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Mining Critical Raw Materials from Waste Printed Circuit Boards by Thermo-Mechanical Disassembly

Alice PROIETTI[1,a*], Denise BELLISARIO[1,b], Leandro IORIO[2,b], Dounia NOQRA[1,d], Giorgio PATRIZII[1,e], Fabrizio QUADRINI[1,f] and Loredana SANTO[1, g]

[1]University of Rome Tor Vergata, via del Politecnico 1, Rome, Italy

[2]Mercatorum University, p.zza Mattei 10, Rome, Italy

[a]alice.proietti@uniroma2.it, [b]denise.bellisario@uniroma2.it, [c]leandro.iorio@unimercatorum.it, [d]dounia.noqra@uniroma2.it, [e]giorgio.patrizii@uniroma2.it, [f]fabrizio.quadrini@uniroma2.it, [g]loredana.santo@uniroma2.it

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Abstract. EU dependence on critical raw materials poses a serious risk of fragility and vulnerability, mainly in strategic sectors such as energy and defense. Electronic boards contain high concentrations of metals, particularly copper and lead, comprising critical raw materials. Urban mining offers the possibility to recover these metals, but continuous innovation is necessary to make extraction processes more efficient, productive and cheap. Contamination is an issue which reduces the extraction performance and increases pollution and costs. Thermo-mechanical disassembly can be used to separate electronic components from boards, by removing the soldering alloy. A thermo-mechanical disassembly process, currently under patenting (namely "impact desoldering"), allows the separation of electronic components and soldering alloys from the boards by impact. It has been applied to 4 kg of waste boards to separate the printed circuit board from the electronic components, and the soldering alloy has been melted to make an ingot. A further thermo-mechanical process has been also applied to extract copper sheets from the disassembled printed circuit boards, by rolling.

Introduction

The ever-increasing development of advanced technologies, renewable energies and digitalization requires a huge availability of high-valued raw materials, which can be difficult to collect because of several geopolitical issues. Such materials are commonly called Critical Raw Materials (CRMs) and, since 2011, they are grouped into a list which is updated every 3 years because of economic and social factors. At first 14 CRMs were identified and, since then, their number increased with an average rate of 1.67 until now, with the current list including 34 materials [1]. According to the European Commission, a classification of raw materials can be done based on their supply risk (SR) and economic importance (EI); in this frame, CRMs are those materials that have a SR≥1 and an EI≥2.8 [2]. The Critical Raw Materials Act (CRMA), promoted by the European Union, aims to regulate CRMs management and to support industries during their acquisition. In this regard, new strategies are developed in such a way to mitigate the supply risk, fostering the recovery of CRMs from urban waste [3]. This process is called urban mining. Indeed, CRMs are typically used in electronic systems, whose end-of-life represent a serious environmental challenge [4]. In fact, in 2022 the global electronic waste (e-waste) reached 62 million metric tons (MMTs), representing a huge source of materials since it included 31 MMTs of metals, 17 MMTs of plastics and 14 MMTs of other materials and only the 22.3% of e-waste is currently recycled [5]. Wasted electronic boards are the 7% of the total amount of e-waste; they are complex systems [6] that integrate high-value precious metals such as gold (Au), palladium (Pd) and silver (Ag) as well as strategic metals like copper (Cu) and tin (Sn) [7]. Electronic boards consist of a non-conducting fiberglass laminate that integrates conducting Cu traces, which is named printed circuit board (PCB), and the electric components that are secured on the substrate. Among these devices there are chips, connectors and capacitors, that contain precious and high-value metals [8]. Figure 1 shows a typical PCB with its components. To recycle such

