

Full Length Research Paper

Application of activated carbon from empty fruit bunch (EFB) for mercury [Hg(II)] removal from aqueous solution

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Mercury is a heavy metal and is used widely in the industry, making it a global problem. It accounts for approximately 70% of man-made emissions. Activated carbon was found to be efficient for the adsorption of Hg(II) in aqueous solution. The characterization of Hg(II) uptake showed that the mercury binding is dependent on initial pH, agitation speed, amount of dosage and also the interaction between pH and contact time. From the experiment, the initial concentration of mercury was set to be 1.6 mg/L. It was found that the minimum residual concentration of mercury was 0.0075 mg/L (99.53%) at the condition of pH 6.5, agitation speed of 100 rpm, contact time of 70 min and AC dosage of 20 mg. This value was considered acceptable as it met the requirement of the Department of Environment, Malaysia.

Key words: Mercury, activated carbon, empty fruit bunch (EFB), aqueous solution.

INTRODUCTION

Mercury is a heavy metal and is used widely in the industry, making it a global problem. The major sources of mercury release according to a report published by the United Nations are coal-burning power plants and waste incinerators. They account for approximately 70% of man-made emissions. Besides that mercury is also being used to extract gold in gold mines and this pollutes the waterways and hence will combine to have an effect on humans and the environment. The presence of mercury in the water body can have serious effects on environment and human health and the full effect of consuming mercury is still being determined. Therefore, elimination or reduction of mercury in the water body is necessary to protect both the environment and public health. In Malaysia, the permissible discharge for mercury is 0.005 mg/L for standard A and 0.05 mg/L for standard B (Federal Subsidiary Legislation Malaysia, 1979). Humans are exposed to mercury through food, more specifically

fish. Fish can contain methyl mercury if the fishing areas are contaminated by it. Mercury exhibits extreme biological toxicity towards life forms (Ellis and Robert, 1996). The existing methods that are used to treat the removal of heavy metals from wastewater include precipitation, membrane filtration, ion exchange and adsorption. However, some of these methods are usually inefficient or too expensive to be used (Srinivasan et al., 1988; Arulanantham et al., 1989; Kadirvelu et al., 2000; Marshall et al., 1993). Therefore, powdered activated carbon (PAC) derived from empty fruit bunch (EFB) was used to study the potential of the adsorbent.

