IOP Conference Series: Earth and Environmental Science

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To cite this article: Nisrina Nadia Maizatul *et al* 2020 *IOP Conf. Ser.: Earth Environ. Sci.* **549** 012057

Removal of Cu from Printed Circuit Board (PCBs) Leachates using Activated Carbon Derived from Foxtail Palm Fruit

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Abstract. Printed circuit boards (PCBs) are the e-waste generated from the end-of-life electronic equipment such as laptops and mobile phone. PCBs contain relatively abundant of valuable metals such as gold and platinum. However, e-waste is considered as an environmental contaminant as it consists plenty of hazardous materials such as cadmium and copper which can pose health threat to human and also environment. It has been reported that PCBs contain a large amount of copper (Cu) in the circuit boards as it is used as a base metal. Moreover, exposure to Cu will lead to adverse impact of human health. Therefore, the objective of the study is to determine the Cu concentration using FAAS and also to remove the Cu from PCBs leachate using activated carbon derived from foxtail palm fruits. In order to remove the Cu, hydrometallurgical process on PCBs will be conducted to leach the metal into solution. Once the metal has been leached to the solution, the metal removal process using activated carbon through adsorption process was conducted. In this study, foxtail palm fruit was tested as an effective low-cost adsorbent for Cu removal. The effect of adsorbent dosage (1 g and 5 g) with fix contact time (40 min) of the prepared activated carbon in selected metal removal were investigated. The Cu in PCBs leachate solution before and after metal removal process were quantified using flame atomic absorption spectrophotometer (FAAS). Result obtained showed that, the percentage removal of Cu was recorded to be higher at high adsorbent dosage which was 14.417% in 5 g dose and 11.219% in 1 g dose. Thus, it can be concluded that the higher the adsorbent dosage used, the greater the percentage removal of Cu metal.

1. Introduction

Malaysia was estimated to generate 53 million of e-waste in the year 2020 and this due to the shorten product lifespan. Frequent changes of electronics products resulted from continuous innovation, technological advancement, new features and trend also indirectly influence to shorten of product lifespan. Furthermore, there is no formal system in place for household e-waste management although e-waste from the industries are controlled and regulated according to the Department of Environment under Ministry of Energy, Science, Technology, Environment and Climate Change. Personal computers or laptops and mobile phones are among abundant e-waste where printed circuit boards (PCBs) are their backbone of the gadget. These wastes are environmentally hazardous however, they contain substantial amount of valuable metal, including precious metal such as gold (Au), silver (Ag) and platinum (Pt) make it worth for recycling. However, e-waste is considered as an environmental contaminant as it consists plenty of hazardous materials such as copper (Cu), cadmium (Cd) and lead (Pb) which can pose detrimental health threat to human and also environment [1]. Furthermore, there are many ways of heavy metal from e-waste could be introduced to the environment. For instance, the toxic material of the e-waste could seep into the soil and also groundwater due to the action of improper management of e-waste where it is being thrown and dump in landfills and thus it also could induce pollution to the water and the aquatic organisms through runoff and sewage [2]. There are several pathways of heavy metal uptake in human body such as it may occur through inhalation, ingestion of the contaminated food and also by absorption through the skin [3]. According to Koh *et al.* [4] organism also needs heavy metals in the body to perform different function such as iron, copper and zinc are all required by humans. However, highly exposure to heavy metal such as Cu would cause adverse health effect such as vomiting, stomach cramps, nausea and worst liver damage and kidney disease [4]. Hence, it can be considered as both secondary resources and environmental toxicant. Generally, recycling and metal removal process involve hydrometallurgical processes where metal is leached from the PCBs to the solution and recovered later. Adsorption process using activated carbon is a good alternative to remove metal in PCBs as the metal concentration in the leachate solution is low [1]. Figure 1 below shows the flow process of hydrometallurgy.

