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Waste solder and printed circuit board: the emerging secondary sources for recovery of metals

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ABSTRACT

Purpose: Purpose: The goal of the paper is to focus on waste solder and printed circuit board: the emerging secondary sources for recovery of metals

Design/methodology/approach: The worldwide reserves of high-grade ores are diminishing. At the same time the demand for heavy metals is ever increasing with the progress of the industrialized world. The rapid progress of electronic packaging technology is resulting in huge amounts of electronic waste (E-Waste) particularly in the form of solders and printed circuit boards (PCBs). Such E-waste contains various metals. The waste solders and PCB can act as large stockpiles of metals. Hence, they can be important secondary sources of valuable metals. Thus recycling of waste solders and PCB is not only useful for resource recovery from waste materials, but also for the protection of the environment.

Findings: Comparing with the pyrometallurgical processing, hydrometallurgical method is more exact, more predictable, and more easily controlled. Bio-hydrometallurgical processes are emerging as potential environmentally friendly approaches

Research limitations/implications: Several promising metal recovery processes were developed to recover the precious metals from E-waste. There is a need to fill the gap areas in achieving a cleaner and economical recycling process. Also more studies are needed in the area of metal separation and recovery from PCB leach liquor.

Orginality/Value: This review article will provide a concise overview of current disposal and recycling operations.

Keywords: Electronic waste; Solder; Printed circuit board; Metal recovery; Pyrometallurgy, Hydrometallurgy; Bio-hydrometallurgy

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MATERIALS

1. Introduction

The Electrical and Electronic Equipment (EEE) producing industries is the fastest growing sectors of manufacturing industry [1-3]. The technological innovations are causing the shorter life expectancy for the electronics. This results in large amounts of electronic waste. There are two important components of EEE-waste: Solders and PCBs [4]. Typical solders account for 4-6% of the total PCB weight. They are used for joining different components in PCBs [4]. Before 2006 the lead (Pb) containing solders was used extensively in the electronic packaging industry. The high toxicity of Pb raised the health and environmental concerns [5]. The directives on European waste EEE and RoHS restricted the use of Pb in electric and electronic equipment. Thus, tin-lead (Sn-Pb) solder has been substituted by various Pb-free solders [6]. The most favorable Pb-free solder alloys are mainly consisted of Sn with small additions of other elements such as silver (Ag), copper (Cu), and zinc (Zn).

However, too excessive of such elements released into the environment poses a risk to ecosystems and human health [6,7]. PCB is the integral component of any electronic equipment. The proportion of waste PCBs is about 3% of electronic waste [8]. The PCBs are made up of the Cu-clad laminate consisting of glass-reinforced epoxy resin. A number of metallic materials, including precious metals are also included on the PCBs. The concentration of precious metals, especially gold (Au), Ag, palladium (Pd) and platinum (Pt) is much higher than their respective primary resources. Every year, 20-50 million tonnes of waste EEE are generated, and the number is growing at an exponential rate [9]. This makes waste EEE an economically attractive urban ore for recycling. Additionally, waste EEE also contains various heavy metals and flame-retardants. Therefore, the landfilling and incineration of such waste pose grave danger to the eco-system [4]. The proper disposal of waste EEE is an important issue, not only because of the large volume of such waste generated, but also because of the metals and toxic substances it contains. This kind of waste can cause soil, air and groundwater contamination, resulting in risks to human health [10-12].

On the contrary, due to the presence of precious metals, the waste EEE can be considered as secondary ores. The recycling of waste EEE through appropriate technologies is emerging as a profitable business [13]. This will help in the preservation of natural resources by decreasing, the demand for virgin materials. The energy required for recovery of metals from waste EEE is low as compared to metal extraction from the ores. This will reduce the mining activities and thereby the greenhouse gas emissions. Also, it will decrease the economic and environmental burdens of disposal [14]. The metal composition of different electronic scraps from literatures is shown in Table 1.

The composition of electronic waste varies considerably with its age, origin and manufacturer. It is not possible to determine the average scrap composition. Hence, the values given as typical averages in the literature are actually representing only a scrap of a certain age and manufacturer [14].

Table 1.
Metal contents of PCBs in various studies

Reference	Metal contents (mg/g)									
	Cu	Pb	Al	Sn	Zn	Fe	Ni	Ag	Au	Pd
Jadhav and Hocheng [71]	0.0705	0.0035	0.0125	0.0063	0.00008	--	0.0001	--	--	--
Arshadi and Mousavi, [69]	0.392	0.015	0.014	--	0.203	0.019	0.0063	0.0001	0.0002	--
Adhapure et al. [72]	536	0.31	--	--	152	--	--	--	--	--
Arshadi and Mousavi, [64]	0.392	0.015	0.014	--	0.203	0.019	0.0063	0.0001	0.0002	--
Xiu et al. [41]	181.9	20.3	--	51.0	32.0	--	--	--	--	--
Yang et al. [60]	253	1.3	--	3.3	--	1.5	0.2	0.007	0.10	--
Liang et al. [68]	126	31	14	--	56	12	24	0.033	0.014	--
Xiang et al. [74]	231	28.9	26.0	18.8	17.5	8.13	1.92	0.217	0.0144	0.00360
Wang et al. [67]	586	2.53	--	--	2.00	--	0.155	--	--	--
Park and Fray [75]	160	20	50	30	10	50	10	1.0	0.25	0.10
Ilyas et al. [66]	85	31.5	7.1	0.0068	80	83	20	0.029	0.012	--
Kinoshita et al. [57]	100	--	--	--	0.08	--	9.7	--	6.7	--
Mecucci and Scott, [24]	72.80	0.972	--	1.253	--	--	0.0094	0.0179	0.0106	0.0906
Brandl et al. [65]	80	20	237	23	26	--	15	--	--	--

Due to the complex composition, waste EEE recycling requires a multidisciplinary approach intended to separate fibers, metals and plastic fractions and reduce environmental pollution [9]. This review is an attempt to present information on recycling methods currently being used to treat waste EEE (solder and PCBs) for resource recovery. The recycling of waste EEE is difficult due to its heterogeneous composition. Several researchers put forth various methods for the recovery of metals from waste EEE, including physical separation processes [15-17] pyrometallurgical processes, hydrometallurgical processes and bio-hydrometallurgical processes [12,14,18-21].