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systems, several steps are required. First, mechanical processing like dismantling, upgrading and refining are necessary [9]. The former permits the separation of materials and components into different categories; it is performed using hammers, tongs, screwdrivers and conveyors. In alternative, thermal treatments, as infra-red heating, and chemical reagents, as a solution of fluoroboric acid and hydrogen peroxide (HBF_4– H_2O_2) or nitric acid (HNO_3), can be used. After dismantling, physical processes are carried out, that involve the use of crushers and grinders to obtain small parts, and a subsequent step of separation is carried out to divide metallic and non-metallic parts. Magnetic, eddy current and density separation techniques can be used for this purpose. Chemical processes are also used to separate organic and metal parts with the 4 most relevant techniques being pyrolysis, gasification, depolymerization using supercritical fluids [10] and hydrogenolytic. Subsequently, refining is performed; in this step metals are melted by heating (pyrometallurgical processes) or dissolved by a liquid (hydrometallurgical processes) as well as by a combination of them [11,12]. Hydrometallurgical treatments involve acid, cyanide or caustic leaching of solid material. In the end, high-value metals are isolated and concentrated. An innovative approach is based on bio-hydrometallurgical leaching processes, which use microorganisms to extract metals by generating weaker organic acids. These processes can be used to produce ferric iron generated by bacterial oxidation of Cu and zinc (Zn) from PCB. This solution permits to overcome the limits of pyrometallurgical and hydrometallurgical processes by increasing the amount of PCB that can be processed [13]. Finally, the purification of the dissolved metals is carried by liquid/liquid extraction (solvent extraction), precipitation/cementation and electrolyte refinements (electrowinning/electro-recovery). Hazardous materials such as cadmium (Cd), lead (Pb) and chromium (Cr) must be opportunely managed whereas for rare earth metals, such as cerium (Ce), yttrium (Y) and neodymium (Nd), bio-metallurgy and solvent extraction methods are commonly used [14,15].

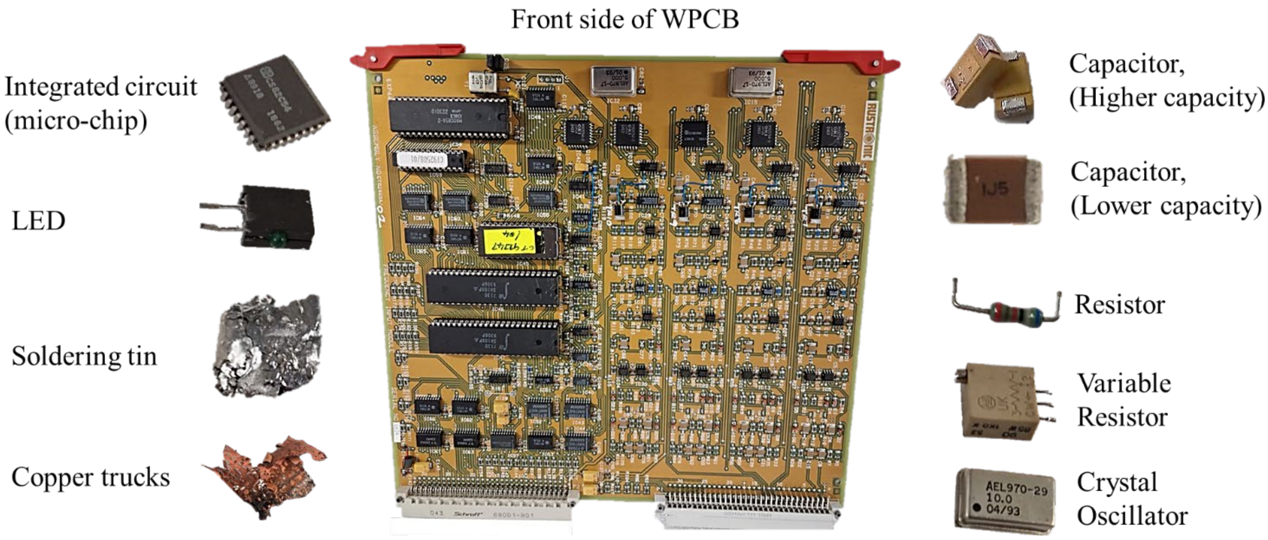

Fig. 1. An example of a board with both the PCB and the electronic components.

Several strategies can be adopted to recover the most high-value materials in electronic boards. Chemical and bio-leach liquor obtained by conventional approaches can be followed by precipitation to recover iron (Fe) and Zn, and cementation to recover Cu from the dismissed PCB [16]. Valuable constituents such as Au, Cu and glass fibers (GFs) can be recovered by zero-waste loop strategies which combine swelling in a closed reactor to obtain a mixture that has to be purified by leaching and physical processes (sieving or magnetic separation) [17]. For precious metals such as Au the commonly adopted practices are based on obtaining the metallic phase from salts through inorganic materials that works as adsorbers or nano structural materials (graphene oxide, organosilica frameworks); they have a high adsorbent efficiency thanks to their intrinsic characteristics, such as high surface area and large functional groups density compared to bulk materials [18]. There are also innovative approaches which use an eutectic solvent as a novel and green carrier within an emulsion liquid membrane system [19]. Moreover, gas assisted microflow can also be used during purification to recover Au and Ag [20]. Due to its use in photovoltaic applications recovery of Ga is also important even though it is present in a very low content into electronic boards [21]. New trends are based on automatic systems; in particular, a facility for disassembly and off-gas purification has been used to separate the electronic components from the substrate [22] avoiding hazardous volatile substances.

