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Removal of Pb(II), Fe(II) and Zn(II) using activated carbon produced from foxtail palm fruit chemically activated by KOH and H_3PO_4

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Abstract

The increasing rate of urbanization and continuous developments are the main factor which led to heavy metals contamination into the environment especially in the water bodies. However, the contamination of heavy metal can be treated using adsorption process using activated carbon. Thus, this study was based on using powdered activated carbon, which prepared from foxtail palm fruit and chemically activated using potassium hydroxide and phosphoric acid. The main parameters such as effect of chemical activating agent, effect of initial concentration of heavy metal and effect of sorbent dosage that influenced the sorption process were studied. From the result, activated carbon that was chemically activated by phosphoric acid shown the best removal compared to activated carbon that was chemically activated by potassium hydroxide. The percentage removal of Pb(II), Fe(II) and Zn(II) were 95.8%, 99.9% and 22.8% respectively using 0.5 g of activate carbon. The result indicates that the adsorption process using activated carbon that produced from plant can be applied for heavy metal removal from aqueous solution.

1. INTRODUCTION

The rapid development of industries such as metal plating facilities, mining operations, fertiliser industries, textile industries, tanneries and batteries industries has discharged wastewater streams containing heavy metals directly or indirectly into the environment (Fu and Wang, 2011). The discharge from the listed industries contain heavy metal may reach into water resources hence, affects the quality of water. Heavy metals are dangerous because of their persistence, not biodegradable and tend to accumulate in living organisms (Cai *et al*., 2012; Fu and Wang, 2011). Water pollution from heavy metals is a major concern especially in developing countries including Malaysia.

Pb is among heavy metals that was listed as priority pollutants by the U.S Environmental Protection Agency (USEPA) (Igwe and Abia, 2006) and originated from the mining and smelting activities, transport emission as well as from old lead paints (Duruibe *et al*., 2007). It leaches through poor housing pipeline and move from one place to another. The movement of Pb in food was proven by their accumulation in foods that grown in particular locations with high levels of Pb in soils or from fertilizers (Sears *et al*., 2012). High acid food that are stored in ceramic containers also may contributed to the presence of Pb in our diet (Ravichandran, 2011). According to Shrestha *et al*., (2011), the permissible level of lead in drinking water and surface water intended for drinking as set by EU, EPA and WHO are 0.01, 0.05 and 0.01 mg/L respectively. However, recent study of Mohod and Dhote (2013) have confirmed that permissible level for Pb in drinking water and surface water is 0.05 mg/L. Pb concentration that is beyond the limits are not allowed to consume, use or any intentions to do so.

Although zinc is not very abundance, but it is readily available because it occurs in concentrated deposits of sulphide ores, usually with iron, lead and many other metals (Parish, 1977). Zinc enter the environment through natural processes however, most come from human activities as zinc is co-product of lead mining (Lajis, 2013). Zinc attaches to soil, sediment and dust particles in the air and it can be moved into the ground water as well as river (Lajis, 2013). Whereas, iron is the most abundant metal in the Earth's crust. Iron being used in the steel industries such as to make bridge, building and most of the skyscrapers. Iron generation from casting operation is one of the instances that contributed to the heavy metal pollution to the environment. The permissible limit for zinc in the drinking water set by WHO is 3 mg/L, nevertheless there

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is no guideline provided for iron limit. However, EU has set only 0.2 mg/L of iron may contain in the drinking water.

Thus, a number of treatments are conducted in order to remove the heavy metal and the treatments are ion-exchange, coagulation and flocculation, oxidation-reduction, chemical precipitation and electrochemical methods. However, most of these treatment processes are costly and in some cases that tend to generate secondary waste by-product (Vunain *et al.*, 2017). Ultimately, adsorption method using activated carbon is a well-established has a great potential for heavy metal removal. However, the consumption of activated carbon is costly due to its production. Hence, production of activated carbon from agricultural waste or unwanted part of the plant is necessary in order to reduce the production cost furthermore it can be one of the environment sustainable solutions for the disposal issue (Mohamad *et al.*, 2017). Due to the abundance of foxtail palm fruit in Malaysia, as its trees are planted most along the road as an ornamental plant, the fruits were chosen as a potential raw material for the activated carbon production and expected to have the ability for Pb, Zn and Fe removal.

