CHEMICAL ENGINEERING TRANSACTIONS

VOL. 70, 2018

A publication of AIDIC

The Italian Association of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Timothy G. Walmsley, Petar S. Varbanov, Rongxin Su, Jiří J. Klemeš

ISBN 978-88-95608-67-9; **ISSN** 2283-9216

DOI: 10.3303/CET1870046

Metals Recovery from Printed Circuit Boards: The Pursuit of Environmental and Economic Sustainability

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The possible optimization of metal recovery from Printed Circuit Boards (PCBs) and Central Processing Units (CPUs) has been investigated. Usual practice is to primarily recover metals with the highest market price. In contrast, the present work shows how strategic considerations of the value share (%) of metals content and data regarding the environmental impact of their recovery can instruct about the best strategies to adopt, pointing at the metals to be recovered as a priority depending on the case. An accurate PCBs' characterization carried out through microwave digestion with a mixture of HNO_3, HF and HCl, is a first essential step of the procedure. Then metals are recovered through chemical leaching with different chemical substances, exploiting both chemical and physical steps. In particular, a proposal to improve environmental and economic sustainability of the PCBs' treatment is presented, which contrary to expectations first considers the recovery of Cu, Pb and Sn by leaching with 6M HNO_3, followed only later by the recovery of gold and other precious metals. Although unusual, the recovery procedure can be adapted accordingly, allowing more profits, easier management and higher metals recovery rates.

1. Introduction

Large volumes of electronic goods, such as computers and mobile phones, are being disposed every day. Every year about 20 - 50 million tons (Ghosh et al., 2015) of electrical and electronic waste (WEEE) are produced, which makes e-waste management a serious emerging problem. At the same times, it also represents a business opportunity, given the increasing production of WEEE and their content in valuable materials (Ardi and Leister, 2015) that can be recovered making use of suitable strategies (Innocenzi et al., 2018). PCBs in particular are an important part of electronic equipment, accounting for approximately 3 % of the total generated e-waste (Ongondo et al., 2011). They usually contain metals (40 %), organics (30 %), and ceramics (30 %) (Khaliq et al., 2014); due to their composition, the European WEEE Directive (2003) considers them as hazardous materials that need to be processed in an environmentally sustainable manner. Unfortunately, only few Mt of e-waste are formally collected by the take-back system while the rest of the e-waste is in general disposed of by landfilling, incineration or uncontrolled dumping (UNU, 2015). An improper disposal can have severe impacts on both environment and humans; for instance, the undesired migration of the metals towards other matrices may result in several forms of persistent contamination that would require to be properly quantified (Ferrucci et al., 2017) and dealt with (Vocciante et al., 2017) by referring to specific strategies.
Although it is always possible to consider at least a cheap and non-invasive control of water movements (Vocciante et al., 2016a) to monitor of the displacement of contaminants in soil and avoid groundwater contamination by toxic leachate, the possibility to recover raw materials from this type of waste is certainly attractive. Indeed, because of the high demand and the progressive natural resource depletion, the global PCBs' recycling market has experienced a rapid growth in recent decades. However, the heterogeneous mix of organic materials, metals and glass fibers of PCBs makes their management and recycling particularly problematic. On the other hand, PCBs are the most "useful" parts of WEEE as they contain many metals of economic interest, representing over 80 % of the economic value of the PCBs (Terena et al., 2017). Since also mining activity causes effects on environment, the recovery of the metals contained in PCBs becomes a priority, as also set by

Please cite this article as: Pietrelli L., Francolini I., Piozzi A., Vocciante M., 2018, Metals recovery from printed circuit boards: the pursuit of environmental and economic sustainability , Chemical Engineering Transactions, 70, 271-276 DOI:10.3303/CET1870046

