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APPLICATION OF NANOCOMPOSIT OF AL_2O_3/ACTIVATED CARBON PREPARED BY HYDROTHERMAL PROCESS FOR PHENOL REMOVAL

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ABSTRACT

Nancomposite of Al_2O_3/activated carbon was prepared from the mixture of alumina oxide (Al_2O_3) and activated carbon from palm oil shell under the mixture of aquabidest and ethanol (1:1). The mixture was treated by hydrothermal process at 250 ºC for 3 h. Resulted Al_2O_3/ activated carbon was characterized using SEM/EDX for studying the morphology structure of activated carbon and composite showing a like flower structure attached on the surface. The chemical compositions were 73.18%, oxygen 23.72%, aluminium 2.30%, dan silica 2.00%. The efficiency of composite was investigated for removal of phenol in aqueous solution. The maximum adsorption of phenol was obtained at pH 2, adsorbent weight 2 g, contact time 60 min and phenol concentration 100 ppm.

Keywords: *composite of Al_2O_3/activated carbon, palm oil shell, activated carbon, phenol*

INTRODUCTION

Phenol is aromatic compound containing a dangerous pollutant that can cause unpleasant smell and taste, toxics and non-biodegradable in water. Phenol is widely used in the household products starting from material in industry such as plastic, pharmacy textile industries. Most of the rest phenol discharged in the water effluence that can cause an environmental problem [1,2,3]. Phenol is highly irritation to skin, eyes and mucous membrane and potentially dead when it adsorb in human body [4,5,6]. Adsorption is separation method using high porosity of adsorbent. The process may occur by transfer of sorbate through pore on the surface of adsorbent under chemisorption and physisorption process [6]. Few methods have applied to reduce the phenol in the environment by physical and chemical process based on the BAPEDAL No.3. 1995. One of the popular methods used for treating phenol is adsorption methods using adsorbent [7,8,9]. Adsorption is separation method using high porosity of adsorbent. The process may occur by transfer of sorbate through pore on the surface of adsorbent under chemisorption and physisorption process [10]. The percentage of Adsopsi :

$$\%Adsorpsi = \frac{C_o - C_e}{C_o} \text{ x } 100\% \qquad (1)$$

Where Co and Ce are initial concentration and final concentration of phenol B (mg/L).

Activated carbon is widely used as adsorbent for separation and purification methods. The process of adsorption is generally influenced by the pH solution, contact time, concentration and adsorbent dosage [11]. Improving the adsorption characterization of adsorbent can be treated by modification using metal oxide such as Al_2O_3. Increasing the adsorption factors such as active size Al^{3+} and O^{2-} may increase the adsorption capacity [12]. Extension of adsorbent properties by combining of Al_2O_3 and activated carbon may increase the adsorption capacity though improving the BET surface area, pore size distribution and active size.

Hydrothermal is one of an important method for activated carbon and composite process. It is a simple process with low temperature using water as reaction media. it can accelerate the evaporation of particles in the formation of activated carbon [13,14]. Research to prepare composite Al_2O_3/activated carbon and investigate its adsorption capacity in the removal of phenol. The efficiency of the Al_2O_3 was evaluated at different adsorption parameters involving pH solution, contact time, concentration and adsorbent dosage. The amount of removal was determined by the UV-Vis make a interesting topic.

METHODS

1. Materials

Palm oil shell was obtained in Riau province, Indonesia. Chemicals such as potassium hydroxide (KOH), ethanol, nitric acid, phenol and Al_2O_3 from Merck and directly used in the experiments.

2. Instrumentation for Characterization

Characterization of activated carbon and composite of Al_2O_3/activated carbon was carried out by the Scanning Electron Microscopy-Energy Dispersive X-Ray (SEM-EDX). The amount of phenol removal was calculated by the UV-Visible spectrophotometer.

3. Research Methods

a. Activated carbon preparation

Activated carbon was prepared by the impregnation of palm kernel in 50% potassium hydroxide. The mixture was refluxed at 85 ^oC for 24 h. Thereafter, sample was washed, neutralized to pH 6-7 by dropping 5M of nitric acid and dried in oven overnight. Dried sample of palm kernel was pyrolized at 500 ^oC for 4h as contact time to obtain activated carbon.

b. Preparation of Al_2O_3/activated carbon

Combining of activated carbon and Al_2O_3 was carried out in the hydrothermal reactor. The mixture was added aquabidest and ethanol (1:1). The mixture was stirred for 1 h and placed in graphite furnace. The furnace was slowly heat up to 250 ^oC for 3 h. Thereafter, the composite was washed with distilled water and dried into oven at 110^oC for 24 h. The composite was kept for further analysis.

c. Characterization of composite Al_2O_3/ activated carbon

Characterization of activated carbon and composite of Al_2O_3/activated carbon was selected with SEM EDX used to study the morphology surface of composite.

