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Preparation of Activated Carbons from Coconuts Shell for Pb (II) Adsorption

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Abstract. The purpose of this study is to prepare activated carbon from coconut shell to be used as an adsorbent for heavy metals. The coconut shell was carbonized at a temperature of 700^0C and the resulting carbon is finely milled and sieved through No 200 mesh. Sulfuric acid is used as an activator for this process. The characteristics of the resulting activated carbon were tested and the results are as follows: moisture content 1.641%, ash content of 0.554%, iodine adsorption of 960.21 mg/g and the adsorption of methyl blue 212.1 mg/g. The small particle size of the activated carbon increases the surface area measured using BET method to 398.721 m^2/g. Adsorption kinetic and adsorption efficiency of the activated carbon on Pb by batch method follow the pseudo second-order equation order model with adsorption capacity at equilibrium equals to 24.491 mg per g Pb (II) adsorbent.

Introduction

Environmental contamination by lead is generally derived from human activities. Lead is widely used as raw material for batteries, ammunition, medical supplies, etc. When these toxic and hazardous materials were dumped in a landfill, infiltration and percolation of rainwater in the landfill produce leachate which contains dissolved organic matter, inorganic and heavy metal macro components also like Pb, Cd, Cr and others [1]. Leachate which is not treated before discharge is a source of groundwater and surface water contamination because the lead can be dissolved in the water and go into human body through food chain.

A number of treatment methods for the removal of metal ions from aqueous solutions have been reported, mainly by elimination, ion exchange, electrolysis, electrochemical precipitation, evaporation, solvent extraction, reverse osmosis, chemical precipitation and adsorption process [2]. Various attempts have been made to reduce the content of heavy metals through processes such as adsorption using zeolites [3], natural adsorbent rice hulk ash [4], carbon aerogen [5] and activated carbon. Activated carbon has been widely used to adsorb heavy metals such as Cu and Zn [6], and aluminum [7]. Several studies have been done to make activated carbon from some types of plant [8], among them is the work by Amuda [9] who used pyrolyzed coconut shell, which is a material that is easy to be found in Indonesia and is renewable.

In this study activated carbon is prepared from coconut shell which passes No 200 mesh and sulfuric acid (H_2SO_4) as activator during the process. With the smaller particle size, surface area is expected to be even greater, thus increasing the adsorption capacity. Characteristics of the activated carbon i.e. moisture content, ash content, adsorption of iodine and the adsorption of methyl blue will be tested and compare with SNI standard 06-3730. Activated carbon produced will be tested in Pb solution adsorbance with variation of contact time and adsorbent weight.

Materials and methods. Materials used in this study are coconut shell as a source of carbon, H_2SO_4 solution as an activator and a solution of Pb $(NO_3)_2$ analytical grade (Aldrich) as the test solution adsorption.

Activated carbon preparation. Coconut shell is cleaned from impurities, then washed thoroughly and dried. A total of 1000 grams of coconut shell carbonized in a furnace at a temperature of 700^0C for 2 hours. After experiencing the carbonization process, the carbon sample is milled and sieved through No 200 mesh. Carbon samples that pass 200 mesh were then soaked with sulfate acid (H_2SO_4) 4 M at 1:3 ratio, then heated and stirred using a magnetic stirrer at a speed of 200 rpm at a temperature of 100^0C for 1 hour. The sample is sieved again upon cooling to ambient temperature then heated at a temperature of 200^0C for 2 hour. The resulting carbon is washed with double distilled deionized water until neutral pH is reached. Furthermore, the carbon is soaked in 2% (w/v) $NaHCO_3$ to remove the remaining sulfuric acid. The activated carbon is washed again until it reached neutral pH, and then heated in a furnace at 500^0C temperature for 2 hours.

Activated carbon characteristics. The measured characteristics of the activated carbon in this study include: moisture content, ash content, adsorption of iodine and methyl blue were tested and the results were compared with the SNI 06-3730 standard. The surface area of activated carbon was measured using BET isotherm.

Batch adsorption experiment. For the determination of adsorption equilibrium time, as much as 0.1 g activated carbon is added into six samples of 50 mL Pb solution with a concentration of 50 mg/L. Each sample is stirred at 150 rpm at temperature of 30^0C for 15, 30, 45, 60, 75 and 90 minutes. Samples were filtered until clear and then the concentration of the filtrate was measured using the AAS.

The effectiveness of adsorption is reached by performing the test with adsorbent weight variation of 0.1 to 0.5 g per 50 mL of Pb, stirring at 150 rpm for 60 minutes. Samples were filtered until clear and then the concentration of the filtrate was measured using atomic absorption spectroscopy (AAS).

Analysis and data evaluation. The percentage of the removed Pb(II) ions (R_{em}%) in solution was calculated using Eq.1.

$$R_{em}(\%) = \frac{(C_0 - C_t)}{C_0} \times 100 \tag{1}$$

While adsorption capacity was calculated using Eq.2.

$$q_t = \frac{(C_0 - C_t)V}{W} \tag{2}$$

where C_0 and C_t are the initial Pb(II) concentrations in mg/L and at a given time t, respectively; *V* is the volume of Pb (II) solutions in mL and W is the weight of activated carbon in g.

Adsorption dynamics. The Pb ion adsorption on activated carbon can be modeled kinetically as the pseudo first-order equation, pseudo-second-order equation based on adsorption capacity with the rate of adsorption [10], and intra-particle diffusion. Diffusion films for adsorption systems are controlled by diffusion [10].