2. Recovery of metals from solders

Various pyrometallurgical processes are in use for recovery of metals from solders. A Pb refining process was used for the Pb containing solders. The solders were first melted to recover the metals from Sn-Pb solders. Then the re-powdering processes were employed [5,22]. Similarly, the melting processes were employed for the Pb-free solders. During this process the solder was melted at 220-230°C. The metal components were recovered using specific gravity difference. However, these processes consume high energy. Also they suffer from the emission of carbon dioxide and harmful gases generated from the combustion of the organic flux. Also, it is not possible to recover metals as individual components using these pyrometallurgical processes [5,22].

The hydrometallurgical routes have been recognized as efficient processes as compared to pyrometallurgical processes, for waste solder recycling. Cheng et al. [6] studied the leaching behavior of heavy metal elements from Sn-Ag-Cu, Sn-Zn, and Sn-Pb solder alloys and their joints. They used H_2SO_4, NaCl and NaOH solutions during this study. It was found that the leaching amount of Sn from solder joints was more than that from solder alloys in most of the test solution conditions. More amounts of Zn and Pb were leached, when Sn-Zn, Sn-Ag-Cu, and Sn-Pb solder alloys and their joints were treated with sulfuric acid (H_2SO_4), sodium chloride (NaCl) and sodium hydroxide (NaOH) solutions. But the amounts of Ag and Cu were relatively small. The leaching amount of Sn from Sn-Zn joint is larger than that from Sn-Pb joint. The NaCl solution was found effective for the leaching of Sn from the Sn-Ag-Cu solder. Rhee and coworkers investigated the feasibility of Sn recovery from Pb frame scrap using sodium hydroxide with sodium persulfate ($Na_2S_2O_8$) as an oxidant [23]. Mecucci and Scott [24] reported the recovery of Sn, Pb and Cu from scrap PCBs using nitric acid (HNO_3), where the contents of Sn and Pb were approximately 0.4 and 0.3 mass%, respectively, in the PCBs (i.e., they were minor components in the PCBs). Yoo et al. [5] used HNO_3 for leaching of metals from Pb free solders. They tried to separate Ag, Cu and Sn. During the process the Sn precipitated as stannic oxide or stannic acid therefore the concentration of Sn was low in all the HNO_3 leaching experiments. The Ag and Cu components were successfully separated from Sn by HNO_3 leaching. They found that more than 99% of the Ag and Cu in the waste Pb-free solder were dissolved using 2 kmol/m^3 HNO_3 under the leaching conditions of 75°C, 100 kg/m^3 pulp density and stirring at 400 rpm for 120 min. Yoo et al. [5] used NaCl and Cu powder for separation of Ag from Cu. The remaining Cu containing solution was subjected to electro-winning for recovery of the Cu. Along with HNO_3 leaching; several other methods have been developed for waste solder recycling. Takahashi and co-workers developed the recycling process of solder paste using organic solvents such as toluene [22]. Other processes involve the use of chemicals such as NaOH and $Na_2S_2O_8$ [5,24]. Though all these methods are effective, use of such chemicals is harmful to the environment. The consumption of high energy and emission of acidic or basic wastewaters is unavoidable [25]. Jadhav et al (study is under consideration) puts forth a suitable and economic alternative with reduced environmental impacts for recovery of metals from Pb free solders. They applied a hydrometallurgical process using acetic acid for recovery of metals from Sn-Cu and Sn-Cu-Ag solders. They reported that the low concentrations of acetic acid are also effective for metal dissolution. This study revealed that organic acids are attractive extracting agents since they are biologically degradable.

The pyro- and hydrometallurgical processes have disadvantages and these processes are not environmentally benign. These processes can be replaced by bio-hydrometallurgical processes [25]. Hocheng et al. [25] reported an environment-friendly bioleaching process for recovery of metals from solders. They used Sn-Cu, Sn-Cu-Ag, and Sn-Pb solders for bioleaching study. They compared the culture supernatants of *Aspergillus niger* and *Acidithiobacillus ferrooxidans* and found that that metal dissolution based on the acidolysis/complexolysis mechanism was more efficient as compared to the oxidation mechanism. The metal removal by *A. niger* culture supernatant was faster for Sn-Cu-Ag solder as compared to other solder types [25]. Considering the toxicity of metals toward microbial cells, Hocheng et al. [25] proposed a two-step bioleaching process. In this process organic acid production is separated from the metal leaching process.

