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RECOVERY OF COPPER FROM PRINTED CIRCUIT BOARDS OF COMPUTERS

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Abstract
Electronic waste contains many types of metals that can be recovered**.** In this study the recovery of gold as a precious metal and copper as a base metal from printed circuit boards (PCBs) of consumed computers was investigated.The results of chemical analysis of (PCBs) sample, after preparation by manual separation, cutting, grinding, and sieving, showed that it contain; copper (36.268%). Hydrometallurgical method by leaching and precipitation process was applied for copper recovery. The parameters for leaching by nitric acid were; temperatures (50, 60, 70 and 80 ^{o}C), times (5, 10, 15 and 20 min), solid to liquid ratio (1:20, 1:25, 1:30 and 1:35), and at constant stirring speeds (400 rpm). The optimum parameters resulted were 50 ^{o}C temperature, 5 min time and 1:20 S:L, and the leaching efficiencies of copper 59.7% was achieved with these optimum conditions. Copper precipitation, from nitric acid solution, were conducted using pure zinc powder at room temperature without stirring for 24 hour. Recovery efficiency of copper 99.93% was achieved

Introduction
In recent decades, with the accelerating technological development on daily basis, the quantity of electrical and electronic products has increased exponentially and in the form of new and innovative products. At the same time, device consumption rate increased and average life-time of the products has decreased. All this led to the presence of large quantities of electrical and electronic waste. International organizations, such as organization for economic corporation and development (OECD), estimated the amount of electronic waste materials produced globally in 2014 has been 50 million tons. The electronic waste materials represent about 1% - 3% which estimated about 1,636 million tons of the produced waste annually in the world[1]. Generation of electronic waste in united states from 2000 - 2012 shown in figure (1) [2].The increase in their quantities required wide area to dump, as well as its harmful to human health and underground water and soil. Many countries have do more attention and worked to the treatment of electronic waste to preserve the environment and take advantage of the metals and materials in them through the recycling of these materials and recover what is possible [3]. The toxic materials that present in electronic waste such as mercury, lead, chromium and cadmium [4]. Recycling of electronic waste have three major advantages (1)Energy and Resource Conservation, (2)Environmental Concerns and (3)Economic Value of Selected valuable metals(PMs and BMs)[5]. Electronic waste consists of different types of valuables metals metals especially copper, aluminum, gold, silver and palladium. All these metals are connected, covered with or mixed with different kinds of plastic materials and materials of the ceramics [6]. Some parts, such as printed circuit boards (PCBs) that commonly used in different pieces of electronic and electrical equipment and represents good percent by weight of many devices, contains a large quantity of valuables metals (copper, aluminum, gold, silver and palladium), as well as other materials such as polymers. Containment of these components in PCBs makes them difficult to processing, separation and extraction [7].Now a days, there are different techniques and processes to recycle and extraction of valuables metals such as copper and gold from PCBs based on physical methods, such as screening, magnetic separation, density-based Separations, eddy current separation and shaking table separation or chemical methods such as pyro metallurgy, hydrometallurgy, bioleaching or a combination of these techniques, followed by purification and refining[8]. All of these processes involve a series of steps, started with the collection and dismantling of waste, sorting and isolating parts containing toxic and hazardous substances and elements, and then cutting (shredding) and grinding (milling) the remaining parts. Hydro-metallurgical process include acid leaching of electronic waste material. Different acid used as leaching solvent such as; hydrochloric acid and nitric acid [9]. In 2014, the amount of electronics waste generated in Iraq was estimated at about 112,000 tons (3.1 kg per inhabitant) [1]. In Iraq, there is low awareness about e-waste materials, there is no information about it's collection and recycling. This study, as an initial step, focusing on acid leaching as an assistance on studying the electronic waste recycling and management in Iraq. The aims and scope of the study are summarized by the

following: (1)Recovery of copper metals from printed circuit boards waste of computers by hydrometallurgical process. (2) Determining the best operational parameters of hydrometallurgical process (temperature, time, and solid/liquid ratio).