d. Adsorption of phenol

Adsorption process for removal of phenol onto Al_2O_3/activated carbon was evaluated by the different pH solution (2, 7 and 10), adsorbent dosage (0.5, 1.0, 1.5 and 2 g), contact time (15, 30, 45 dan 60 min), concentration (0, 20, 40, 50, 60, 80, dan 100 ppm). Experiment was carried out by the batch system at room temperature. The adsorption capacity was determined by the UV-Visible at 260 nm.

e. Stock solution of phenol

Stock solution of phenol was prepared by the following procedure. Dissolve 1.00 g of phenol with distilled water in a calibrated 1000ml volumetric flask and continued to the mark with distilled water.

f. Effect of pH solution

Each of the experiment data was obtained from 50 ml of phenol concentration of 50 ppm. The solution was set up with different pH solution (2, 7 and 10) using buffer solution. Each of solution was added 0.5 gram Al_2O_3/activated carbon and stirred for 30 min at room temperature. The result was measured with UV-Visible to find the optimum condition of pH versus percentage of phenol removal.

g. Effect of adsorbent dosage

The volume of phenol solution was set up 50 ml with concentration of 50 ppm with the optimum pH obtained from previous experiment. Each of solution was added adsorbent with different weight (0.5, 1.0, 1.5 and 2 g). The mixture was stirred for 30 min as contact time at room temperature and di result was measured with UV-Visible to obtained optimum condition of percentage of phenol removal versus adsorbent dosage.

h. Effect of contact time

The solution was prepared with 50 ml of phenol solution, concentration 50 ppm, the optimum of pH condition. Each of solution was stirred at room temperature with different contact times (15, 30, 45 dan 60 min). The maximum condition of phenol adsorption was obtained from percentage removal versus contact time.

i. Effect of concentration

The effect of concentration of phenol was investigated with 50 ml volume, contact time 30 min, optimum pH condition and 50 g of adsorbent dosage. Concentration was set up from 20, 50, 70 and 100 ppm at room temperature. The maximum adsorption of phenol was obtained from the percentage of phenol removal versus concentration.

RESULTS AND DISCUSSION

1. The effect of Al_2O_3 onto activated carbon

The formation of composite of Al_2O_3/ activated carbon is assumed as carbon metal composite which is dominated with metal aluminium oxide and carbon. Comparison amount of aluminium and carbon active in the mixture may support to increase the pores and loaded aluminium inside of pores [15]. The presence of ethanol and distilled water, in the hydrothermal process was assumed to initiate the bonding between activated carbon and metal oxide. However, the ethanol can be evaporating during the hydrothermal process [12].

Hydrothermal process can accelerate the evaporation of rest material inside the pore. Thereafter, the aluminium oxide filled the pore of activated carbon as an immobile material on the surface of activated carbon. During the hydrothermal process, the carbon-carbon bond was broken up and inserted by the aluminium oxide. This is due to the high pressure inside the hydrothermal. The assumption of reaction is illustrated in Figure 1. Interaction of functional group of activated carbon with Al_2O_3 can be illustrated in Figure 2. The result of reaction may increase the active size especially is oxygen functional.