the latest European laws that promote the development of processes in line with the strategies of the Circular economy (European Commission, 2017), BAT techniques, and the "near-zero discharge" of hazardous wastes (see, for example, Pietrelli et al., 2018).
So far, several processes for the recovery of valuable materials from PCBs have been developed, which can be broadly categorized as thermal (Hall and Williams, 2007), mechanical (Zangh and Forssberg, 1997), hydrometallurgical (Tuncuk et al., 2012), or as mixed processes (Cui and Zang, 2008). There are both regulatory and economic drivers behind the use of these technologies mainly as consequence of the valuable metals content and environmental limitations. Each of the cited technologies has disadvantages that severely limit their utilization. For example, thermal processes, such as pyrometallurgical technologies, produce dioxins and require high energy (Menad et al., 1998), while hydrometallurgical processes rely on the use expensive and hazardous chemicals.
Regardless of the technology used, most precious metals are commonly considered as those mainly deserving to be recovered, thus the whole process is designed according to this strategy. However, PCBs' characterization can play an important role be pointing out that, beyond the precious metals, other metals having less value but more market, such as Copper, Tin, Nickel and Zinc, can be recovered and recycled as well. This is even more relevant considering that, during the past years, the gold content in PCBs has gradually decreased thanks to the optimization of surface conduction mechanisms. In the '80s, the gold contact layer thickness was in the range 1 - 2.5 µm while today it is between 0.3 - 0.6 µm (Cui and Zhang, 2008), and it is reasonable to hypothesize that it will decrease further in the future.
PCBs' availability also plays a key role in the choice of the treatment process finalized to metal recovery, especially in terms of costs. Indeed, they are considered waste but currently it is not possible to find PCBs available on the Italian market: considering the high price of PCB (5 - 10,000 € t^{-1}) and CPU (10 - 15,000 € t^{-1}) waste (ESG, 2018), the treatment process is significantly influenced by the "raw material" costs.
Therefore, waste management and sustainability of the PCBs' recovery process depend strongly on their nature and raw price. In this study, a characterization of the considered PCBs has been conducted firstly: leaching was performed through microwave digestion with a mixture of HNO_3, HF and HCl, followed by ICP-OES analysis. PCBs' characterization showed that Cu is the most abundant metal (164.9 - 290.1 g kg^{-1}), whereas Au is extremely variable (21.2 - 3,270.1 mg kg^{-1}). Based on metals content evaluation, strategic considerations and strategies about recovery process can be drawn in terms of metals to be recovered as a priority, PCB typology, treatment costs and effective value of metals content.

2. Materials and methods

Whole end-of-life PCBs were collected from a WEEE dismantling center and separated by type as mobile phones (MP), motherboards (MB), modems (M), Central Processing Units (CPU) and mixed PCBs (MIX). Then, the metal content of each class was determined by grinding 10 PCBs per type in particles < 1 mm to ensure a good leaching of metals. Three samples of the obtained powder were weighted, added with a mixture of HNO_3, HF and HCl in the ratio 2:4:6, and the matrix dissolved by using a microwave digestion system (Mileston Start D Microwave) for twenty minutes. HF in particular was needed because aqua regia is ineffective for dissolving SiO_2. After digestion, the leaching solution was evaporated and recovered by HNO_3, and the resulting solutions were analysed by Atomic Absorption Spectrophotometer (Perkin Elmer 5100). To obtain a representative value of the metals content in a real case, a sample consisting of 30 kg of mixed PCBs from a collecting/dismantling operating plant was burnt at 600 °C in inert atmosphere and pulverized in particles < 0.02 cm with a ball mill (RESCH SM2000). Then, by using the above-described procedure, six specimens of the fine powder were solubilized and analyzed. Further experiments were performed by using 5 kg of entire CPU and 7.5 L of 0.5 % cyanide solution (KCN) in the presence of oxygen and adding a commercial chemical catalyst (gold stripper from Patachemicals) to reduce the leaching time. As previously described exhaustively (Pietrelli et al., 2012), leaching from whole PCBs was carried out by adding 5.5 kg of PCBs (from the collection and disassembly center) in a 50 L rotating reactor containing 6M HNO_3 (solid/liquid ratio 1:6) at room temperature. Control of NOx emissions was performed by scrubbing with a solution of NaOH.

3. Results and discussion

The stream composition of the processing plant can be determined only approximately, as it changes day by day; in any case, there is a clear difference between PCB types (Table 1). The majority of PCBs are made by bonding a layer of copper over the entire substrate and removing unwanted copper by etching, leaving only the desired copper traces. Therefore, copper is the main metal component, amounting to 164.9 - 290.1 kg t^{-1} (Guo et al., 2009). In PCBs from mobile phones, the amount of copper is higher than in the other PCBs and the difference can be explained by the extensive use of multi-layered boards in recent years allowing size and weight reduction in mobile phones. The gold content in mixed PCBs was 853 mg kg^{-1}, an intermediate value between contents found in modem (21.3 mg kg^{-1}) and CPU (3,270 mg kg^{-1}). By considering 19 PCBs from mobile phones, Chancerel and Rotter (2009) reported an average weight of Au = 2.2 g $unit^{-1}$, significantly higher than the 0.58 g $unit^{-1}$ obtained in this study (-73.3 %, n = 10). The low gold content found in the mixed PCBs can be due to the "cannibalization" of WEEE (particularly regarding CPU and mobile phones), to the detriment of who deals with disposal and treatment. Therefore, the gold content obtained by processing mixed PCBs might be greater, even if highly variable. Regarding the other metals reported in Table 1, the high silicon content (96.6 - 134.6 kg t^{-1}) is due to the glass fibers, which constitute the support of the PCBs, while Pb, Sn and Ag are contained in the solder paste. Many metals present in PCBs are included in the priority ranking of hazardous substances that could endanger public health or the environment (Comprehensive Environmental Response, Compensation, and Liability Act, CERCLA). In particular, the content in lead and chromium is high, confirming that the treatment and recovery must be performed under safe conditions.

