THE USE OF FACTORIAL DESIGN FOR ANALYSIS OF MERCURY REMOVAL EFFICIENCY USING PALM OIL FUEL ASH

Imla Syafiqah,Hafizuddin Wan Yussof*

Faculty of Chemical Engineering & Natural Resources, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia.
Corresponding Author Email: hafizuddin@ump.edu.my

ARTICLE DETAILS

ABSTRACT

A two-level half fractional factorial design was employed to evaluate the factors that have the greatest effect on the mercury removal efficiency. Several factors can influence the mercury removal using palm oil fuel ash (POFA). The aim of this study was to examine the main and interaction effects on mercury removal efficiency using POFA. The parameters which affect the mercury removal are pH (2 - 6), contact time (1 hr - 4 hr), initial concentration of the Hg^{2+} solution (1 mg/L – 5 mg/L), adsorbent dosages (0.10 g – 0.25 g) and agitation speed (100 rpm – 200 rpm) were investigated. Half factorial design using Response Surface Methodology (RSM) was used to determine the significant contribution of the above factors towards mercury removal efficiency. From the statistical analysis, the main factor of contact time and the interaction between adsorbent dosage and initial concentration of the Hg^{2+} resulted in the strongest effect on mercury removal. However, the interaction between agitation speed and pH was the least influencing factors. Based on the predicted and the experimental results presented, the experimental values were in good agreement with the predicted values proposed by the model with an error less than 10 % and proved to be an adequate model. The result indicated that fractional factorial design (FFD) was useful to improve the mercury removal efficiency by considering all the interactions of variables involved. In summary, this demonstrated that POFA could be potential as low-cost adsorbent for the removal of mercury.

KEYWORDS

1. INTRODUCTION

Activated carbon is mostly employed to remove wastewater pollutants. However, commercial activated carbon is expensive. Palm oil fuel ash is created from palm oil mills by the burning of oil palm shell and fibre used as fuel in boiler to generate steam [1]. The high cost of activated carbon has inspired a search for suitable inexpensive adsorbents. Adsorption is an effective and low-cost method for removal of heavy metals from water and wastewaters [2]. POFA are known for their good adsorption properties toward pollutants from the environment. Thus, many researchers are looking for cheaper substitutes, which are relatively inexpensive, and at the same time endowed with reasonable adsorptive capacity [3].

Mercury is an important environmental pollutant that had worldwide concern due to its volatility, persistence and bioaccumulation in the ecosystem [4]. The contamination of drinking water and other natural water resources is a severe problem, as they are non-biodegradable resources that can enter into the food chain. Thus, an effective technique should be developed for the selective monitoring and effective removal of mercury contamination in water or wastewater generated by both natural processes and human activities [5]. Currently, the primary processes employed for mercury removal in water systems are chemical precipitation, coagulation, ion exchange, adsorption, and biological methods [6].

A factorial design is a research in which only an effectively chosen fraction of the experimental combinations required for the complete factorial experiment is selected to be run [7]. The software provides a function known as full factorial design which is useful to study and determine the influences of several variables necessary in a process [8]. Besides, to solve this problem and obtain a probable optimum, design of experiment (DOE) offers a better alternative to study the effect of variables and their responses with minimum number of experiments. The software provides a function known as full factorial design which is useful to study and

determine the influences of several variables necessary in a process.

Although, there are few reports on mercury removal using POFA, no research has been found using response surface methodology to determine the interaction of several factors [9, 10]. The aim of this study was to examine the main and interaction effects on mercury removal efficiency using POFA. A 2^{5-1} factorial design were employed to examine the most significant factors among agitation speed, contact time, initial concentration of the Hg^{2+} solution, adsorbent dosages and pH which affect the mercury removal. A model was then developed and analysed using analysis of variance (ANOVA) to identify the correlation between experimental data and predicted value.