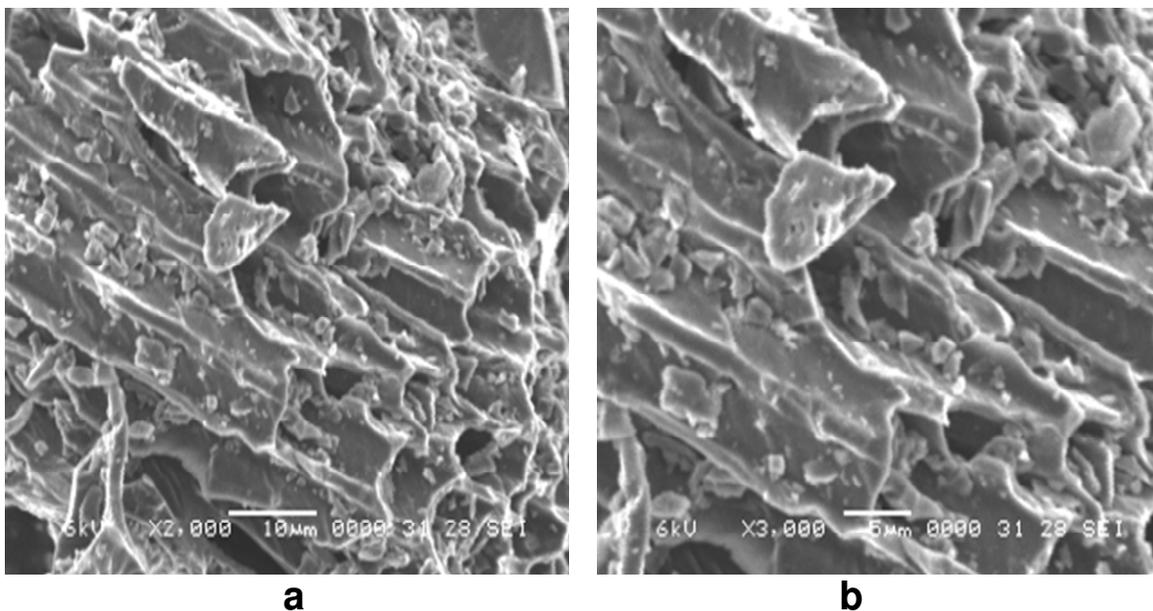
MATERIALS AND METHODS

The EFB sample was obtained from Seri Ulu Langat palm oil mill. The sample was dried in an oven at 105°C for 24 h in order to dehydrate the sample until a constant weight was been observed. The sample was then crushed, grinded and sieved to less than 250 µm particle size. The prepared EFB without the impregnation was carbonized with N₂ gas at 900°C for 30 min in the furnace. The N₂ flow rate was 0.25 L/min, then, it was activated with CO₂ gas for 15

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Table 1. Experiment parameters and their variation.

S/N	Parameter	Variation		
		Low (-1)	Medium (0)	High (1)
1	Dosage (mg)	5	7.5	10
2	pH	3	6	9
3	Time contact (min)	10	65	120
4	Agitation speed (rpm)	50	100	150
5	Concentration of mercury (mg/L)	0.1	0.1	0.1

**Figure 1.** SEM representative images of ACs after adsorption (2000 and 3000x).

min at a flow rate of 0.1 L/min. The temperature was set at 900 °C as it is the activation temperature.

Batch mode adsorption experiment

The adsorption experiment was carried out by taking 50 ml of mercury solution of desired concentration (0.1 mg/L) and desired weight of adsorbent using 100 ml conical flasks. Mercury aqueous solution was used in the experiments instead of water samples as the optimization of the experimental conditions can be observed clearly. The mercury sample was prepared by dissolving the mercury at a known quantity in deionized water. The working solutions were prepared by diluting the stock solution using distilled water. Afterwards, the conical flask was agitated at desired speed using mechanical shaker. The pH condition was adjusted by using 0.1 M sulphuric acid (H_2SO_4) or 0.1 M sodium hydroxide (NaOH).

Experimental design

Table 1 shows the experimental parameters (adsorbent dosage 10 to 30 mg, pH 5 to 8, contact time 20 to 120 min and the agitation speed 1.6 mg/L) and the variations that were used in the adsorption experiments were selected as the optimal condition in maximizing the removal of mercury. The number of runs for complete randomiz-

ed experiment was determined using Design Expert 6.0.8 central composite design software. Central composite is useful in this experiment as the product outcome has already been known and therefore screening was not required.

The design characteristics which were used to obtain a complete randomized experiment are as follows: 4 factors; half fraction; replicate 2; center point per block.

RESULTS AND DISCUSSION

Figure 1 shows the characterization of the activated carbon after batch mode adsorption study at different magnifications. It can be seen that after the batch mode study, the AC were more oriented and the porous structure could be clearly seen.

Analysis of batch mode adsorption experiment

The experimental result from Table 2 showed that the minimum residual occurred at pH 6.5, agitation speed of 100 rpm, contact time of 70 min and AC dosage of 20 mg.

Table 2. Result of the experiment.

Run	A: pH	B: Agitation speed (rpm)	C: Contact time (min)	D: Dosage (mg)	Hg(II) (mg/L)	Average (mg/L)	Removal (%)
1	5	50	20	10	0.704		
2	5	50	20	10	0.669	0.6865	57.09
9	5	50	120	30	0.104		
10	5	50	120	30	0.119	0.1115	93.03
5	5	150	20	30	0.342		
6	5	150	20	30	0.211	0.2765	82.72
13	5	150	120	10	0.073		
14	5	150	120	10	0.071	0.0720	95.50
17	6.5	100	70	20	0.004		
18	6.5	100	70	20	0.011	0.0075	99.53
3	8	50	20	30	0.470		
4	8	50	20	30	0.560	0.5150	67.81
11	8	50	120	10	0.516		
12	8	50	120	10	0.508	0.5120	68.00
7	8	150	20	10	0.536		
8	8	150	20	10	0.461	0.4985	68.84
15	8	150	120	30	0.063		
16	8	150	120	30	0.058	0.0605	96.22

At these conditions, the final concentration of mercury was 0.0075 mg/L where it managed to remove the mercury up to 99%. Furthermore, it showed that the AC had successfully removed the mercury and met the requirement of the DOE Malaysia regulations.