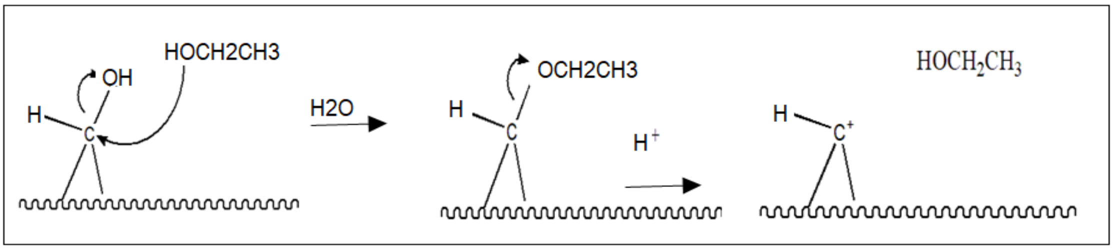

Figure1. Interaction of functional group of activated carbon and ethanol

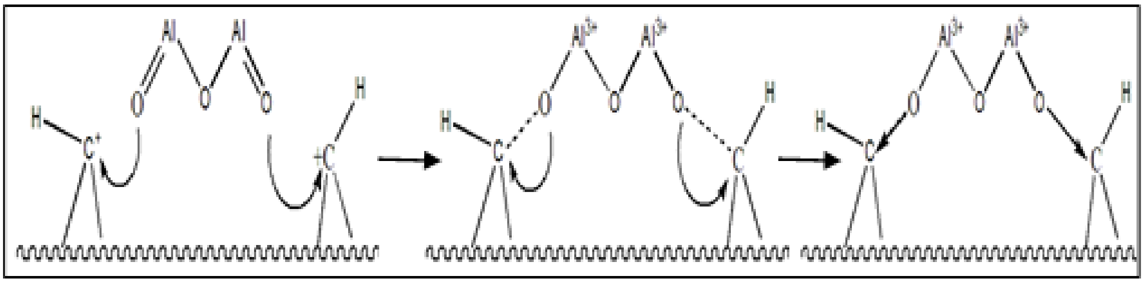

Figure 2. Interaction of Al_2O_3 with functional group of activated carbon

Morphology structure of activated carbon and composite Al_2O_3/activated carbon were applied by the SEM-EDX which is an important factor for studying the surface structure. Development of structure of activated carbon and composite Al_2O_3/ activated carbon can be shown in Figure 3. Development of pore of activated carbon was clearly seen on the surface with irregular size as displayed Figure 3.a. Some of metals oxide from Al_2O_3 was trapped on the pore surface. The Al_2O_3 appeared like a flower which grew up on the surface as shown in Figure 3 b and 3c.

Table 1. The elemental analysis of composite of Al_2O_3

Element Symbol	Atomic Conc	Weight Conc.	Oxide Symbol	Stoich. Wt.Coc
C	55.62	41.94	C	59.70
O	29.62	2.76		
Si	5.51	9.72	Si	13.83
K	3.84	9.42	K	13.40
Al	5.42	9.17	Al	13.06

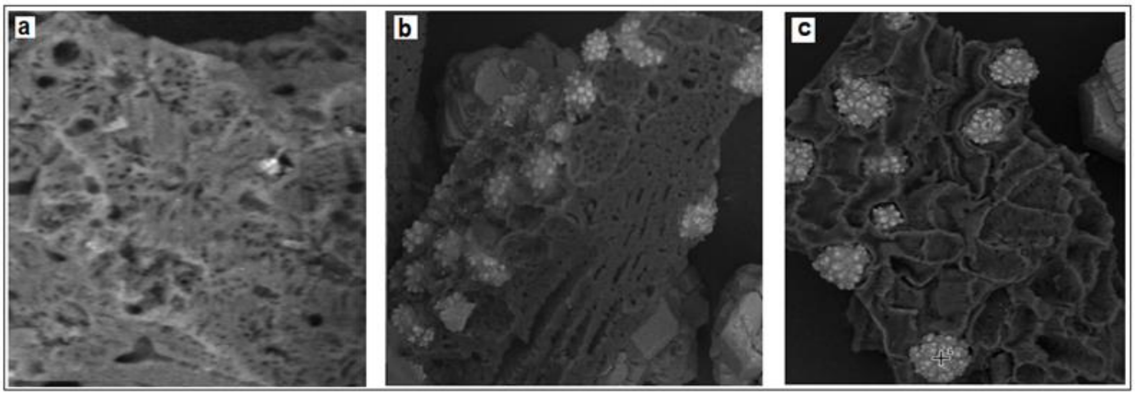

Figure 3. SEM morphology analysis a) activated carbon, b) composite of Al_2O_3/activated carbon, c) the growing of Al_2O_3

2. Adsorption process of phenol

The amount of phenol removal onto Al_2O_3/activated carbon was determined by the UV-Visible spectrophotometer. The experiment data were carried out by different of adsorption parameter involving pH solution, adsorbent dosage, contact time and concentration [3].

3. The effect of PH solution

The pH solution is one of essential factor for determination the optimum of adsorption phenol onto activated carbon. Initial condition of reaction was carried out with adsorbent dosage = 2 g, contact time, concentration 100 ppm. Percentage of removal phenol ws determined using the formula as Eg. 2 and 3.

$$\% R = \frac{Co-Ce}{C_o} \; x \; 100 \qquad (2)$$

$$q_e = \frac{Co-Ce}{m} \; x \; v \qquad (3)$$

The equilibrium concentration was performed as a unit mass of the removal percentage (% R), and the amount of the equilibrium adsorption, qe (mg/g) was calculated according to the following the equation (2) and (3), respectively.

The change of percentage adsorption of phenol is shown in Figure 4.