3. Recovery of metals from waste PCBs

Currently, three processes are in use for recycling of waste PCBs. These are pretreatment, physical recycling, and chemical recycling. The PCBs pretreatment stage includes disassembly of the reusable and toxic parts. Then in next step mechanical and/or metallurgical processes are used for the refining waste PCBs. The mechanical recycling system is based on the differences of the physical characteristics of the materials present in waste PCBs. The metal distribution is a function of size ranges therefore the size and shape of particles play crucial roles in mechanical recycling processes. Hence a crushing stage is necessary for further easier management of PCB waste. Except Al, all the other metals are mainly distributed in the fine fractions (< 5 mm). Almost all the mechanical recycling processes have a certain effective size range and mechanical separation processes are performed in a variety of technique [9,26]. After crushing the waste PCB samples, they are subjected to various treatment processes such as shape separation, magnetic separation, density based separation and electric conductivity based separation to recover the metals [9]. In the shape separation process the particles of waste PCB were separated depending on the difference in the particle velocity, the time the particles take to pass through a mesh aperture. Also, it depends on the particle cohesive force to a solid wall and the particle settling velocity in a liquid [9]. During the magnetic separation process a magnetic force is applied to separate the magnetizable particle. Magnetic separators, in particular, low intensity drum separators are widely used for the recovery of ferromagnetic metals from non-ferrous metals and other non-magnetic wastes [9,27]. For the density-based separations the difference in density of the components is the basis of separation. Several methods are employed to separate heavier materials from lighter ones. The materials are separated depending on their different specific gravity and by their relative movement in response to the force of gravity [9,28]. The electric conductivity-based separation separates materials of different electrical conductivity (or resistivity). There are three typical electric conductivity-based separation techniques in use. They are Eddy current separation, corona electrostatic separation and triboelectric separation [9,29]. But all these processes suffer from loss of valuable metals during the separation and dust problems. Therefore, there is a need to develop more efficient processes for recovery of the metals from waste PCBs. The precious and non-ferrous metals are being recovered using pyrometallurgical processes. These processes use high temperatures for recovery of metals. Various pyrometallurgical processes, including incineration, smelting etc. are in use [14,30-32]. In the pyrometallurgical operations the waste PCBS are smelted to form a precious metal-bearing Cu bullion. This Cu bullion is then subjected to electrolytic refining to produce high purity Cu. The precious metals are then recovered from the slimes collected from Cu electro-refining process. In addition to Cu and precious metals, a variety of metals can be extracted in modern smelters [14,30-32]. However, these processes need waste feed containing high concentrations of copper and precious metals. These processes cannot be operated economically for the waste PCBs having low concentration of metals. They are energy intensive. The formation of dioxins and furans due to the presence of halogenated flame retardants in a PCBs is unavoidable. This process leads to the formation of volatile metals and dust as well. Due to these drawbacks the pyrometallurgical processes may present environmental problems [31-33].

Various chemical recycling methods were employed for the recovery of metals from waste PCBs. They include pyrolysis, gasification, combustion, depolymerization by supercritical fluids and hydrometallurgical processes [34]. Pyrolytic treatment involves the thermal decomposition of organic components through destructive distillation in the absence of air. This process yields hydrocarbon liquids and gases, which can be used as fuel or chemical feedstock [34,35]. In a vacuum pyrolysis process low decomposition temperature can be employed. This process reduces the reaction time required for pyrolysis [34,36].

The metal-nonmetal separation was also carried out using supercritical fluids. The supercritical water [37], super critical carbon dioxide [38] and supercritical methanol [39] have been used for separation of metal-nonmetal from PCBs. These supercritical fluids allow organic species; oxygen and water to form a homogeneous phase, resulting in a more efficient oxidation due to the removal of the mass transport limitations. An enhanced oxidation of waste printed circuit boards in the presence of NaOH was shown by Chien et al. [37]. The bromine remained in the liquid phase, whereas the Cu remained in the solid residue as copper oxide and copper hydroxide. In another study Xing and Zhang [40] controlled the temperature, water content and holding time. By controlling these parameters, they achieved complete decomposition of brominated epoxy resins to HBr enriched with water. Hence, they separated glass fibers and Cu efficiently by applying sub- and supercritical water treatment methods respectively. A combination of supercritical degradation with an electrokinetic process was used by Xiu et al. [41] to recover Cu and Pb under optimum conditions. Both the recovery rate and purity of the reclaimed metal ions were shown to be sufficiently high. The lack of formation of hydrocarbons and noxious substances, commonly observed in PCB pyrolysis, strengthens the environmentally-friendly nature of the supercritical fluid recycling process. Moreover, a high content of metals was retained in the solid residue [34].

The laboratory and pilot scale studies based on hydro-metallurgical processes were proposed for the metal recovery from EEE-waste [14,24,42,43]. The hydro-metallurgical processes are more environmentally friendly over pyro-metallurgical processes [14,43]. The cyanide, thiourea and thiosulfate leaching are commonly employed hydro-metallurgical processes for treatment of waste PCBs. Some non-cyanide lixiviants are also used for leaching metals from waste PCBs [44]. A highly efficient recovery of Au and Ag was achieved by a simple process by applying cyanide. Cyanide is largely used lixiviant for Au mining for years because of the low cost and several other advantages over non-cyanide lixiviants. Considering this ability of cyanide several researchers used it for recovery of precious metals from waste PCBs [45]. Quinet et al. [43] used cyanide leaching for recovery of metals from E-waste at pH > 10 and temp 25ºC. They achieved 93, 95 and 99% metal recovery for Ag, Au and Pd respectively. In a commercial cyanide process, Galvastripper Galva (2013) used potassium cyanide (concentration of 6–8%) at 25ºC, pH 12.5, and S/L ratio 1/20. They achieved 60-70% recovery of Au in 2-4 h from the PCB of cell phones [46].

Thiosulphate is emerging as a potential substitute to cyanide for Au recovery. Presence of Cu^{2+} and NH_3 in a thiosulphate solution acts as a catalyst enhancing the Au recovery [47]. Ha et al., [48] used thiosulphate for the first time for recovery of Au from waste PCB. Ficeriova et al. [49] obtained 98 and 93% Au and Ag extraction from thiosulphate leaching of E-wastes in 48 h. They pretreated the E-waste. The thiosulphate leaching of waste material to recover Au and Ag was economical and eco-friendly too [49]. Tripathi et al. [50] conducted thiosulphate leaching. The results obtained showed recovery of 90 and 50% Au and Ag respectively. Petter et al. [46] used PCB of cell phones for recovery of Au and Ag.