In managing PCBs desoldering or melting, the removal of the soldered joints is the first essential step to proceed with further recovery of metals from electric and electronic components. It can be carried out by physical desoldering which mainly includes heating a medium (as in thermal convection), infrared and laser heating, or chemical methods [23]. Moreover, the separation of the multi-layer structures of the PCB, after the removal of electrical and electronic components is crucial. Good results have been reached in terms of reduced time and enhanced process efficiency through microwave bath using solvents such as dimethyl formamide or dimethyl acetamide if compared to commonly adopted ultrasonic bath and thermostatic bath [24].

Following the recent trends in the fields of urban mining, this work uses an innovative approach to recover electric and electronic components from PCBs through desoldering by preheating in oven and further impact (Figure 2). Moreover, a thermo-mechanical disassembly strategy, already adopted in previous studies for carbon fiber reinforced composites recycling [25-27], was used for the multilayer structure of the PCBs frame, the copper traces and the soldering alloy recovery. Finally, the recycled Sn was successfully processed to manufacture a bulk ingot.

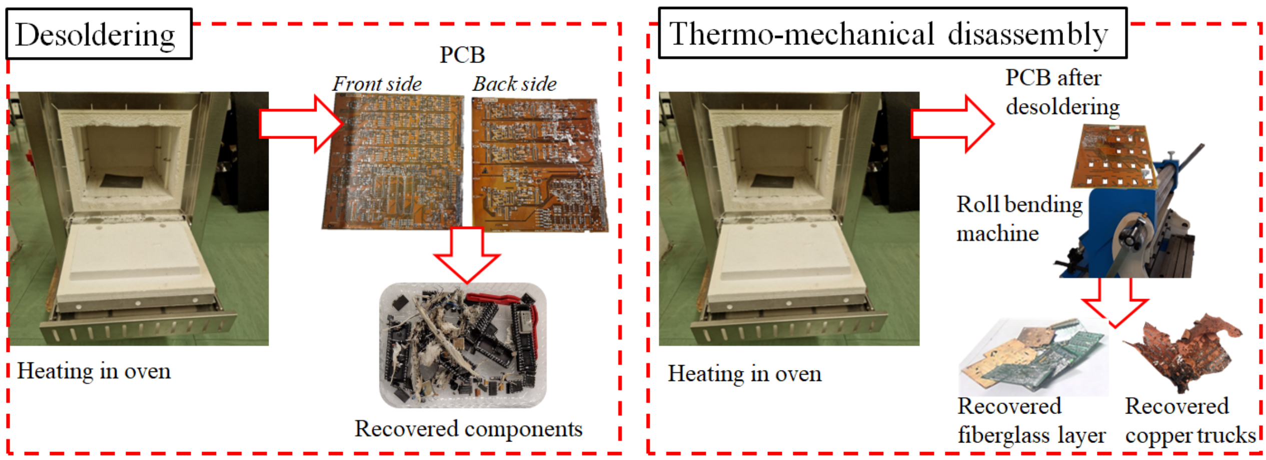

Fig. 2. Desoldering and thermo-mechanical disassembly to recover the constituents of the electronic board.

Materials and Methods

Printed circuit boards. Different kinds of electronic boards were used in the experimental phase with a total amount of 4 kg. They have been collected from electronic devices such as personal computers. All of them were made of 3 main categories of materials: electric and electronic components, soldering alloys and copper traces, and fiberglass laminates for the substrate. In Figure 1 an example of the processed bords is shown.

Desoldering and thermo-mechanical disassembly of PCBs. A two-step process has been carried out to recover the main constituents of the electronic boards (Figure 2). In the first step, the desoldering one, they were heated in a muffle furnace Nabertherm L45/12/B140 (Lilienthal, Germany) for 15 min at 250 °C. During heating, the soldering alloy melted and components were removed through impacts. This procedure allowed us to recover not only electronic components but also the soldering alloy. A maximum of 3 heating cycles in oven were applied to each PCB. In the end, an ingot was obtained by pouring the recovered soldering alloy. Subsequently, the PCBs were disassembled by heating in oven with the same process parameter already adopted in the previous step and then, deformed by a roll bending machine, the rollers of which had a nominal diameter of

30 mm. The applied deformation permitted the separation of the fiberglass layers, exposing the copper traces. Each PCB underwent several heating and deformation cycles.