2. MATERIALS AND METHOD

2.1. Preparation of Activated Carbon

Foxtail palm fruits (Figure 1) were collected around Kubang Kerian, Kelantan, Malaysia. The fruits were washed several time with distilled water as to remove surface impurities and then dried in an oven at 100°C overnight. The dried fruits were hard to crush, hence, need to be carbonized at 300°C. The carbonization was conducted for two hours in order to get the char samples and allowed to cool in that furnace for three hours (Zakir, 2013). Next, the char samples were crushed using miller blender, sieved to pass through a 1.18 mm mesh-sieve and kept stored in desiccator for further chemical activation process.

Figure 1: (a) Foxtail palm fruit tree and (b) the foxtail palm fruits.

The prepared char was weighed about 100 g and added to the conical flask containing 200 ml of 20% phosphoric acid, H_3PO_4. The conical flask was shaken about 5 minutes for homogeneity and left overnight for impregnation. After that, the sample was carbonized for 2 hours in furnace with temperature of 500°C (Awoyale *et al.*, 2013). Finally, the produced activated carbons were rinsed with distilled water to remove any excess H_3PO_4. The carbon samples were dried at 100°C overnight and kept in air thigh container prior using for heavy metal removal. The procedures were repeated using other chemical activating agent, potassium hydroxide (KOH). Methods in preparing activated carbon were adopted from Zakir, (2013) and Awoyale *et al.*, (2013) with modification in order to suit with the material in this study, foxtail palm fruit.

2.2. Preparation of Synthetic Heavy Metals Solution

The synthetic heavy metal solution of Pb(II), Fe(II) and Zn(II) were prepared by dissolving separately 1.5985 g $Pb(NO_3)_2$, 2.7200 g $FeSO_4$ and 2.0844 g $ZnCl_2$ respectively in a volumetric flask and diluted up with 1000 ml of distilled water to make 1000 mg/L of stock solution. The stock solution was diluted to the desired concentration for further experiment.

2.3. Adsorption Studies

In each experiment, a total volume of 25 ml solution containing a known concentration of a single heavy metal, Pb(II) with the initial concentration of 50 ppm was placed in the conical flask. The solution was adjusted to the pH 5 using 0.1M HNO_3 and/or 0.1M NaOH. The pH was not controlled during the experiment. Then, 0.5g of activated carbon was added to the solution for the adsorption process. The conical flask was then rotated for 30 mins at 200 rpm to ensure the homogeneity. Same steps were repeated for Fe(II) and Zn(II) removal. The conducted experiments were repeated for the different value of initial concentration used and different amount of activated carbon loaded to the solution for the removal purposes. The treated heavy metal solution was analysed using AAS spectrometer. The experiments were conducted triplicate.

2.4. Determination of heavy metal removal

The percentage (%) removal of heavy metals by the activated produced from foxtail palm fruit were calculated by the following equation.

Percentage removal of heavy metal, % =

$$\frac{C_i - C_f}{C_i} \times 100 \qquad \text{(Equation 1)}$$

Where C_i is the initial reading of the heavy metal concentration, C_f is the final reading of the heavy metal concentration.

3. RESULTS AND DISCUSSION

3.1. Effect of Chemical Activating Agents

Figure 2 showed the percentage removal of 50 mg/L Pb(II) using two different type of activating agents. The maximum removal of 95.8% was obtained using 0.5 g activated carbon that activated by phosphoric acid, H_3PO_4. Only 35.0% of Pb(II) removal occurred when activated carbon that activate by KOH was applied to the solution. The higher value of percentage removal for Pb using activated carbon that chemically activated by H_3PO_4 has been supported by previous study which stated that H_3PO_4 allows the micropores and mesopores development hence resulted more active site for adsorption occurred (Kumar and Jena, 2016; Yorgun and Yildiz, 2015). Therefore activated carbon chemically activate by H_3PO_4 was selected for subsequent experiment.

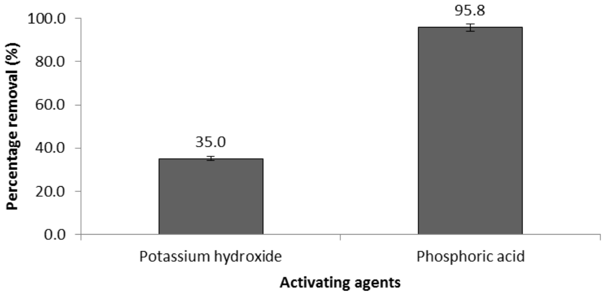

Figure 2: Effect of different chemical activating agents.