Several researchers showed use of thiourea as potential lixiviant for precious metal recovery. The electron pairs between nitrogen and sulfur atoms of thiourea forms a coordination bond with Au and Ag and leach the metal in solution [44]. The leaching yields obtained for Au and Ag from ores/concentrates and wastes using thiourea lixiviant is promising. Also, thiourea is highly selective to precious metals [51]. Separation of metals in alkali solution is difficult and unstable; therefore the leaching is carried out in acidic medium. Few researchers suggested that the addition of ferric ion into thiourea leaching system for Au and Ag recovery is advantageous [52]. Lee et al. [53] obtained complete Au and Ag extraction from thiosulphate leaching of E-wastes. They pretreated the E-waste. They used roasting, size reduction and magnetic separation processes for the treatment of E-waste. Li et al. [54] conducted thiourea leaching changing several parameters such as particle size, thiourea concentration, Fe^{3+} ion concentrations and temperature. The results obtained showed recovery of 90 and 50% Au and Ag respectively. Gurung et al. [51] used PCB for recovery of Au and Cu.

Various mineral acids are useful non-cyanide lixiviants for the extraction of base metals, Cu in particular from waste PCBs [44]. It has been extensively investigated. Table 2 shows the summary of research work carried out by several researchers for extraction of metals using various mineral acids and oxidants. Mecucci and Scott [24] used HNO_3 for Cu and Pb extractions (>95%) from waste PCBs. Madenoglu [55] and Sheng and Etsell [56] used aqua regia (HNO_3/HCl media) for extraction of Cu and Au from PCBs. Few researchers used hydrogen peroxide (H_2O_2) along with inorganic acids. H_2O_2 is a strong oxidant. The combination of H_2O_2 with acids enhances metal extraction. The oxidation reaction (Eq. 8) is highly exothermic (DH0 = 411.2 KJ/Mol) and control of temperature may be needed. Quinet et al., [43]; Deveci et al., [57] and Behnamfard et al. [58] used H_2O_2 in combination with sulfuric acid. They found that the concentration of H_2O_2 and temperature are the most influential factors affecting metal extraction from e-waste. Oh et al. [42] proposed H_2SO_4 leaching of PCBs in the presence of H_2O_2 as a first stage process where Cu, Fe, Zn, Ni and Al were extracted at high recoveries of >95%. In the second stage, they targeted at the recovery of precious metals using ammoniacal thiosulfate ($CuSO_4$-NH_4OH-$(NH_4)_2S_2O_3$) as lixiviant. They used 2 M NaCl solution to leach out Pb. Kinoshita et al. [59] developed a process of selective leaching of Ni and Cu from non-mounted printed wiring boards. They used HNO_3 and studied the leaching performance in terms of various experimental parameters. Initially Ni was selectively leached with 0.1 M HNO_3 solution. The remaining Cu was transferred into 1.0 M HNO_3 solution in the second step of leaching. Kinoshita et al. [59] used LIX984 reagent for selective extraction of Cu. With the advance of the leaching of the base metals, Au flakes were detached spontaneously from the boards and recovered easily in high yield with excellent purity [59]. Yang et al. [60] used shredded particles of waste PCBs for Cu leaching using H_2SO_4 solution using H_2O_2 as an oxidant at room temperature. They investigated the influence of system variables such as H_2SO_4 concentration, amount of H_2O_2 addition, waste PBCs particle size, presence of cupric ion, temperature and time on copper recovery. Yang et al.

[60] suggested that use of shredding pieces of waste PCBs smaller than 1 mm was beneficial for efficient leaching of Cu. Xiu et al. [41] initially treated waste PCBs by supercritical water (SCW). In next step they used HCl for leaching of metals. Two methods of SCW pre-treatment were studied: supercritical water oxidation (SCWO) and supercritical water depolymerization (SCWD). They found that SCW pretreatment significantly enhanced metal leaching.

Bio-hydrometallurgy has been successfully applied and is an emerging commercial technology for metal recovery from waste PCBs [61]. Bioleaching and biosorption are the two forms of bio-hydrometallurgy that are used for metal recovery [62]. In bioleaching microorganisms interact with the metals to dissolve those metals [63]. A variety of microorganisms can participate in the leaching process [64]. Brandl et al. [65] applied *Thiobacillus thiooxidans*, *Thiobacillus ferrooxidans*, *Aspergillus niger*, and *Penicillium simplicissimum* for recovery of metals from electronic waste materials. They incubated the bacteria for 17 days and fungi for 21 days with PCB powder to study the metal recovery process. The mobilization of metals occurred due to the formation of inorganic and organic acids. Initially the high concentration of waste PCB inhibited the microbial growth. However, the adaptation of fungi as well as bacteria helped their growth even at high waste PCB concentrations.

Ilyas et al. [66] selected moderately thermophilic strains to recover the metals from electronic scrap. They applied a mixed consortium of the metal adapted cultures for metal leaching. This study supports pre-adaptation of the microorganisms for enhancing the metal solubilization rates. The PCB powder is alkaline in nature. The addition of sulfur to the leaching medium resists the increase in pH of the medium [66]. This study also provided evidence for the presence of Pb and Sn in the precipitates that appear during bioleaching. In another study, Wang et al. [67] demonstrated the possibility of copper recovery using *Acidithiobacillus ferrooxidans*, *Acidithiobacillus thiooxidans* and their mixture for a bioleaching process. They revealed that the metal recovery is greatly influenced by process variables such as Fe^{3+} concentration, quantity of stock culture added, and pH. Liang et al. [68] developed a novel strategy to increase the bioleaching performance.

They studied the appropriate PCB addition times and amounts of the bioleaching process. Moreover, increased redox potential and lowered pH in the filtrate were observed, suggesting the mechanism involved in enhancing metals recovery in mixed culture. Arshadi and Mousavi [69] showed the simultaneous gold and copper recovery from PCBs.