Table 1: Content of valuable metals in PCBs (kg t^{-1}) and economic value (€ t^{-1} at Jun 2017). MP = mobile phone, M = modem, MB = motherboard, CPU = Central process unit, MIX = mixed PCBs. CERCLA = priority ranking of hazardous substances (2013); - = not analyzed; n.i. = not included.

Metal	CERCLA	MP	M	MB	CPU	MIX
Cu	125	290,120	164,950	211,380	166,333	287,300
Zn	74	4,680	11,820	670	-	502
Cd	7	31	360	130	-	360
Ni	53	15,740	14,140	2,810	78,237	6143
Pb	2	14,450	29,010	18,030	-	27,342
Fe	n.i.	31,610	57,580	1,810	-	9,900
Cr	77	1,310	250	70	-	3,620
Si	n.i.	96,610	134,600	103,430	-	110,000
Al	181	19,810	36,230	18,980	-	10,200
Au	n.i.	1,740	21.3	120	3,270	853
Ag	217	1,210	1,760	660	0.7	425
Sn	n.i.	28,540	62,160	33,410	1.0	55,500
Sb	219	-	-	-	-	1,067
Mn	140	13.4	-	-	-	32.6
Pd	171	124.5	-	-	-	250
Pt	n.i.	6.8	-	-	-	12.1
Economic value [€ t^{-1}]		8 - 10,000	1 - 2,500	4 - 5,500	10 - 15,000	-

The type and content of metals in WEEE (gold in particular) can vary greatly depending on equipment age, origin and manufacturer. However, by considering the PCBs' characterization conducted in this study, the value share (V_i) for each metal and each PCB type can be determined by the following equation:

$$V_i = 100 * \frac{W_i * Pr_i}{\sum_i W_i * Pr_i} \quad (1)$$

where W_i is the weight percentage of metal *i* in the PCBs (according to Table 1) and Pr_i represents the current price of the metal according to the London Metal Exchange (October 2017).

Table 2: Calculated value share (%) based on metals content of the PCB classes considered collected from the recycling center. Metal price data were taken from the London Metal Exchange (October 2017).

Metal	MP	MB	CPU	MIX	Metal	MP	MB	CPU	MIX
Cu	75.87	73.30	43.73	85.28	Cr	4.2×10^{-2}	2.2×10^{-6}	-	0.004
Zn	0.005	1.8×10^{-4}	-	6.7×10^{-5}	Si	2.42	5.06	-	3.81
Cd	1.8×10^{-7}	1.8×10^{-6}	-	2.3×10^{-5}	Al	0.09	0.15	-	0.03
Ni	0.5	0.03	-	6.7×10^{-5}	Au	18.8	16.32	56.27	5.48
Pb	0.05	0.15	-	0.11	Ag	0.19	0.11	-	2.9×10^{-3}
Fe	0.04	2.7×10^{-4}	-	0.01	Sn	1.96	4.88	-	5.36

Table 2 shows that gold has 56 % of value share in case of CPUs, while for mobile phones (MP), motherboards (MB) and mixed PCBs (MIX), copper has a higher value share, ranging between 73 % and 90 %. This indicates that the major economic driving force for PCB waste recycling should not be strictly related to the gold content.

Knowing the ecological footprint of a non-renewable resource is needed to evaluate the environmental impact of use and consumption of such resource. Although approximations made in the calculation might affect the accuracy of the obtained outcome, the evaluation is highly encouraged since provides a result (impact number) that is easily understandable and summarizes many relevant factors otherwise difficult to quantify, highlighting aspects often unexpected (Vocciante et al., 2016b).

In Italy, the total amount of recovered WEEE in 2011 was 260,000 t (4.3 kg inhabitant^{-1}), about 32 % of the total WEEE, of which refrigerators (R1) accounted for 68,432 t; washing machines (R2) for 66,132 t; TVs (R3) for 84,274 t; mobile phones, PC, and small devices (R4) for 40,288 t, and lamps (R5) for 962 t (C.C.R., 2011).