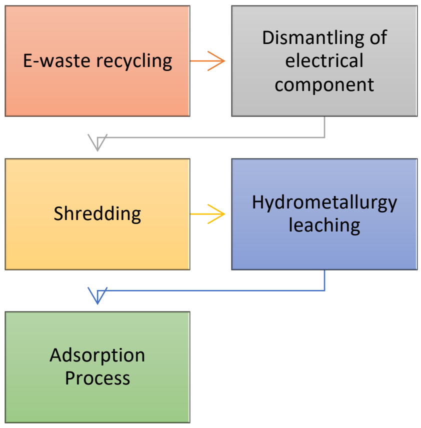

Figure 1. Flow Process of Hydrometallurgy

Furthermore, a lot of study mentioned that activated carbon capable to remove metal from low concentration solution [1, 5, 6]. In this way, many activated carbon derived from agro-waste have been proposed since the use of commercially available activated carbon is costly. Only few studies using commercial activated carbon to treat real e-waste leachate [7] The agro-waste that will be used is *Wodyetia bifurcata* or known as Foxtail palm fruit. It is a species of palm in the Arecaceae family where popularly planted in Malaysia and around the world as a landscape plant.. The foxtail palm fruit tree is shown in Figure 2.

Figure 2. Foxtail Palm Fruit Tree, Wodyetia bifurcata

Foxtail palm tree produce flowers and bunches of fruits that contain seeds.The foxtail fruits do not have any specific usage and always being left until rot. Nevertheless, agro-waste from palm species such as palm kernel shell has been studied as biosorbents for the removal of methylene blue dyes [8]. Therefore, the adsorption of Cu from PCBs leachate using activated carbon derived from foxtail fruits may contribute to: 1) converting the non-used foxtails fruit waste to the valuable product; 2) Cu can be removed; 3) the PCBs leachate will be purified and can be discarded without environmental damages.

2. Materials and Method

2.1 Preparation of Activated Carbon

Prior to use, the collected foxtail palm fruits were washed gently with distilled water to get rid of any dirt and surface impurities followed by overnight oven at 100°C to dry the sample and later were cooled down in the room temperature. Next, the dried fruit were carbonized at 300°C for two hours in the furnace and were cooled at room temperature. Figure 3a and b below show the fresh foxtail palm fruits and the carbonized palm fruits respectively.

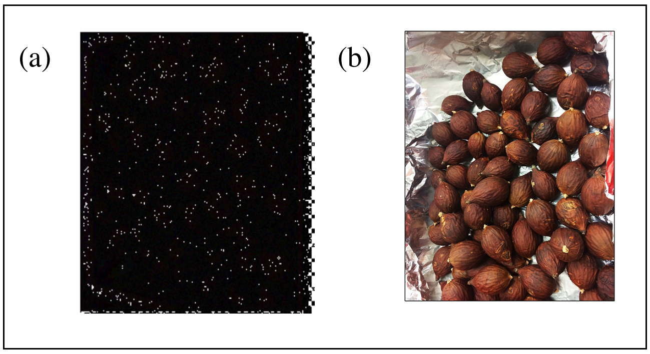

Figure 3. The Foxtail Palm Fruits in (a) Raw and (b) Carbonized

The char was crushed into small pieces using pestle and mortar and was grinded using miller blender to get smaller bits of the sample. The crushed fruit was sieved to pass through a 250 μm mesh sieve and stored in desiccator for further chemical activation process [9]. The prepared char was weighed about 40 g and then was soaked and impregnated in a beaker that contains 80 ml of concentrated HNO_3 [10]. The mixture was mixed vigorously for 30 minutes until became paste with constant stirring. The paste was left for overnight in fume hood for the chemical to fully reacted. After that, the slurry was weighed and place in dry crucibles and carbonized at 500°C for two and half hours in furnace [9]. Finally, in order to remove any excess of HNO_3, the produced activated carbon was washed using distilled water until reached pH 7. The activated carbon was dried at 100-150°C for three hours in the oven, kept in tight polyethylene bag and stored in desiccator for further use [10].

2.2. Preparation of Printed Circuit Boards (PCBs) Leachate

The PCBs were collected from one of the biggest E-waste recycling center in Kelantan which is Laksana Cergas Sdn. Bhd. This center is situated at industrial area in Pengkalan Chepa, Kota Bharu, Kelantan, Malaysia. The obtained PCBs were sorted, crushed and grounded to the smaller pieces which in range of (1-5 cm). Then, the sample went through hydrometallurgical techniques where about 20 g of grounded PCBs were soaked in 1000 mL of aqua regia and were allowed to leach for about 1 hour. The aqua regia solution were prepared using 1:3 of HNO_3: HCl. At the end of the experiment, the leachate solution was filtered and stored in 2000 ml of Duran reagent flask in the dark for further use. Figure 4 shows the preparation process of metal leaching from PCB to leachate solution.