Results and Discussion

The experimentation has been carried out on a total of 20 boards, different in size (from the minimum of 10x6.5 mm^2 to the maximum of 160x100 mm^2). An oven has been used for PCB heating, at the temperature of 250°C to reduce the processing time for each PCB, and to compensate for the fact that the oven door was continuously opened. To maximize the mass of disassembled components, up to 3 consecutive impacts have been carried out on each single PCB. The impact for desoldering has been produced on a cold metallic plate to have the immediate solidification of the soldering alloy after its removal from the PCB. In Figure 3, the result of the desoldering process is shown in the case of the best component separation. When desoldering is optimal, the alloy is fully removed from all the holes of the PCB, where the electronic components were initially placed. Machining cannot produce this result, leaving consistent contamination on the PCB, as well as on the components.

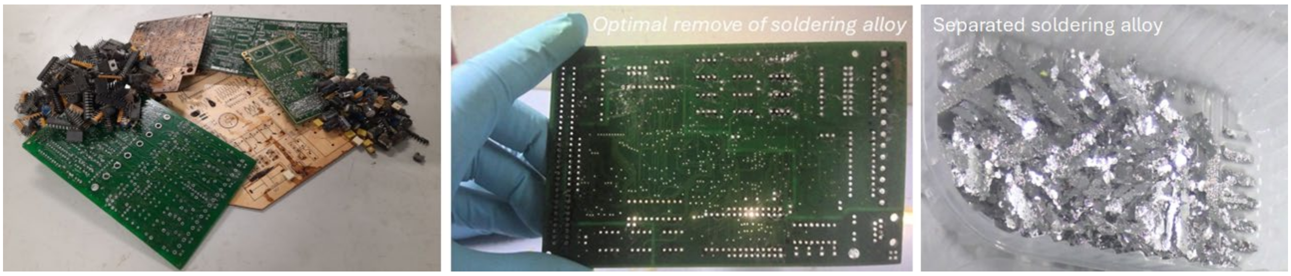

Fig. 3. Separation of electronic components and soldering alloy by impact.

Not all the boards have been optimally desoldered, also after 3 impacts. In the end, 40% of the boards are almost perfectly desoldered, and another 35% are sufficiently treated, but one in four boards, comprising the largest, is only partially desoldered. In badly desoldered PCBs, there are large areas where the components have been completely removed as well, but such portions (from 20 to 50%) are almost unaffected. In the case of large boards, this issue depends on the PCB deformability after heating, and the difficulty of transferring the impact energy on the components. In the end of the desoldering campaign, the processed PCBs have a weight of 2.3 kg, 58.7% of the initial mass. The disassembled components are 1460 g (37.1%), and the soldering alloy is 106 g (2.7%). A mass of about 60 g (1.5%) has been lost during the process for several reasons, such as the ejections of very small components and alloy droplets, or the degradation of such plastics. This mass loss could be reduced by using an automated impact desoldering machine, that is currently under patenting; it would permit an accurate calibration of process parameters, such as force, distance and speed. However, the recovered soldering alloy is particularly clean as shown in Figure 3.

The soldering alloy, separated by impact from the board, has been cast into a sand mold to extract a small ingot (72.4 g). After pouring, a residual of 21.8 g resulted because of slag, impurities, and embedded wires (Figure 4). To conclude the extraction phase by thermo-mechanical processes, copper has been separated from clean waste PCBs by another innovative technique, namely "disassembly", originally applied to the recycling of carbon fiber reinforced laminates. This is a thermo-mechanical process, as well as the impact desoldering, and can separate single fiberglass plies of the PCBs, and copper layers from them. A hand-operated roll bending machine was used for this purpose. In the future, an automated machine will be prototyped in such a way to optimize process parameters like rolls' speed. Good results have been obtained for such PCBs, as large ones were difficult to separate because of difficulties in having homogeneous heating in the laboratory procedure. When possible, copper layers were fully separated from the inside of the waste PCBs as well as copper tracks from the top and the bottom (Figure 5).

Fig. 4. Manufacturing of an ingot with the recovered soldering alloy.

Fig. 5. Copper extraction from waste PCBs by thermo-mechanical disassembly.

Summary

Experimental results show that impact desoldering is able to separate waste PCBs from electronic components, even at laboratory scale and by hand operation. Contamination from the soldering alloy is minimal both on the PCBs and the components, thus enhancing successive recycling steps to recover CRMs or other high value substances. Secondary raw materials have been extracted in the form of an ingot for the soldering alloy, and copper sheets. Further studies will be performed in the future in such a way to increase the level of components separation. Moreover, in-line control will be implemented to increase the amount of recovered CRMs.

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