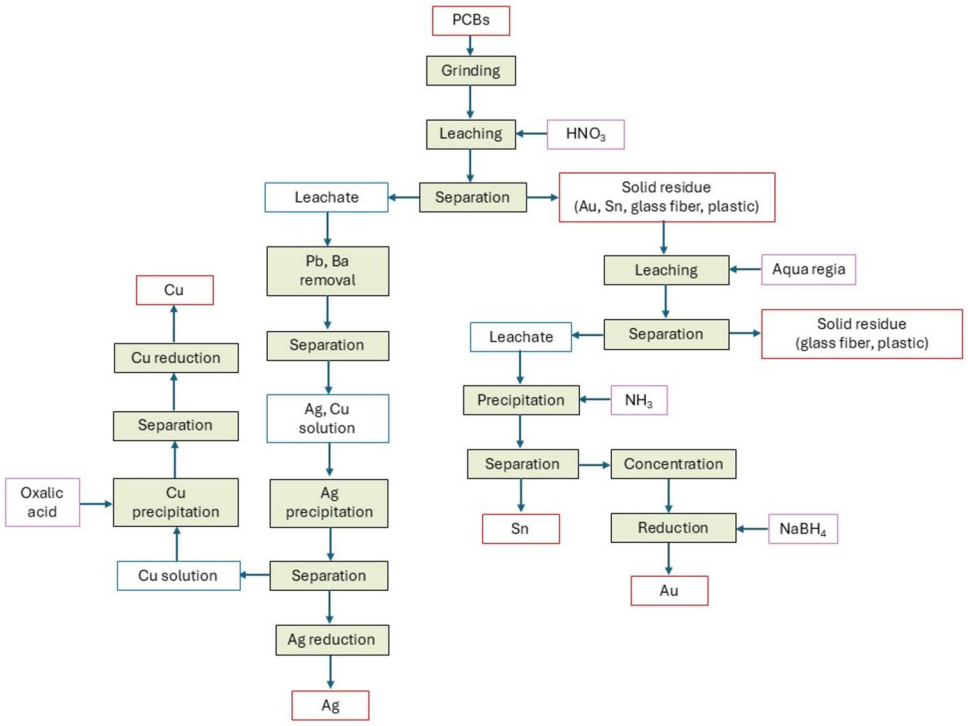

Fig. 2 Proposed hydrometallurgical process

Table 5 Quantity of each fraction which can be obtained per ton of MPs

Valuable fraction	kg/ton_{MPs}
Metallic fraction	
Ferromagnetic	108
Non-ferromagnetic	12
PCB	
Cu	73
Sn	4
Ag	0.6
Au	0.14
Plastic fraction	
PC	183
Polyamide	42
ABS	30
PMMS/PU/silicon	9
Lithium-ion batteries	252
Glass	50
LCD displays	90

terms of energy efficiency, while maintaining high recovery yields: process steps are in fact carried out at room temperature, thus reducing energy consumptions and emissions. Moreover, both leaching and precipitation are relatively simple operations and they can be potentially integrated in an e-waste treatment facility without requiring high capital investments.

Table 6 Chemical speciation, purity and yield of the elements of interest from the PCBs hydrometallurgical recovery process

Element	Chemical state	Purity grade	Recovery yield
		%	%
Au	Metallic	> 98	98
Sn	Compound	> 70	98
Ag	Salt	> 98	98
	Metallic	> 98	98
Cu	Salt	> 98	97
	Metallic	> 98	98

Dismantling was found to be difficult to automatize, because each device has different characteristics; however, it could be strongly simplified if the recycling stage would be considered during the production stage (eco-design).

Further research will be needed to verify the possibility to (partially) automatize the disassembly step in a cost-effective way, to further low down the recycling costs and reduce the gap toward process industrialization.

Conclusions

MPs are complex and heterogeneous matrices, because they are constituted by a significant number of elements and compounds in different quantities (metals, plastic, glass fiber,

etc.); furthermore, they have different characteristics according to producer, model, and production year.

In this paper, the valorization of EoL MPs was investigated through materials' selective separation followed by the recovery of Au, Ag, and Cu contained in PCBs by means of hydrometallurgical techniques. From 1 ton of MPs, about 250 kg batteries, 90 kg LCD displays, 50 kg glass, 180 kg polycarbonate, and 120 kg metal scrap can be obtained. The hydrometallurgical process allows to recover the elements present in highest quantities and of strategic interests (Au, Ag, Cu and Sn) by selecting the unit operations to fine-tune both purity and chemical speciation of the obtained products. Metallic Au, Ag, and Cu were obtained with a purity grade > 98%. Cu, in particular, was selectively precipitated with oxalic acid from the solution coming from Ag recovery and then recovered as metallic Cu by a mild thermal treatment, without the addition of further chemicals.

Magnetic separation was found to be an effective pretreatment step which allows to separate Fe, to simplify the separation/purification of the recovered elements and to significantly increase Sn purity.

This study demonstrates the feasibility of recovering high-purity Au, Ag and Cu from EoL MPs through a combined manual dismantling and hydrometallurgical process. While the process achieves quantitative recovery of the target metals, future research should focus on automating dismantling steps and on developing eco-friendly alternatives to hazardous chemicals to enhance scalability and sustainability. These advancements will further align the process with circular economy principles and contribute to sustainable e-waste management.

Author contributions All authors have contributed equally to the work.

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Declarations

Conflict of interest There are no conflicts of interest/competing interests to declare.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

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