3.2. Effect of Dosage on Activated Carbon Used

The activated carbon dosage is important parameters that need to be studied as the amount used reflected to the cost. The removal of 50 mg/L Pb(II) using 0.5 g and 2.0 g of activated carbon was given in Figure 3. It shows insignificance difference of Pb removal. The Pb removal was 95.8% and only increased to 98.9% once the amount of activated carbon used was increased from 0.5 g to 2.0 g. This result explained that although quadruple amount has been added to the Pb solution, 100% of Pb removal was to no avail. Nevertheless, previous studies by Manoochehri *et al.* (2012) and Hegazi (2013) found that as the amount of activated carbon increased, the percentage of Pb(II) removal increased accordingly. Therefore, 0.5 g of activated carbon dosage was used for subsequent experiment.

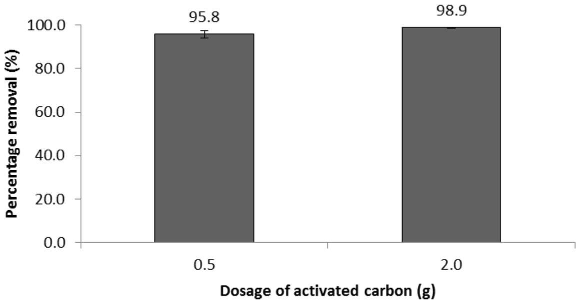

Figure 3: Effect of dosage on activated carbon used.

3.3. Effect of Pb Initial Concentration

The effect of initial Pb concentration has been studied and presented in Figure 4. The result show that removal of Pb(II) was decreased as the Pb initial concentration was increased. This due to the depletion of active site availability as the initial concentration of was increased (Mohamad *et al.*, 2017).

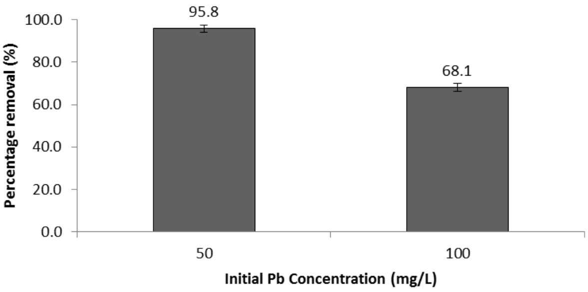

Figure 4: Effect of initial Pb concentration.

3.4. Heavy Metal Removal

The ability of 0.5g activated carbon produced from foxtail palm fruit to remove another heavy metal such as Fe(II) and Zn(II) were carried out and the result showed in Figure 5. Almost complete Fe(II) removal was achieved (99.9%), however only 22.8% of Zn(II) removal occurred at initial concetration of 50 mg/L. The adsorption capacity of Pb(II), Fe(II) and Zn(II) showed in Figure 6.The result clearly indicated that the actual amount of Fe(II) adsorbed (mg/g) on the activated carbon higher than Pb(II) whereas Zn(II) was the lowest. This could be due to the higher ability of Fe(II) and Pb(II) in competing with Zn(II) for binding site with activated carbon (Chen *et al.*, 2011).

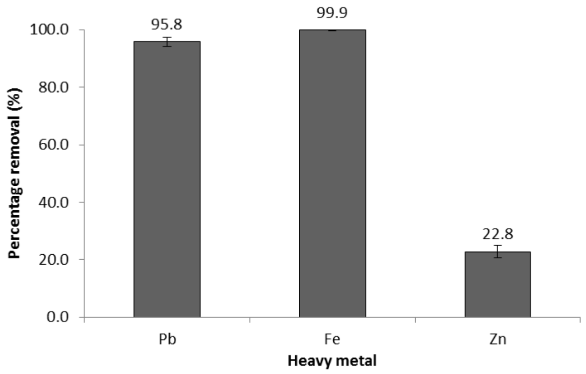

Figure 5: Heavy metal removal.

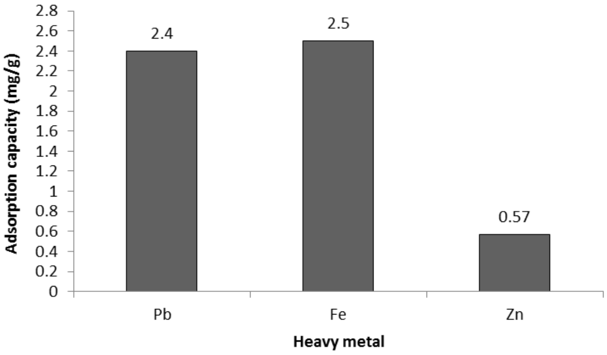

Figure 6: Adsorption capacity of various heavy metal.

4. CONCLUSION

This study has demonstrated that activated carbon produced from foxtail palm fruit could be one of the potential activated carbon in removing heavy metal pollutant. The excellent removal of 99.9% Fe(II) using only 0.5 g of activated carbon which activated by H_3PO_4.

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