Table 2.
Comparison of various hydrometallurgical processes applied for metal recovery from waste PCB

References	Lixiviant used	Time (h)	Metals
Jadhav and Hocheng [73]	Hydrochloric acid	22	Cu (100%), Zn (100%), Ni (100%), Sn (100%), Pb (100%), Fe (100%), Al (100%), Ag (100%), Pd (100%), Au (100%)
Xiu et al. [41]	Hydrochloric acid + Supercritical water	2.5	Cu (99%), Zn (90%), Sn (90%), Sr (90%), Cd (90%), Pb (80%)
Behnamfard et al. [58]	Sulfuric acid + Hydrogen peroxide	3	Cu (99%)
Yang et al. [60]	Sulfuric acid + Hydrogen peroxide	3	Cu (90%)
Deveci et al. [57]	Sulfuric acid + Hydrogen peroxide	4	Cu (>98%)
Lee et al. [53]	(ii) Aqua regia leaching (iv) H_2SO_4 leaching	Not specified	(ii) Au (100%), Ag (88.51%),Cu(100%) (iv) Au (6.05%), Ag (90.37%), Cu (100%)
Park and Fray [75]	Aqua-regia	3	Zn (100%), Ni (100%), Au (100%), Pd (7.8%), Ag (7.4%)
Madenoglu [55]	HNO_3/HCl leaching	Not specified	Au, Cu
Quinet et al. [43]	(i) For Pd recycling; HCl/NaCl with HNO_3/H_2O_2 leaching	Not specified	(i) Pd (93–95%)
Kinoshita et al. [57]	Nitric acid	24-78	Ni (99%), Cu (98%), Au (98%)
Mecucci and Scott, [24]	Nitric acid	6	Cu (100%), Sn (100%), Pb (100%)

Table 3.
Advantages and disadvantages of various processes for recovery of metals from waste PCBs

References	Advantages	Disadvantages
Bioleaching process	1. It avoids use of toxic chemicals. 2. Generation of secondary waste is lower. 3. The process is environmentally friendly.	1. The process is often time consuming. 2. The establishment of economical industrial plants is not possible yet. 3. Maintenance of an acidic environment is prerequisite for bioleaching to keep metals in solution. 4. The occurrence of precipitate is common which creates problems for the purification of leached metals. 5. Need to use low pulp density. 6. The metal toxicity towards microorganisms creates problems.
Physical/mechanical separation process	1. The objective of physical recycling process is to recover the nonmetallic materials as it is, without any loss of valuable metals. 2. Less environmental hazards. 3. Low energy consumption and easy operation.	1. The efficiency of the separation is poor as the particle shape and size play a crucial role in the separation.
Pyrometallurgical process	1. Its ability to accept any forms of scrap.	1. Integrated smelters cannot recover aluminum and iron as metals, since they transfer into the slag. 2. Formation of dioxins is unavoidable. 3. Ceramic components and glass in the e- wste increase the amount of slag from blast furnaces, which thereby increases the loss of precious metals and base metals from the scrap. 4. Only partial separation of metals can be achieved and hydrometallurgical techniques and/or electrochemical processing are subsequently necessary. 5. Precious metals stay for a long time in the pyrometallurgical process and are obtained at the very end of the process.
Hydrometallurgical Process	1. It is a fast process. 2. All types of metals can be recovered. 3. High metal recovery is possible. 4. Suitable for small scale applications.	1. Generation of secondary atmospheric pollution is unavoidable. 2. Needs large amount of energy and chemicals.

For this purpose in first step they applied *Acidithiobacillus ferrooxidans* for copper recovery. In the second step a pure culture of *Bacillus megaterium*, a cyanogenic bacterium, was used to produce cyanide as a leaching agent for gold recovery. Arshadi and Mousavi [64] and Arshadi and Mousavi [70] carried out a multi objective optimization of a bioleaching process for recovery of copper and nickel from mobile phone PCBs.

They selected two modified quadratic models to predict the interactions and behavior of the influencing parameters on the bioleaching of Cu and Ni from e-waste samples. Recently Jadhav and Hocheng [71] developed an enzymatic bioleaching process for recovery of metals from PCB powder. They used a glucose oxidase enzyme to execute the bio-Fenton process. Metal solubilization was achieved through this bio-Fenton process.

The advantages and disadvantages of physical/mechanical, pyrometallurgical, hydrometallurgical and bio-hydrometallurgical processes are described in Table 3. The choice of method depends on the type of E-waste and the metal content. Most of the above-mentioned reports used powdered sample/pulverized sample of waste PCBs for metal recovery. However, little is known about a use of large pieces of PCBs for recovery of metals. The effect of above mentioned leaching reagents on PCB piece during the hydrometallurgical recycling process is not reported yet. Recently, two studies explained the metal recovery from large pieces of PCBs. After complete recovery of metals, the remaining board (nonmetallic part) can be easily recycled; which otherwise difficult while using pulverized PCBs [72,73]. The precipitate formation is common in metal recovery processes from pulverized PCB samples, especially during the bioleaching process [72,73]. The occurrence of such precipitate makes it difficult to distinguish between the precipitate and residual PCB powder and make the overall metal recovery process more complicated. This can be avoided by using large pieces of PCBs [72,73]. Adhapure et al. [72] and Jadhav and Hocheng [73] pre-treated waste PCBs to remove chemical coating present on the PCBs. Then the PCBs were subjected to bioleaching [72] and acid leaching [73] process for metal recovery. Adhapure et al. [72] used a mixed bacterial culture for recovery of metals from large PCB pieces. They required 10 days for removal of metals. Jadhav and Hocheng [73] tried various inorganic and organic acids for recovery of metals from large PCB pieces. They reported that hydrochloric acid was more effective for the recovery of metals as compared to other acids tested. If developed properly, this process has future commercial application.