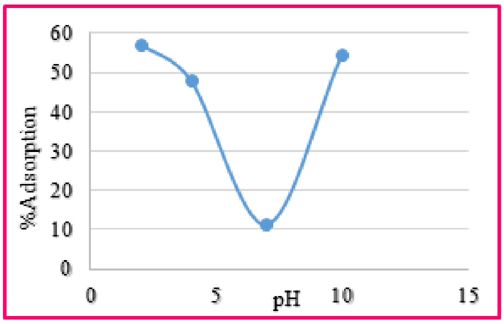

Figure 4. adsorption of phenol at different pH solution

The optimum adsorption was obtained at pH 2 with percentage adsorption = 57.13%. This pH was used for study the next equilibrium reaction for different parameters such as adsorbent dosage = 2 g, contact time, concentration 100 ppm. At the acidic condition phenol was change to be Phenolic ion with negative charge and aluminum hydroxide change to be Al^{3+} ion. The electrostatic force between phenolic ion and Al^{3+} ion achieved a maximum sorption of phenol onto composite. However, the sorption of phenol sharply decreases at increasing pH up to pH 7. At this moment, it assumed the Al^{3+} is not able to bond phenol instead of hydroxide ion to form

$Al(OH)_3$. At the basic condition pH 10, the aluminum was change to be $Al(OH)^{4-}$ with large of negative charge caused due to the increase of phenol adsorption. The reaction between phenol and activate size of composite can be illustrate as Figure 5.

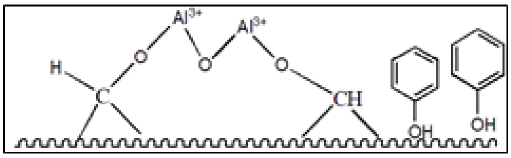

Figure 5. interaction between composite and phenol

4. The effect of adsorbent dosage

The effect of amount of adsorbent (0.5, 1.0, 1.5 , 2g) was carried at room temperature. The percentage of adsorption of phenol is shown in Figure 6. The adsorption of phenol sharply increases with the increase of adsorbent dosage with the maximum removal of 67.34 %. This result was assumed due to increase of activate size of composite.

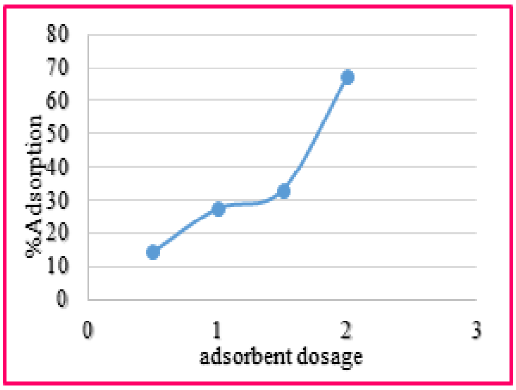

Figure 6. adsorption of phenol at different adsorbent dosage

5. The effect of concentration

The percentage of phenol removal versus concentration is shown in Figure 7. The amount of adsorption steadily increases with the increase of concentration. The maximum adsorption phenol was obtained at 100 ppm with sorption value of 53.69%.

6. The effect of contact time

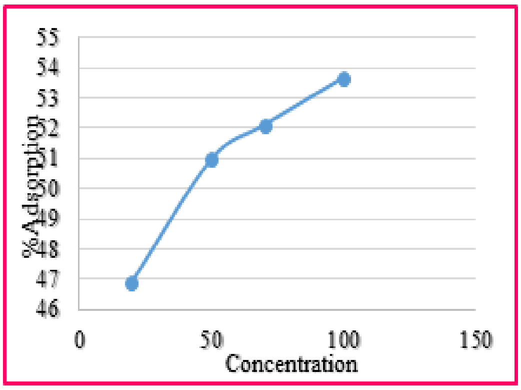

Figure 7. adsorption of phenol at different adsorbent dosage

The amount of phenol removal onto composite at different contact time was shown in Figure 8. The percentage of adsorption steadily increases with the increase of contact time. This reason can be described that increasing contact time may take any longer reaction occurred and give longer time to contact between phenol and composite in the adsorption process.

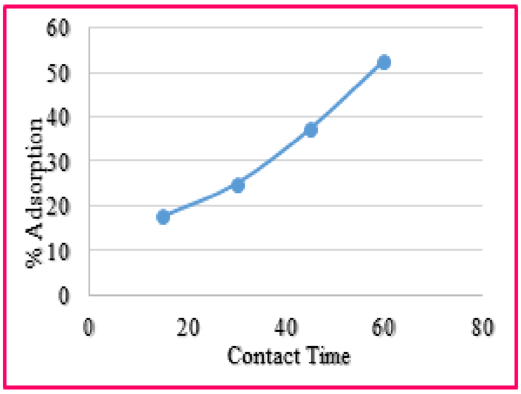

Figure 8. adsorption of phenol at different contact time

CONCLUSION

Composite of Al_2O_3/activated carbon has been successfully designed as an adsorbent to remove a phenol in aqueous solution. The optimum pH solution was obtained at pH 2 with the highest percentage of phenol removal. The influence of concentration, contact time and adsorbent dosage was evaluated and shown the adsorption of phenol onto composite steadily increase. The result show that the extension of aluminium oxide loaded onto activated carbon from palm kernel waste has a good prospect as an adsorbent.

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