Effect of acidity (pH) on the uptake of mercury (II) ions

Figure 2 shows the residuals of mercury concentrations at different pH values, it can be observed that the lowest concentration removal of Hg(II) ions was at pH 6.5.

The adsorption capability of ACs is mainly determined by the functional groups introduced by oxidation. The attachment of the functional groups on ACs surface enhances the lead adsorption capacity. The increase of residual concentrations at pH 8 most likely reflects a reduction in the quantity of negative surface charges on ACs. The negative charge density on the surface of ACs decreases as the pH increases and this contribute to low adsorption of mercury.

Effect of agitation speed on the uptake of mercury (II) ions

It was observed that the residual concentration decreased at speed of 50 to 100 rpm and increases from 100 to 150 rpm.

In Figure 3, the lowest residual concentration occurred at mild agitation speed of 100 rpm. At the agitation speed of 100 rpm, the barrier between the solid-liquid phases

was overcome. Agitation at this speed led to the decrease of the boundary layer and increase in the transportation of Hg(II) ions at the adsorption site. Hence, the removal of Hg(II) ion from aqueous solution can be done at 100 rpm in order to achieve maximum removal of ACs. This speed has managed to overcome the resistance at the boundary layer between solid-liquid phases.

Effect of contact time on the uptake of Hg(II) ions

The equilibrium time was measured from 20 to 120 min and the effect of contact time was determined by plotting the residual concentration of Hg(II) against contact time as shown in Figure 4. Adsorption equilibrium time is defined as the time required for heavy metal concentration to reach a constant value. As shown in Figure 4, the removal of the residual increases as time increases.

Effect of AC dosage on the uptake of Hg(II) ions

The dosage can be related to the availability of the adsorption site for lead adsorption (Inglezakias et al., 2007). The relation was that with the increase of ACs dosage, the adsorption sites for Hg(II) ions increased, which greatly enhanced the adsorption of the mercury as seen in Figure 5.

Interaction between pH (A) and contact time (C)

Based on the ANOVA analysis, only in the interaction

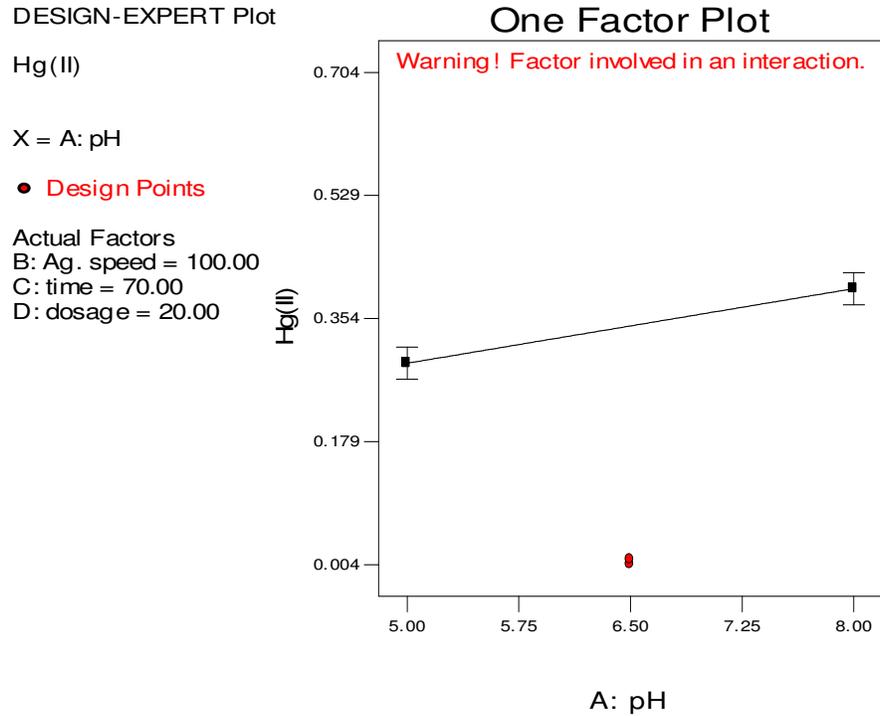


Figure 2. Effect of pH on uptake of mercury.