4. Conclusions

The heterogeneous composition and hazardous material contents of waste EEE necessitate the proper recycling of this waste. Besides the hazardous substances, a lot of valuable materials are contained in waste EEE, make them worth being recycled. The precious metals make up more than 70% of the value and copper 20%. The metals present in waste EEE is in highly pure form. Their recovery will reduce the demand of virgin materials and thereby reduce the mining activities. Therefore, developing a non-polluting, efficient, and low-cost processing technology for recycling of waste EEE cannot only avoid environmental pollution, but also help recycle valuable resources. A disassembly stage is always required to remove dangerous components. Crushing and separation is then carried out for improving successful further treatments. Traditionally, recovering of valuable metals by waste PCBs was carried out on a large scale for positive economic revenue. Physical/mechanical recycling is a promising recycling method without environmental pollution and with reasonable equipment invests, and diversified potential applications of products. However, low recovery of metals limits its application. Comparing with the pyrometallurgical processing, hydrometallurgical method is more exact, more predictable, and more easily controlled. Bio-hydrometallurgical processes are emerging as potential environmentally friendly approaches. There is a need to fill the gap areas in achieving a cleaner and economical recycling process. Also more studies are needed in the area of metal separation and recovery from PCB leach liquor.

Additional information

Selected issues related to this paper are planned to be presented at the 22nd Winter International Scientific Conference on Achievements in Mechanical and Materials Engineering Winter-AMME'2015 in the framework of the Bidisciplinary Occasional Scientific Session BOSS'2015 celebrating the 10th anniversary of the foundation of the Association of Computational Materials Science and Surface Engineering and the World Academy of Materials and Manufacturing Engineering and of the foundation of the Worldwide Journal of Achievements in Materials and Manufacturing Engineering.

References

[1] J. Cui, E. Forssberg, Mechanical recycling of waste electric and electronic equipment: a review, Journal of Hazardous Materials 99 (2003) 243-263.
[2] R. Murugan, Milling and separation of the multi-component printed circuit board materials and the analysis of elutriation based on a single particle model, Powder Technology 183 (2008) 169-176.
[3] J. Guo, J. Guo, Z. Xu, Recycling of non-metallic fractions from waste printed circuit boards: A review, Journal of Hazardous Materials 168 (2009) 567-590.
[4] B. Ghosh, M. Ghosh, P. Parhi, P. Mukherjee, B. Mishra, Waste Printed Circuit Boards recycling: an extensive assessment of current status, Journal of Cleaner Production 94 (2015) 5-19.
[5] K. Yoo, J. Lee, K. Lee, B. Kim, M. Kim, S. Kim, B. Pandey, Recovery of Sn, Ag and Cu from Waste

Pb-Free Solder Using Nitric Acid Leaching, Materials Transactions 53 (2012) 2175-2180.
[6] Q. Cheng, F. Yang, J. Zhao, L. Wang, X. Li, Leaching of heavy metal elements in solder alloys, Corrosion Science 53 (2011) 1738-1747.
[7] S. Lim, J. Schoenung, Human health and ecological toxicity potentials due to heavy metal content in waste electronic devices with flat panel displays, Journal of Hazardous Materials 177 (2010) 251-259.
[8] L. Flandinet, F. Tedjara,b, V. Ghettac, J. Fouletier, Metals recovering from waste printed circuit boards (WPCBs) using molten salts, Journal of Hazardous Materials 213-214 (2012) 485-490.
[9] J. Sohaili, S. Muniyandi, S. Mohamad, A Review on Printed Circuit Board Recycling Technology, Journal of Emerging Trends in Engineering and Applied Sciences 3 (2012) 12-18.
[10] K. Brodersen, D. Tartler, H. Bergmann, et al. The ASM European Office (ed) Proceedings of the 1992 conference on the recycling of metals, the European Council of ASM International and its Technical Committee. ASM, Belgium, 45-51 (1992).
[11] C. LaCoursiere Electronic waste recovery business (E-128). Business Communication Company Inc. (BCC), Norwalk, CT, USA (2005).
[12] J. Lee, H. Song, J. Yoo, Present status of the recycling of waste electrical and electronic equipment in Korea, Resources Conservation and Recycling 50 (2007) 380-397.
[13] A. Marques, J. Marrero, C. Malfatti, A review of the recycling of non-metallic fractions of printed circuit boards, SpringerPlus 2 (2013) 521.
[14] J. Cui, L. Zhang, Metallurgical recovery of metals from electronic waste: a review, Journal of Hazardous Materials 158 (2008) 228-256.
[15] P. Mou, L. Wa, D. Xiang, J. Gao, G. Duan, A Physical Process for Recycling and Reusing Waste Printed Circuits Boards 2 (2004) 237-242.
[16] D. Xiang, P. Mou, J. Wang, G. Duan, H. Zhang, Printed circuit board recycling process and its environmental impact assessment, International Journal of Advanced Manufacturing Technology 34 (2007) 1030-1036.
[17] J. Li, H. Lu, J. Gio, Z. Xu, Y. Zhou, Recycle technology for recovering resources and products from waste printed circuit boards, Environmental Science and Technology 41 (2007) 1995-2000.
[18] J. Li, P. Shrivastava, Z. Gao, H. Zhang, Printed circuit board recycling: a state of the art survey, IEEE Transactions Electronics Packaging and Manufacturing 27 (2004) 33-42.
[19] Z. Gao, J. Li, H. Zhang, Printed circuit board recycling: a state-of-art survey, in: IEEE International Symposium on Electronics and the Environment, SanFrancisco, CA, (2002) 234-241.
[20] S. Chatterjee, K. Kumar, Effective electronic waste management and recycling process involving formal and non-formal sectors, International Journal of Physical Science 4 (2009) 893-905.
[21] T. Havlik, D. Orac, M. Petranikova, A. Miskufova, F. Kukurugya, Z. Takakova, Leaching of copper and tin from used printed circuit boards after thermal treatment, Journal of Hazardous Materials 183 (2010) 866-873.
[22] H. Takahashi, T. Tanaka, M. Hamada and K. Tohji, 10th Int. Symp. On East Asian Resource Recycling Technology (2009) 761-764.
[23] K. Rhee, J. Lee, C. Lee, D. Yang, K. Chung, Proc. 2nd Int. Symp. Metall. Process. the Year 2000 and beyond and the 1994 TMS Extraction and Process Metallurgy Meeting 2 (1994) 515-526.
[24] A. Mecucci, K. Scott, Leaching and electrochemical recovery of copper, lead and tin from scrap printed circuit boards, Journal of Chemical Technology and Biotechnology 77 (2002) 449-457.
[25] H. Hocheng, T. Hong, U. Jadhav, Microbial Leaching of Waste Solder for Recovery of Metal, Applied Biochemistry Biotechnology 173 (2014) 193-204.
[26] J. Li, H. Duan, K. Liu, S. Wang, Characteristic of low-temperature pyrolysis of printed circuit boards subjected to various atmosphere, Resources Conservation and Recycling 54 (2010) 810-815.
[27] J. Svoboda, T. Fujita, Recent developments in magnetic methods of material separation, Minerals Engineering 16 (2003) 785-792.
[28] B. Wills, Mineral Processing Technology, 4th ed., Pergamon Press, Oxford, England, 1988, 377-381.
[29] R. Meier-Staude, K. Koehnlechner, Elektrostatische trennung von leiter / nichtleitergemischen in der betrieblichen praxis (electrostatic separation of conductor/non-conductor mixtures in operational practice), Aufbereitungs-Technik 4 (2000) 118-123.
[30] C. Hageluken, Recycling of electronic scrap at Umicore precious metals refining, Acta Metallurgica Slovaca 12 (2006) 111-120.
[31] Umicore, Precious Metals Refining (Ed.), 2011. Precious Metals Refining Process, Germany, <http://www.preciousmetals.umicore.com/PMR/Process> (retrieved 27.07.11).
[32] A. Tuncuk, V. Stazi, A. Akcil, E. Yazici, H. Deveci, Aqueous metal recovery techniques from e-scrap: Hydrometallurgy in recycling, Minerals Engineering 25 (2012) 28-37.