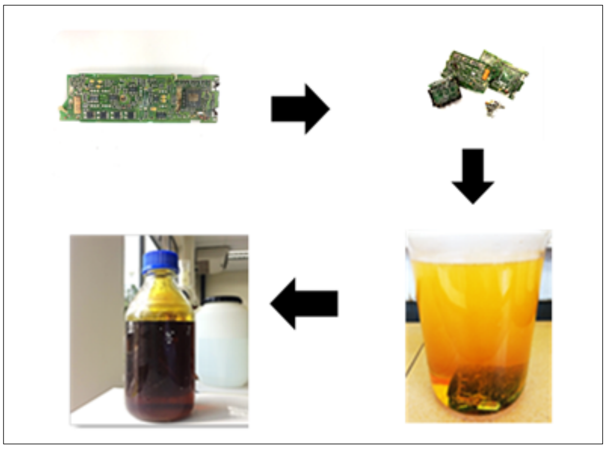

Figure 4. PCB to leachate preparation

2.3. Removal of Cu through Adsorption Experiment

Adsorption experiment were conducted using 100 mL PCBs leachate solution in 250 mL of Erlenmeyer flask by comparing two different adsorbent dosage which were 1 g and 5 g, respectively with a fix contact time of 40 min. In each experiment, the flask was shook at 150 rpm to ensure the homogeneity. The percentage (%) removal of Cu by the activated carbon produced from foxtail palm fruit were determined as equation (1) [11].

$$\textbf{Percentage removal (R), \%} = \frac{Ci-Cf}{Ci} \times 100 \qquad (1)$$

Where C_i is the initial reading of the metal concentration and C_f is the final reading of the metal concentration in the leachate solution.

2.4. Determination of Percentage of Yield

The percentage yield (%) of foxtail palm fruits activated carbon was obtained by taking the final mass of the activated carbon divided by the initial mass of precursor by the following equation (2) [12]:

$$\textbf{\% of yield} = \frac{Wf}{Wi} \times 100 \qquad (2)$$

Where W_f is final mass of the activated carbon at the end of activation process and W_i is the initial mass of the dry impregnated char.

2.5. Characterization
The concentrations of Cu metal in the PCBs leachate solution before and after removal process were quantified using FAAS.

3. Result and Discussion

3.1 Percentage of Yield
The percentage yield of foxtail palm fruit activated carbon which was impregnated with a ratio of 2:1 HNO_3 and carbonized at 500°C was 59.42% which was relatively high compared to the study by Kouotou *et al*. [12] on oil palm shell activated carbon which the percentage yield was recorded as 56.47 % at 630°C of activation temperature [12]. The slightly different of the yield from these palm species is might be due to higher temperature use when conducted for the oil palm shell activation process. The increase of temperature degrades the microstructure of the activated carbon [12]. Hence, more volatiles are release resulting the lower the percentage of yield [11].

3.2 Effect of Contact Time and Adsorbent Dosage
The preliminary study of Cu removal using activated carbon derived from foxtail palm fruits was conducted whereby, two different dosage of activated carbon which were 1g and 5g were chosen to differentiate and to compare the effectiveness of Cu removal on low and high adsorbent dosage. Both of these different dosages were tested for 40 min of contact time. The result is shown in Figure 5 below.

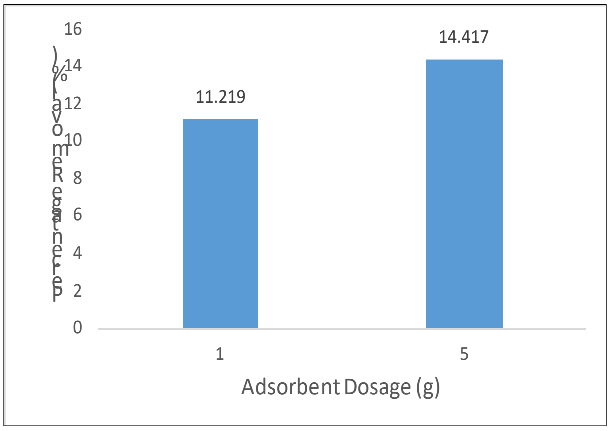

Figure 5. Percentage Removal of Cu after 40 min of contact time with different adsorbent dosage