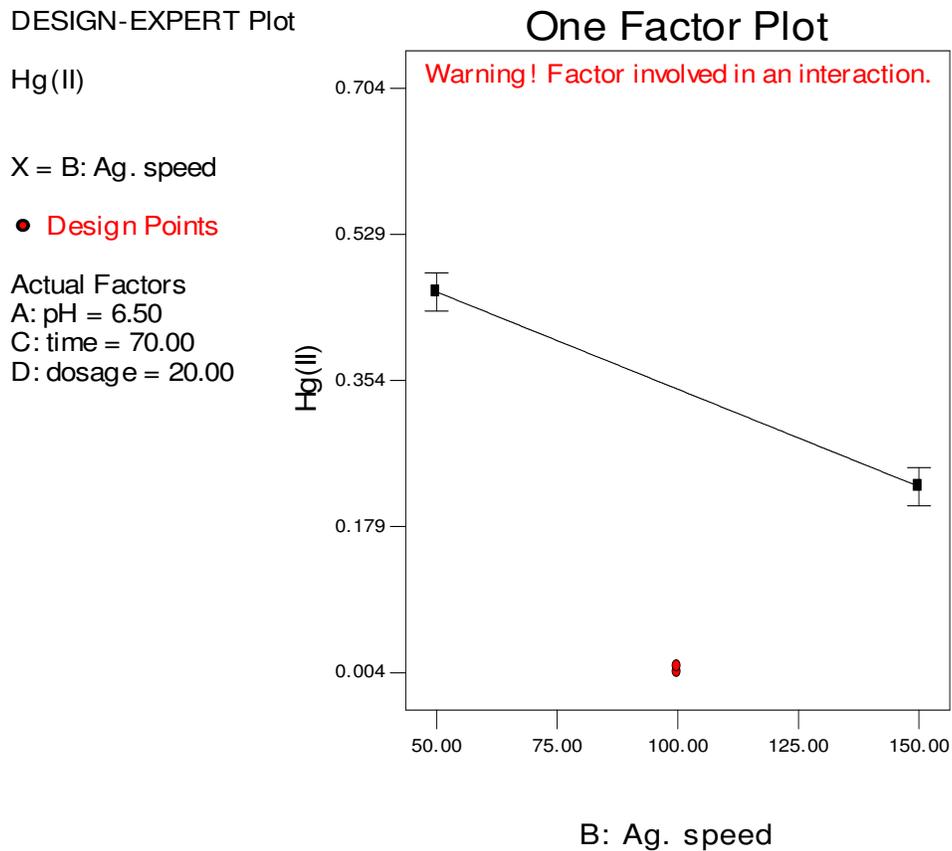


Figure 3. Effect of agitation speed on uptake of mercury.

DESIGN-EXPERT Plot

Hg (II)