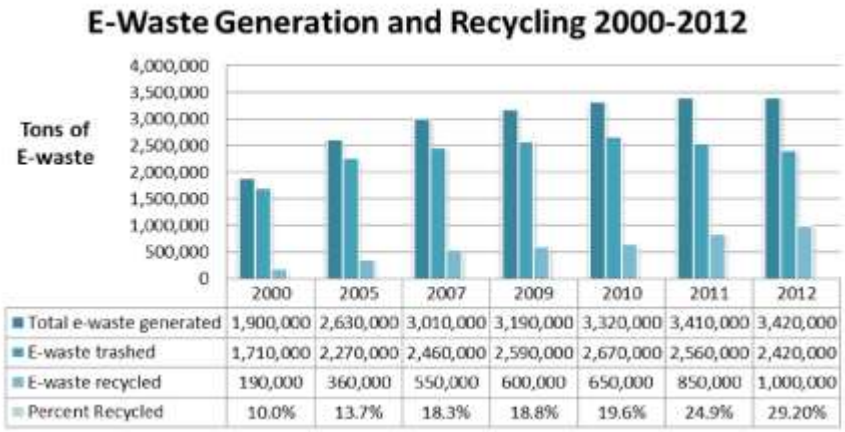

Figure (1) Generation of E-waste from 2000-2012 in united state

Experimental Work

In this study, PCBs of the consumed computers were used for recovery of the copper. The printed circuit boards for this work was obtained from consumed computers found in computers maintenance centers in local markets. The parts of copper that coated with gold in these printed circuit boards was manually separated to prepare it for the subsequent processes. Figure (2) illustrate flow diagram for recovery of copper from printed circuit of computers. Combination process is to be a two-stages, cutting, and grinding to get on the suitable particle size. To get rid of plastic parts, the powder from grinding stage was washed with distilled water in a container and leave for a day where the plastic parts are floated and removed. The residue powder placed in an electric oven at 100 ˚C for 30 min to remove moister. The dried powder was sieved to particle size less than (150) µm by using auto sieve shaker model (AD60-01). The chemical analyses of the PCBs powder is carried out by flame atomic absorption spectrometry (NOVAA-350).The analysis results are shown in table (1)as weight percentage.

Table 1,The chemical analysis for the printed circuit boards powder

Components	**Au**	**Ag**	**Cu**	**Sn**
Wt.%	**0.0327%**	**0.0111%**	**36.268%**	**0.0430%**

The samples of PCBs powder leaching with nitric acid in concentration 24% to dissolved copper and reduce the concentration of some metals such as silver and tin.

For leaching process, three parameters were studied in this work, temperature, time and solid to liquid ratio. For each parameters four level was selected for leaching copper with nitric acid as shown in table 2.

Table 2, Process parameters with four levels used in leaching process of copper by nitric acid.

Controlled factors	**Levels**		
	L1	L2	L3
Temp(°C)	50	60	70
Time (min)	5	10	15
Solid: Liquid Ratio (wt:wt)	1:10	1:20	1:30