The percentage removal of Cu for adsorbent dosages (1 g and 5 g) were 11.219% and 14.417%, respectively. This percentage removal was considerly low and this could be the shorter contact time (40 minutes) of the conducted experiment where most of the studies [1,5,6,13] showed highest removal of metals could be achieved with up to 24 hours of the contact time. However, from the result, it is clearly shows that the highest Cu removal percentage was obtained at higher adsorbent dose (5 g). Therefore, percentage removal increases with higher adsorbent dosage use. This is due to the introduction of binding sites for the adsorption process [14]. Moreover, many conducted studies on the Cu removal reported that Cu was limited to only monolayer adsorbent, where it is only in contact with the surface layer of the adsorbent itself [5]. According to the study by Wahi *et al*. [5], different adsorbent dosage (0.2, 0.4, 0.6, 0.8 and 1.0 g) of palm oil empty fruit bunch activated carbon were added to 100 ml of heavy metals solution which contain initial concentration ranging from 10 to 20 mg/L and agitated up to 24 hours of contact time. Then, it is found that, the result from the study also showed the lower percentage removal of Cu. However, the Cu removal percentage was decreases when higher initial Cu concentration is being used [5]. This is because, at high concentration, the adsorbent active sites might get saturated and therefore the metal ions could not freely interact with the adsorbent active sites and thus the percentage removal decrease [15]. Moreover, a similar result was also obtained by a study that was conducted by Benzaoui *et al*. [16] on the adsorption of Cu. A range of adsorbent dose ranging from 1g to 5g were tested in 100 ml of copper solution and agitated up to 70 minutes of contact time. The result of the study shows that the maximum removal rate of Cu was 80% at 3g of adsorbent and brought to no increment of Cu percentage removal after further addition of adsorbent dosage [16]. High adsorbent dosage might overlap the adsoprtion active site as a result of overcrowding of adsorbent particles and hence the metal would be shielded from binding to the adsorbent active sites [6]. Therefore, from this study, it can be understand that the percentage of Cu removal is low because of the Cu might be limited to the monolayer adsorbent, hence resulted on the amount of metal binds to the active site of adsorbent also low [5]. However, the longer contact time along with the higher adsorbent dosage would potentially contributed to the higher Cu removal as it may give higher chance for the metals to adsorp onto the adsorption site and thus the removal of the metals will be increased**.**

Furthermore, there were also studies which doped the activated carbon with other element such as CuO [17,18] to enhance both physical and chemical properties of the produced activated carbon. However, in this experiment the doping process using oxide metal to the produced activated carbon was not recommended as the objective of the study was to remove the Cu metal. Thus, positive charge in both of the elements (CuO and Cu in PCB leachate) inhibit the metal to be attracted with each other [19]. Hence, the Cu metal will unable to be removed from the solution.

3.2.1 Author Recommendation on the Study
The application of Foxtail Palm Fruit, *(Wodyetia bifurcate)* as activated carbon in removing Cu has been studied. This preliminary result has indicated that the foxtail palm fruit could be an effective adsorbent for the adsorption treatment and able to remove Cu metal. The highest percentage removal of the metals can be achieved at longer contact time and higher adsorbent dosage. Hence, other values of different contact time such as 60 till 120 min with different adsorbent dosage of 1 to 5 g will be studied along with the progress of this research. Therefore, the trends of the metal adsorption process could be traced.

4. Conclusion
This study has demonstrated that hydormetallurgical leaching process could leach the printed circuit boards into solution and hence the concentration of Cu before and after the removal process could be determined using FAAS. Moreover, this study also shows that activated carbon produced from foxtail palm fruit is potentially for removing Cu in the PCBs leachate as the Cu removal percentage after 40 min is calculated and recorded with the percentage of yield of 59.42 %. Hence, foxtail palm fruits agro-waste could be reduced. Furthermore, from the result, it is clearly shows that higher removal percentage of Cu could be obtained with high adsorbent dosage and also at longer contact time.

Acknowledgements
This research work was financially supported by the Ministry of Higher Education (MOHE) Fundamental Research Grant Scheme (FRGS) R/FRGS/A0800/00235A/003/2019/00705. Special appreciation to Faculty of Earth Science, Universiti Malaysia Kelantan and also Laksana Cergas Sdn. Bhd. recycling center for providing technical supports and cooperation

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