X = C: time

● Design Points

Actual Factors

A: pH = 6.50

B: Ag. speed = 100.00

D: dosage = 20.00

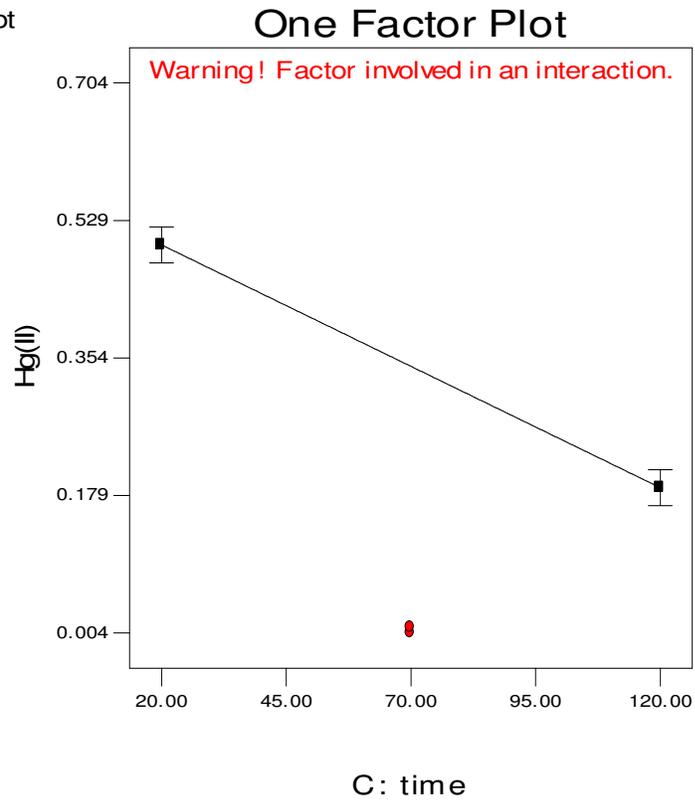


Figure 4. Effect of contact time on uptake of mercury.

DESIGN-EXPERT Plot

Hg (II)

X = D: dosage

● Design Points

Actual Factors

A: pH = 6.50

B: Ag. speed = 100.00

C: time = 70.00

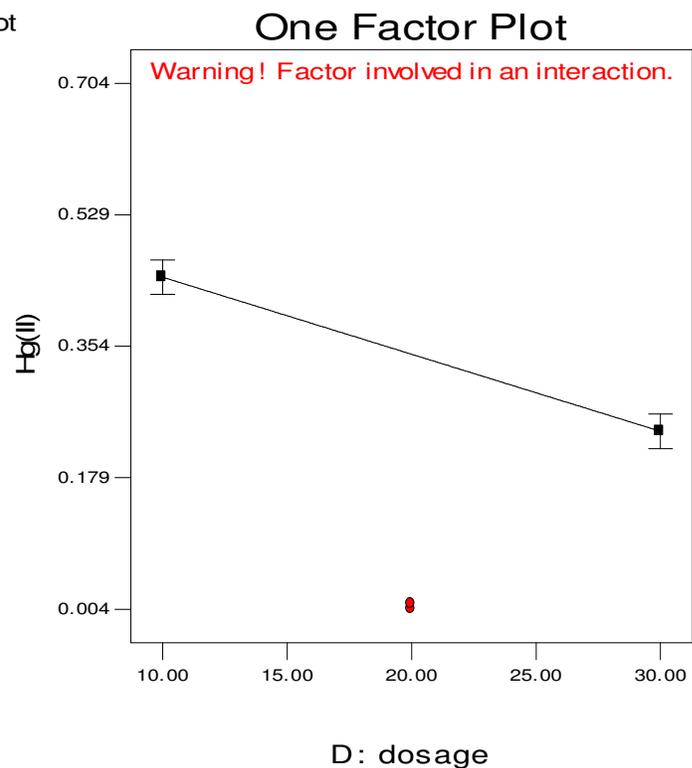


Figure 5. Effect of AC dosage on uptake of mercury.