[33] C. Hageluken, Improving metal returns and eco-efficiency in electronics recycling - a holistic approach for interface optimisation between preprocessing and integrated metals smelting and refining. In; IEEE International Symposium on Electronics & the Environment, 8-11 May, San Francisco (2006) 218-233.

[34] P. Hadi, C. Ning, W. Ouyang, M. Xu, et al., Toward environmentally-benign utilization of nonmetallic fraction of waste printed circuit boards as modifier and precursor, Waste Management 35 (2015) 236-246.

[35] Y. Zheng, Z. Shen, S. Ma, C. Cai, X. Zhao, Y. Xing, B. Guo, X. Zeng, L. Wang,. Influence of the recycled glass fibers from nonmetals of waste printed circuit boards on properties and reinforcing mechanism of polypropylene composites, Journal of Applied Polymer Science 118 (2010) 2914-2920.

[36] Y. Zhou, K. Qiu, A new technology for recycling materials from waste printed circuit boards, Journal of Hazardous Materials 175 (2010) 823-828.

[37] Y. Chien, H. Wang, K. Lin, Y. Yang, Oxidation of printed circuit board wastes in supercritical water, Water Research 34 (2000) 4279-4283.

[38] S. Sanyal, Q. Ke, Y. Zhang, T. Ngo, J. Carrell, H. Zhang, Understanding and optimizing delamination/recycling of printed circuit boards using a supercritical carbon dioxide process, Journal of Cleaner Production 41 (2013) 174-178.

[39] F. Xiu, F. Zhang, Materials recovery from waste printed circuit boards by supercritical methanol, Journal of Hazardous Materials 178 (2010) 628-634.

[40] M. Xing, F. Zhang, Degradation of brominated epoxy resin and metal recovery from waste printed circuit boards through batch sub/supercritical water treatments, Chemical Engineering Journal 219 (2013) 131-136.

[41] F. Xiu, Y. Qi, F. Zhang, Recovery of metals from waste printed circuit boards by supercritical water pre-treatment combined with acid leaching process, Waste Management 33 (2013) 1251-1257.

[42] C. Oh, S. Lee, H. Yang, T. Ha, M. Kim, Selective Leaching of Valuable Metals from Waste Printed Circuit Boards, Journal of the Air and Waste Management Association 53 (2003) 897-902.

[43] P. Quinet, J. Proost, A. Van Lierde, Recovery of precious metals from electronic scrap by hydro-metallurgical processing routes, Minerals Metallurgical Process 22 (2005) 17-22.

[44] A. Akcil, C. Erust, C. Gahan, M. Ozgun, et al., Precious metal recovery from waste printed circuit boards using cyanide and non-cyanide lixiviants-A review, Waste Management (2015) (in print).

[45] C. Flynn, S. McGill, Cyanide Chemistry-Precious Metals Processing and Waste Treatment. US Bureau of Mines, NTIS Publication, PB96-117841 (1995).

[46] P. Petter, H. Veit, A. Bernardes, Evaluation of gold and silver leaching from printed circuit board of cellphones, Waste Management 34 (2014) 475-482.

[47] M. Jeffrey, P. Breuer, W. Choo, A kinetic study that compares the leaching of gold in the cyanide, thiosulfate and chloride systems, Metallurgical and Materials Transaction B 32 (2001) 979-986.

[48] V. Ha, J. Lee, J. Jeong, H. Haia, M. Jha, Thiosulfate leaching of gold from waste mobile phones, Journal of Hazardous Materials 178 (2010) 1115-1119.

[49] J. Ficeriova, P. Balaz, E. Gock, Leaching of gold, silver and accompanying metals from circuit boards (PCBs) waste, Acta Montanistica Slovaca 16 (2011) 128-131.