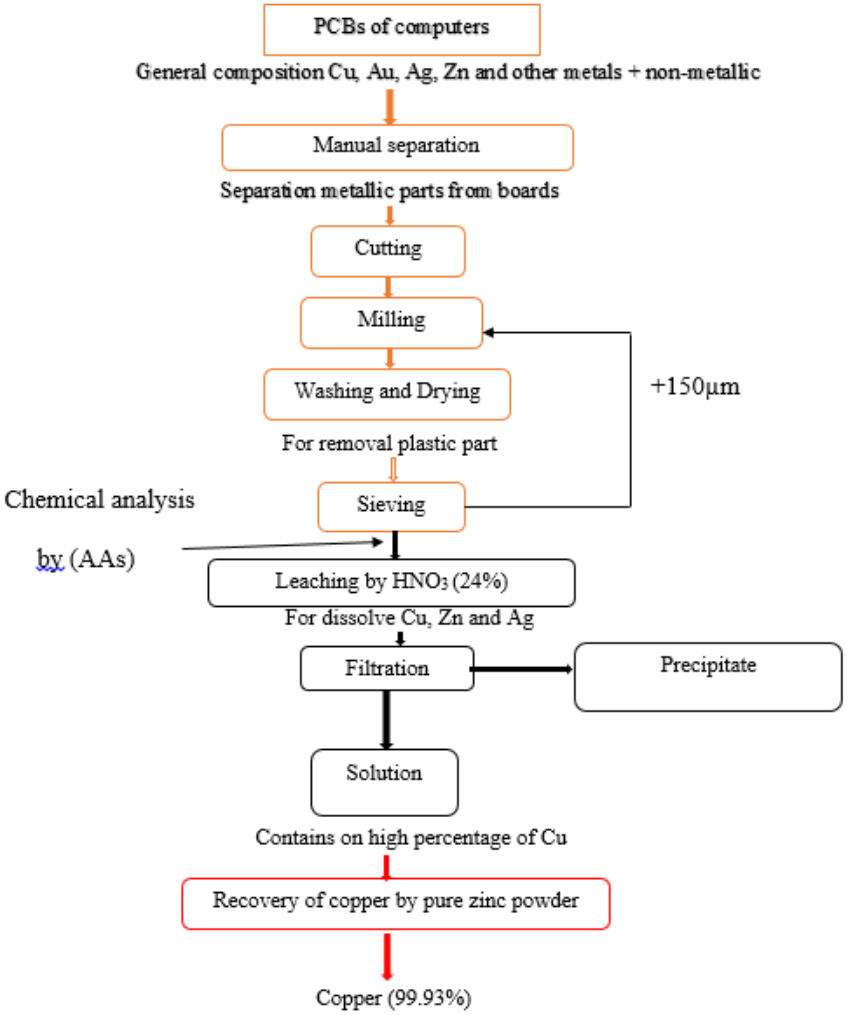

Figure (2)Flow sheet for recovery copper from PCBs of computers

2.1. Taguchi Design of Experiment

To determine the optimum conditions during leaching experiments, design of experiments by taguchi method was used. A L16 taguchi orthogonal array was chosen using Minitab program to conduct leaching experiments with nitric acid solution. The three parameters and their four levels are selected to determine the optimum leaching condition with nitric acid solution as shown in table (3).

Table 3, Leaching process data with nitric acid used in experiments according to TOA design

Exp.	Temp(˚C)	Time(min)	Solid: Liquid Ratio (wt:wt)
1	**50**	**5**	**1:20**
2	**50**	**10**	**1:25**
3	**50**	**15**	**1:30**
4	**50**	**20**	**1:35**
5	**60**	**5**	**1:25**
6	**60**	**10**	**1:20**
7	**60**	**15**	**1:35**
8	**60**	**20**	**1:30**
9	**70**	**5**	**1:30**
10	**70**	**10**	**1:35**
11	**70**	**15**	**1:20**
12	**70**	**20**	**1:25**
13	**80**	**5**	**1:35**
14	**80**	**10**	**1:30**
15	**80**	**15**	**1:25**
16	**80**	**20**	**1:20**

2.2. Leaching Process with nitric acid

The PCBs powder was dissolved in nitric acid solution with 24% concentration. The leaching process involved replication of each experiment. Two grams of prepared PCBs sample was used in leaching with nitric acid. The powder added to nitric acid 500ml glass container which has three holes for; material or reagents adding, thermometer inserting, and the last hole for condense fitting. The magnetic stirrer with heating plate was used to conduct leaching process at predetermined temperature, time, and solid to liquid ratio according to each experiment and constant agitation speed at 500 rpm for all experiments. The solution produced from leaching process were filtered using ash less Whatman filter paper, the result were subjected to chemical analysis using flame atomic absorption spectrometry. Sixteen experiments of leaching process by HNO3 was conducted according to taguchi method, and the experiments repeated twice to obtain leaching percentage.

7- For each experiment the percentage of leaching is calculated by the following formula[10]:

$$\text{Leaching} = \frac{C.V}{C.W} 100\% \qquad (1)$$

C= The gold concentration or copper concentration after process of the leaching (g/l),V=The solution volume of the after process of the leaching (l), W= PCBs sample weight in the solution of leaching (g) and C= The gold concentration or copper concentration in the PCBs (%).

Equation (2) to (3) show the half-reactions occurred in the HNO_3 leaching process [11].

$$M \rightarrow M^{2+} + 2e^- \qquad (2)$$

$$Cu + 4\ HNO_3 \rightarrow Cu(NO_3)_2 + 2\ H_2O + 2\ NO_2 \qquad (3)$$

2.3 Recovery of copper

Pure zinc powder used as precipitant agent to precipitate copper from solution produced from leaching process with nitric acid. Five grams of pure zinc powder added to 25 ml of leachant solution diluted with 75 ml of water in a beaker for 24 hour at room temperature. After copper precipitation indicated by color change of solution, the mixture was filtered and subjected to chemical analysis using flame atomic absorption spectrometry to determine copper content and calculating coper recovery.

The copper recovery percentage can be calculated by using the following formula [10]:-

$$Recovery\ \% = \frac{C_1L_1 - C_2L_2}{C_1L_1} \qquad (4)$$

Where:
C_1= The metal concentration before process of the precipitation (g/l), L_1= The solution volume before process of the precipitation (l), C_2= the metal concentration after process of the precipitation (g/l) and L_2= The solution volume after process of the precipitation (l).