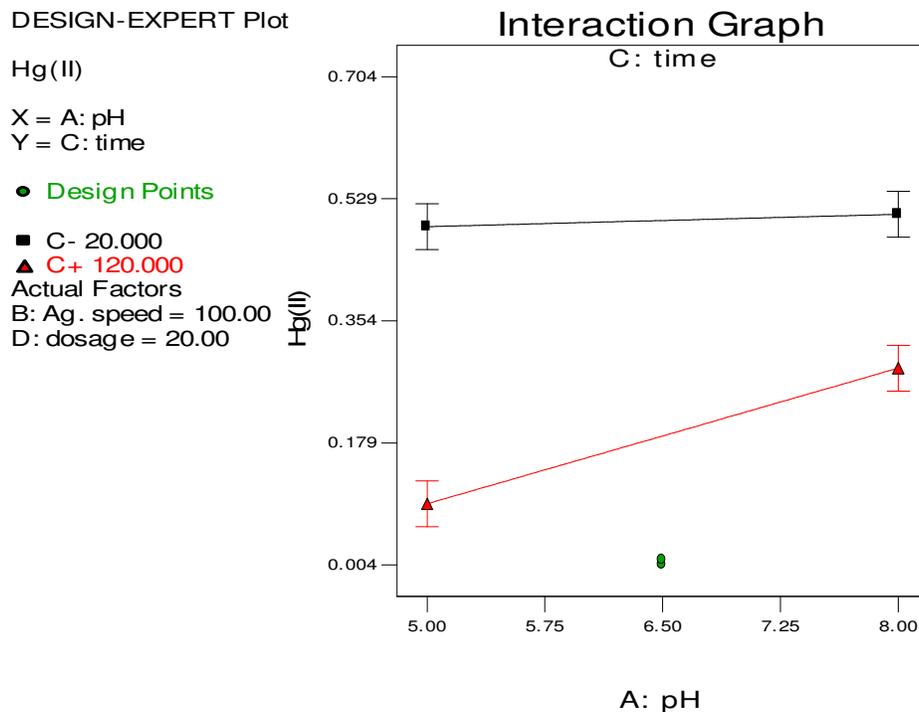


Figure 6. Interaction between pH (A) and contact time (C).

Table 3. Predicted values of mercury(II) residuals for optimization.

S/N	pH	Agitation speed	Time	Dosage	Hg(II)	Desirability
1	6.80	149.86	118.35	29.40	-0.0003	1.000
2	5.73	145.65	115.49	27.44	-0.0252	1.000
3	6.05	149.02	115.82	28.03	-0.0211	1.000
4	5.11	149.68	99.83	26.76	-0.0032	1.000
5	6.52	146.97	119.13	29.03	-0.0090	1.000
6	5.01	107.25	119.03	27.92	0.0028	1.000
7	6.67	149.71	119.91	28.01	0.0017	1.000
8	5.00	94.27	120.00	30.00	0.0087	0.993
9	7.58	150.00	119.66	30.00	0.0359	0.954
10	7.95	150.00	119.10	30.00	0.0596	0.921

model among AB, AC, AD, BC, BD and CD, model AC demonstrated to have significant effect on the model. Model AC is the interaction between pH and the contact time and this is shown in Figure 6.

The interaction model AC shows how at two different times, contact with different pH affects the residual concentration of the mercury. The first line (contact time of 20 min) showed that the removal of the mercury does not reduce much when the pH was varied from pH 5 to 8.

Optimum conditions for the removal of Hg(II) using ACs

The data were fitted into Design-Expert 6.0.8 to develop

a model that can be used to predict the highest percent removal. The predictive percent removal was calculated using this model to further determine the best conditions for the removal of mercury. The details of the predictive remaining concentration of mercury can be seen in Table 3.

Previously, there were many types of adsorbent that were used to remove Hg(II) from aqueous water. This shows that there have been great concerns on the removal of toxic metals from water due to its adverse effects on humans. Table 4 shows the types of adsorbent that were used to remove Hg(II) ions and the percentage uptake of Hg(II).

The percentage removal for each adsorbent is different

Table 4. Comparison between adsorbents for the removal of mercury.

Adsorbent	Condition			Removal (%)	Reference	
	pH	Contact time (min)	Dosage (mg)			
Sago waste	5	105	20	120	70	Kadirvelu et al., 2004
<i>Acetobacter xylinum</i>	5	10	10	-	78	Rezaee et al., 2005
<i>Pseudomonas putida</i>	6-6.5	10 days	10g	-	98	Canstein et al., 1999
Empty fruit bunch	6.5	70	20	100	99	This study

due to the variation in the operating parameters (pH, agitation speed, dosage, temperature, etc.). Thus, this comparative study was conducted to further understand the mechanism of adsorption and compare the types of adsorbents that were previously used to remove Hg(II). It can be seen in Table 4 that *Pseudomonas putida* gave the highest percentage among the three adsorbents that were used when compared with removal percentage of 98. However, considering the contact time of 10 days to achieve the highest removal, it may not be suitable for scale up purposes.

Conclusion

Activated carbon was found to be efficient for the adsorption of Hg(II) in aqueous solution. The characterization of Hg(II) uptake showed that mercury binding is dependent on initial pH, agitation speed, amount of dosage and also the interaction between pH and contact time. From the experiment, the initial concentration of mercury was set to be 1.6 mg/L. It was found that the minimum residual concentration of mercury was 0.0075 mg/L (99.53%) at the condition of pH 6.5, agitation speed of 100 rpm, contact time of 70 min and AC dosage of 20 mg. This value was considered acceptable as it met the requirement of the Department of Environment, Malaysia.

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