Concerning the R4 fraction, 35 M mobile phones and 4.5 M PCs (desktop, notebook, and workstation) are estimated to be sold in Italy every year (Rusconi, 2012). By hypothesizing a 100 % waste rate for the sold PCBs, the amounts of metals potentially recoverable, in Italy, are reported in Table 3. About 850 t y^{-1} of copper (4.4 MUSD) could be recovered, saving 42.1×10^3 t of soil (calculated as total metal on ore concentration); more than 213 kt y^{-1} of soil could be saved by recovering metals from PCBs, 68.8% of which results from the recovery of gold, as shown in Table 3.

Despite the Italian legislation, recyclers are very keen to recycle PCBs and mobile phones, as these devices contain valuable metals. This is probably due to the particular Italian context whose excessive bureaucracy makes it easier to send the PCBs to thermal plants abroad.

Table 3: Metals content in MP and PC, ore concentration (*), ecological burden (metal mined/excavated rock) and soil saved (Total metal/Ore concentration).

Metal	MP		PC		Ore Conc.	Ecological	Soil saved
	Conc. [g t^{-1}]	Total [t]	Conc. [g t^{-1}]	Total [t]	[%]	burden	[t y^{-1}]
Cu	290,120	300.56	211,380	541.3	2	50	42.1x10^3
Zn	4,680	4.85	670	1.71	6.9	14.5	95.1
Cd	31	0.032	130	0.33	40 ppm	25×10^3	9.05×10^3
Ni	15,740	16.31	2,810	7.20	1.2	83.3	1.96×10^3
Pb	14,450	14.97	18,030	46.17	2.9	34.4	2.10×10^3
Fe	31,610	32.75	1,810	4.63	60	1.7	55.71
Cr	1,310	1.36	70	0.18	41	2.5	3.85
Si	96,610	100.09	103,430	264.8	43.7	2.2	802.7
Al	19,810	20.52	18,980	48.6	28.1	3.6	248.8
Au	1,740	1.80	1,200	0.31	15 ppm	67×10^3	141.4×10^3
Ag	1,210	1.26	660	1.69	0.08	1.3×10^3	3.8×10^3
Sn	28,540	29.57	33,410	85.56	10	100	11.5×10^3
Total						9.36×10^3	213.1×10^3

3.1 Process description

Considering both economic growth and environmental sustainability, the treatment of PCBs finalized to metal recovery should be designed and realized in a proper way. Based on the analysis of the characterization data obtained in this work, of experimental tests and the price of the PCBs, a proposal for the PCBs' treatment is schematized in Figure 1. Since each PCB type can be found and bayed separately on the market, it is realistic to design a process consisting of two main lines: CPUs and mixed PCBs.

The expensive CPUs (10 - 15,000 € t^{-1}) can be simply treated with the well-known process of cyanide leaching, followed by metal electro-refining (Cocchiara et al., 2017): as an average, about 2,890 g Au t^{-1} of CPU could be recovered. Following experimental tests, performed with 5 kg of CPU, the process cost (CPU and chemicals purchase, electro-refining and treatment of exhausted solutions) was estimated at 10,800 € t^{-1} CPU.

As for the mixed PCBs, they can be first leached with 6M HNO_3, in order to remove components such as resistors and capacitors: the resulting leaching solution, containing mainly Cu (55.8 g L^{-1}) can be used to recover copper by solvent extraction, ion exchange or selective precipitation (Pietrelli et al., 2012). Copper recovered by leaching is about 50 % of the total content; the remaining amount is located between resin layers, which prevent leaching by HNO_3. Tin is solubilized and recovered as SnO_2/H_2SnO_3 (about 98 % of the total); the recovered Pb is 89.9 % of the total, while leached silver amounts to about 97 %.

The solid residue consists mainly of: i) the board without components (resin, glass and Cu) (51.7 %); ii) a white-colored precipitate of tin oxide plus stannic acid (11.5 %); iii) thin sheets of gold resulting from the dissolution of the Cu or Ni supports (gold is not soluble in HNO_3); iv) components detached from the PCB (capacitors, resistors, transistors, integrated circuits, etc.) (8.1 %); v) slots (28.7 %); vi) a small organic fraction resulting from the decomposition of the surface coating of the PCB.