The pseudo first-order equation. The pseudo first-order kinetic equation by Lagergren is expressed as follows:

$$\frac{d_{qt}}{d_t} = k_f\,(q_e - q_t) \tag{3}$$

with q_e and q_t is the amount of adsorbate adsorbed at equilibrium and at time t (mg/g), the rate constant k_f is the first pseudo first order (min^{-1}). q_e and k_f values are obtained from the plot of ln (q_e - q_t) versus t, with k_f and q_e are the slope and intercept respectively [10].

The pseudo second-order equation. The pseudo second-order adsorption kinetics rate equation is expressed as [10]:

$$\frac{d_{qt}}{dt} = k_2\,(q_e - q_t)^2 \tag{4}$$

where the parameters q_e (mg/g) and k_2 (g mg^{-1} min^{-1}) was calculated from the plots versus t / q_t versus t with $h = k_2q_e$ (mg / g min^{-1}) is the initial uptake rate at $t \rightarrow 0$.

The intra-particle diffusion model. Weber and Morris formulate intra-particle diffusion model to track the movement of the adsorbate from the adsorbent surface to the internal pore due to agitation, as follows:

$$q_t = k_i t^{0.5} + C \quad (5)$$

where q_t (mg/g) is the amount of adsorbate on the adsorbent surface, k_i (mg/ $min^{0.5}$) is the rate constant of intra-particle diffusion, t (min) is time and C (mg/g) is the intercept k_i value obtained by plotting q_t versus $t^{0.5}$[10].

Results and discussion

Activated carbon characterization. Table 1 shows the results of test on the characteristics of activated carbon from coconut shell as compared to the SNI 06-3730 standard.

Table 1. Characteristics of Coconut shell Activated Carbon

Type of test	SNI 06-3730	Result
Water content (%)	15	1.641
Ash content (%)	10	0.554
Adsorption of Iodine (mg/g)	Minimal 750	960.21
Adsorption methyl blue (mg/g)	Minimal 120	212.1
Surface Area (m^2/g)	-	398.721

The purpose of the determination of the water content is to evaluate the hygroscopic properties of the activated carbon. The average water content of the activated carbon produced in this study is 1.641 % . This suggests that the active carbon qualifies the SNI standard, and also showsed that the water content of free and bound water in the material has evaporated during the carbonization process [11]. The purpose of the determination of the ash content is to obtain the metal oxide content in activated carbon. The resulting ash content was 0.554 % that qualifies SNI standard.

The adsorption of iodine is an important indicator in assessing the activated carbon. The resulting activated carbon has the adsorption of iodine solution of 960.21 mg/g. Carbonization at high temperature of 700^0C is applied to produce high adsorption of iodine solution. The adsorption of iodine showed the ability of activated carbon to absorb substances with molecular sizes smaller than 10 A^0 and give an indication of the number of pores with a diameter of 10-15 A^0. This indication is in accordance with the results of the surface area measurements of the activated carbon isotherms by BET (Brunauer Emmett Teller) with Nova Instrument equipment based on adsorption of N_2 gas. The surface area of the activated carbon produced is 398.721 m^2/g with a pore diameter of 15.574 A^0. Adsorption of methyl blue showing the ability of the carbon to adsorb molecules that are large, in which the adsorption of methyl blue was 212.1 mg/g which qualifies the SNI standard. This shows that the activated carbon is reasonably good in terms of adsorption.

Effect of carbon dosages. Figure 1 indicates the effect of amount of adsorbent on the effectiveness of adsorption. The more adsorbent is added, the greater the adsorbed Pb [6, 9].

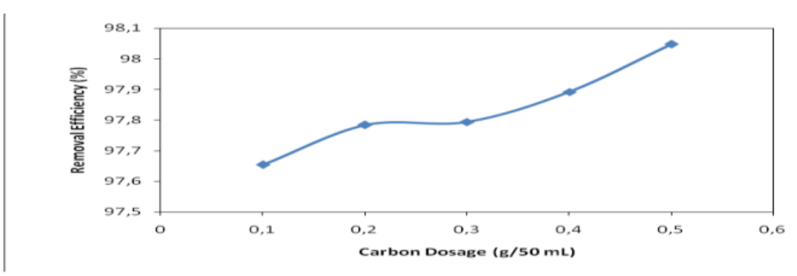

Figure 1. Removal efficiency of Pb by the activated carbons

Kinetics adsorption. The Pb ion adsorption kinetic model by activated carbon is determined from the data at the equilibrium time. The model is used to explain the mechanism of adsorption.

Adsorption kinetic parameter values of three kinetic models are summarized in Table 2. It can be seen that the adsorption of Pb (II) using the activated carbon from coconut shell follows the pseudo second-order models. It can be inferred from the highest value of R^2, which is equal to 0.956. Adsorption capacity at equilibrium obtained was 24.491 mg of Pb (II) per g of adsorbent. This result is similar to previous study [12], where the adsorption capacity was found to be 26.546 mg of Pb (II) per g of adsorbent.

Table 2. Adsorption kinetics model parameter values of Pb (II) ions by activated carbon

Kinetics model		Parameter		R^2
Pseudo first –order	k_f (min^{-1}) = - 0.021	q_e (mg/g)	1.396	0.602
Pseudo second-order	h (mg/g min^{-1}) = 14.78	q_e (mg/g)	5.399	0.956
Intra-particle diffusion	k_i (mg g^{-1} $min^{0,5}$) = 0.036	C (mg/g)	24.14	0.694

Conclusion

It can be concluded from this study that the activated carbon produced from coconut shells is of good quality and meet the quality standards of SNI 06-3730. The effectiveness of activated carbon adsorption of the solution reached 97.65% Pb in a solution of 50 mg/L and 0.1 g of adsorbent. The adsorption of Pb (II) by activated carbon from coconut shell follows the pseudo second-order models with R^2 value of 0.956. The adsorption capacity at equilibrium reached 24.491 mg of Pb (II) per g of adsorbent agrees with published data.

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