[50] A. Tripathi, M. Kumar, D. Sau, A. Agrawal, S. Chakravarty, T. Mankhand, Leaching of gold from the waste mobile phone printed circuit boards (PCBs) with ammonium thiosulphate, International Journal of Metallurgical Engineering 1 (2012) 17-21.

[51] M. Gurung, B. Adhikari, H. Kawakita, K. Ohto, K. Inoue, S. Alam, Recovery of gold and silver from spent mobile phones by means of acidothiourea leaching followed by adsorption using biosorbent prepared from persimmon tannin, Hydrometallurgy 133 (2013) 84-93.

[52] N. Gonen, E. Korpe, M. Yildirim, U. Selengil, Leaching and CIL processes in gold recovery from refractory ore with thiourea solutions, Minerals Engineering 20 (2007) 559-565.

[53] C. Lee, L. Tang, S. Popuri, A study on the recycling of scrap integrated circuits by leaching, Waste Management and Research, 29 (2010) 1-9.

[54] J. Li, X. Xu, W. Liu, Thiourea leaching gold and silver from the printed circuit boards of waste mobile phones, Waste Management 32 (2012) 1209-1212.

[55] H. Madenoglu, Recovery of Some Metals from Electronic Scrap. Ege University, Graduate School of Natural and Applied Sciences, Department of Chemical Engineering, Master Thesis, 2005.

[56] P. Sheng, T. Etsell, Recovery of gold from computer circuit board scrap using aqua regia, Waste Manage Res: 25 (2007) 380-383.

[57] H. Deveci, E. Yazıcı, U. Aydın, R. Yazıcı, A. Akcil, Extraction of copper from scrap TV boards by sulphuric acid leaching under oxidising conditions, Proceedings of the Going Green - CARE INNOVATION 2010 Conference, 8-11 November, Vienna, Austria, 2010, 45.

[58] A. Behnamfard, M. Salarirad, F. Veglio, Process development for recovery of copper and precious metals

from waste printed circuit boards with emphasize on palladium and gold leaching and precipitation, Waste Management 33 (2013) 2354-2363.
[59] T. Kinoshita, S. Akitaa, N. Kobayashib, S. Niib, F. Kawaizumib, K. Takahashi, Metal recovery from non-mounted printed wiring boards via hydrometallurgical processing, Hydrometallurgy 69 (2003) 73-79.
[60] H. Yang, J. Liu, J. Yang, Leaching copper from shredded particles of waste printed circuit boards, Journal of Hazardous Materials 187 (2011) 393-400.
[61] J. Pradhan, S. Kumar, Metals bioleaching from electronic waste by Chromobacterium violaceum and Pseudomonads sp, Waste Management and Research 30 (2012) 1151-1159.
[62] K. Huang, J. Guo, Z. Xu, Recycling of waste printed circuit boards: a review of current technologies and treatment status in china, Journal of Hazardous Materials 164 (2009) 399-408.
[63] F. Gerayeli, F. Ghojavand, S. Mousavi, S. Yaghmaei, F. Amiri, Screening and optimization of effective parameters in biological extraction of heavy metals from refinery spent catalysts using a thermophilic bacterium, Separation and Purification Technology 118 (2013) 151-161.
[64] M. Arshadi, S. Mousavi, Simultaneous recovery of Ni and Cu from computer-printed circuit boards using bioleaching: Statistical evaluation and optimization. Bioresource Technology 174 (2014) 233–242.
[65] H. Brandl, R. Bosshard, M. Wegmann, Computer-munching microbes: metal leaching from electronic scrap by bacteria and fungi, Hydrometallurgy 59 (2001) 319-326.
[66] S. Ilyas, M. Anwar, S. Niazi, M. Ghauri, Bioleaching of metals from electronic scrap by moderately thermophilic acidophilic bacteria, Hydrometallurgy 88 (2007) 180-188.
[67] J. Wang, J. Bai1, J. Xu, B. Liang, Bioleaching of metals from printed wire boards by Acidithiobacillus ferrooxidans and Acidithiobacillus thiooxidans and their mixture, Journal of Hazardous Materials 172 (2009) 1100-1105.
[68] G. Liang, Y. Mo, Q. Zhou, Novel strategies of bioleaching metals from printed circuit boards (PCBs) in mixed cultivation of two acidophiles, Enzyme and Microbial Technology 47 (2010) 322-326.
[69] M. Arshadi, S. Mousavi, Enhancement of simultaneous gold and copper extraction from computer printed circuit boards using Bacillus megaterium, Bioresource Technology 175 (2015) 315-324.
[70] M. Arshadi, S. Mousavi, Multi-objective optimization of heavy metals bioleaching from discarded mobile phone PCBs: Simultaneous Cu and Ni recovery using Acidithiobacillus ferrooxidans, Separation and Purification Technology 147 (2015) 210-219.
[71] U. Jadhav, H. Hocheng, Enzymatic bioleaching of metals from printed circuit board, Clean Technology and Environmental Policy 17 (2015) 947-956.
[72] N. Adhapure, P. Dhakephalkar, A. Dhakephalkar, V. Tembhurkar, A. Rajgure, A. Deshmukh, Use of large pieces of printed circuit boards for bioleaching to avoid 'precipitate contamination problem' and to simplify overall metal recovery, MethodsX 1 (2014) 181-186.
[73] U. Jadhav, H. Hocheng, Hydrometallurgical Recovery of Metals from Large Printed Circuit Board Pieces, Scientific Reports (In Press).
[74] Y. Xiang, P. Wu, N. Zhu, T. Zhang, Wen Liud, Jinhua Wu, Ping Li, Bioleaching of copper from waste printed circuit boards by bacterial consortium enriched from acid mine drainage, Journal of Hazardous Materials 184 (2010) 812-818.
[75] Y. Park, D. Fray, Recovery of high purity precious metals from printed circuit boards, Journal of Hazardous Materials 164 (2009) 1152–1158.