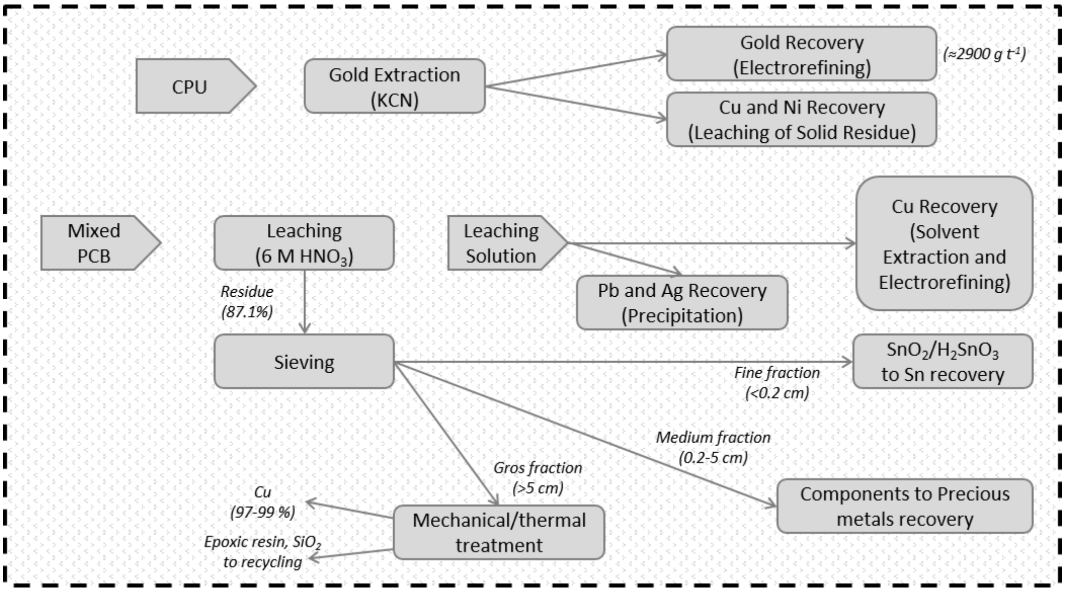

Figure 1: The whole process scheme to recover metals from CPU and mixed PCBs.

The components detached through leaching can be collected, crushed and used again for the recovery of precious metals. By this way, the residue to be treated is c.a. 1/25 of the original material, which allows saving in sizing of equipment and reagents to use. In a hydrometallurgical process, the selective separation of a specific metal is made simpler through the reduction of competing ions and of their concentrations in the leached solution, and also requires less chemicals.

The white precipitate (SnO_2/H_2SnO_3) can be melted in a reducing environment to obtain tin (Pietrelli et al., 2012). PCBs without components can be used to recycle the inner copper by simple mechanical (crushing and sorting) or thermal processes. In fact, a components-free PCB consists solely of epoxy resin, glass fiber and copper (between resin layers). The cost of a mechanical treatment is much lower than that of a hydrometallurgical process, yet the mechanical treatment allows copper recovery with a purity around 98%, at a lower price (5,056 USD t^{-1}) than that obtained via hydrometallurgy (> 99 %, 5,164 USD t^{-1}). Basically, the treatment costs required to obtain Cu with a high degree of purity are greater than the difference between the selling price of the purest and the less pure copper.

The remaining glass fiber plus epoxy resin can be utilized as filler material for polymerization (Pietrelli et al., 2012).

4. Conclusions

In general terms, heterogeneity is the main drawback hampering material recovery from WEEE. However, results show that suitable strategies can be adopted depending on PCB typology, treatment costs and effective value of the metal content. In particular, as copper is present in high amounts (30 % c.a.), it should be considered the main objective of the recovery process (at least for equipment poor in gold, such as Modems or mixed PCBs). Gold recovery may be the first step of the treatment of CPUs, by using the most common extraction processes (KCN, Urea, etc.), whereas, for the other PCBs, it may be recovered (together with other precious metals) from the residue (8.0 %) obtained after leaching with 6M HNO_3. To reduce the chemical consumption, leaching of the entire PCB must be done avoiding the grinding and sieving steps; later, about 50 % of the copper may be recovered from the cleaned PCB through a mechanical or thermal process.

Since PCBs are improperly considered as waste, their characterization recycling oriented may support a rational design, and the environmental and economical sustainability of the recovery/recycling process. By assuming to recover the total content of metals in the sold WEEE, it would be possible to preserve large amount of natural resources, as no excavation would be needed. In this regard, further work is welcomed to optimize the process and highlight the advantages in economic and environmental terms compared to a standard approach.

Acknowledgments

The authors would like to thank Dr. Ing. L.M. Cangeri for his help with the execution of some of the experiments.

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