The reactions of recovered copper from nitric acid solution by zinc pure powder are as

$Zn + Cu(NO_3)_2 \rightarrow Cu + Zn(NO_3)_2$ (5)

$Zn(s) + Cu^{2+}(aq) \rightarrow Zn^{2+}(aq) + Cu(s)$ (6)

Results & Discussion

3.1. Effect of time on copper leaching with HNO_3

The experiments of leaching process by nitric acid solution was carried out twice at different times (5 ,10 , 15 and 20)minutes to discover the suitable time to get on the lowest percentage solubility of gold with good percentage solubility of copper during process of leaching with HNO3. Figure 4.3 shows the time effect on the process of the copper leaching with the solution of nitric acid. Leaching efficiency of copper were increasing and decreasing irregularly at different times. The percentage of copper leaching at 5 min was 53.22%.

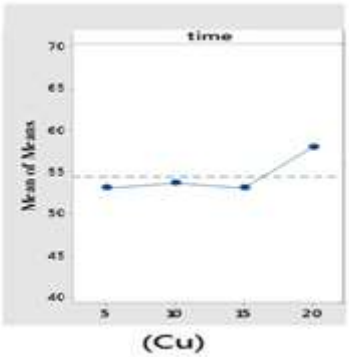

Figure (3) Efficiency of copper leaching by nitric acid solution for every level as a function of time

3.2. Effect of the temperatures on copper leaching with HNO_3

Copper leaching kinetics at four levels of the temperatures (50, 60, 70 and 80) ˚C were studied. Figure 4.3 show leaching efficiency of copper at different temperatures. The leaching kinetics of copper increasing and decreasing irregularly at different temperatures therefore the activation energy of the leaching reactions irregularly with increased temperature. Copper leaching efficiency at 50˚C 57.23% was achieved.

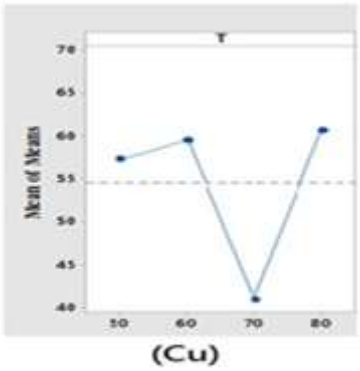

Figure (4) Efficiency of copper leaching by nitric acid solution for every level as a function of temperature

3.3. Effect of the solid to liquid Ratioon copper leaching with HNO_3

The S/L ratio (wt:wt) is considered one of the factors affecting on the copper leaching. Four levels of S/L ratio (1:20, 1:25, 1:30 and 1:35) were studied. The effect of S/L ratio on the copper leaching is shown in figure 4.3. The mean of the leaching for copper in nitric acid solution in general increased with increased S:L ratio. Whenever the ratio of solid to liquid increase the possibility the liquid on extract and dissolve the metal required from sample also increase. Copper leaching efficiency at 1:20 S/L was about 49.21% was achieved.

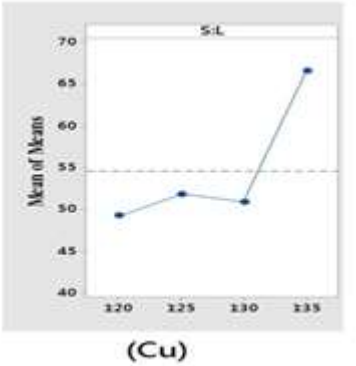

Figure (5) Efficiency of copper leaching by nitric acid solution for every level as a function of solid to liquid ratio

3.4. Recovery of copper

Copper nitrate formed after the copper dissolved in nitric acid solution during the first leaching process for printed circuit boards of computes. Pure zinc powder used to precipitate copper because it has high efficiency in copper precipitate from nitrate solution. Zinc is strong reducing agent to copper (Cu^{+2}) Because zinc is more active to interact with acids as arranged in the periodic table, which comes before copper so that react with copper nitrate to release copper (Cu) and its precipitate and zinc nitrate (Zn $(NO_3)_2$) was formed. The percentages of efficient recovery of copper (99.93%).

Conclusions

The following conclusions are drawn from this work:

1. The optimum dissolution of copper by nitric acid were 59.7%, at a time 5 minutes, temperature 50°C, and solid/liquid ratio 1/20.
2. Recovery efficiency of copper from nitric acid solution by using pure zinc powder was 99.93% at room temperature and time 24 hour.

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