2. MATERIALS AND METHODS

2.1 Materials

POFA was collected from palm oil mill boiler at KilangSawitLepar, Kuantan. It was washed with deionized water for several times to remove foreign particles and oven dried overnight at 110°C to remove moisture. It was sieved through 100 μm and stored in airtight container.

2.2 Preparation of standard solution

A mercury stock solution was prepared at 1000 mg/L. This stock solution was then diluted to 1 mg/L and 5 mg/L using deionized water. All the adsorption experiment was carried out at room temperature. The required quantity of Hg^{2+} solution was dissolved in ultrapure water. Then, a small amount of HNO_3 was added to preserve the solution. The pH of the solution was adjusted using 0.1 M of HCl and 0.1 M NaOH. The mixture of the sample is shaken by using incubator shaker at the constant room temperature. The suspension is filtered using a vacuum pump and the filtrates are analysed using the direct mercury analyser RA-3310 (Nippon Instrument Corporation, Japan).

2.3 Characterizations of activated carbon

Particle sizes distributions of POFA were analyzed using Mastersizer 2000 (Malvern Instruments Ltd, UK). 50 mg of POFA sample is obtained using provided scoop and is placed into the vibratory hopper of Mastersizer. EVO 50 SEM (Carl Zeiss, Germany) instrument was used to study the morphology of the POFA. Small amount of POFA powder was poured on the carbon tape which is attached to the holder. Then the excess powder was blown off with air gun to ensure that only small pieces of the powder remain on the tape. After that, it was put into in the SEM chamber for analysis.

2.4 Experimental set up

The experimental setup used to evaluate the performance of mercury removal by POFA. Adsorption measurement is determined by a known amount of the sample with 50 mL of aqueous Hg^{2+} solution and placed into 250 ml of the conical flask. The mixture of the sample is shaken by using incubator shaker at the constant room temperature of 25℃. The suspension is filtered using a vacuum pump and the filtrates are analyzed using the direct mercury analyzer. The quantity of adsorbed mercury ion on adsorbent is calculated as the difference between initial concentration and concentration at any time.

2.5 Experimental design and statistical analysis

For the evaluation of five factors at two levels, 2^{5-1} factorial design with 16 experiments was run in a standard order as shown in Table 1. The resulted values and statistical analysis were processed using Design Expert software (Version 7.1.6, State-Ease Inc., Minneapolis, USA).

Table 1: 2^{5-1} Fractional factorial design matrix

Std. Order	Factors					Response
	pH	Contact time (h)	Initial concentration of the Hg^{2+} solution (mg/L)	Adsorbent dosage (g)	Agitation speed (rpm)	Mercury removal efficiency (%)
1	- 1	- 1	- 1	- 1	+1	74.67
2	+1	- 1	- 1	- 1	- 1	42.67
3	- 1	+1	- 1	- 1	- 1	97.33
4	+1	+1	- 1	- 1	+1	97.33
5	- 1	- 1	+1	- 1	- 1	73.67
6	+1	- 1	+1	- 1	+1	71.67
7	- 1	+1	+1	- 1	+1	65.33
8	+1	+1	+1	- 1	- 1	80.93
9	- 1	- 1	- 1	+1	- 1	59.67
10	+1	- 1	- 1	+1	+1	44.87
11	- 1	+1	- 1	+1	+1	74.33
12	+1	+1	- 1	+1	- 1	44.00
13	- 1	- 1	+1	+1	+1	37.73
14	+1	- 1	+1	+1	- 1	90.53
15	- 1	+1	+1	+1	- 1	98.03
16	+1	+1	+1	+1	+1	93.20
Factor	**Level**					
	-1		0		+1	
A - pH	2		4		6	
B - Contact time (h)	1		2.5		4	
C - Initial concentration of the Hg2+ solution (mg/L)	1		3		5	
D - Adsorbent dosage (g)	0.10		0.175		0.25	
E - Agitation speed (rpm)	100		150		200	

3. RESULTS AND DISCUSSION

3.1 Characterization of the materials

Figure 1 shows the particle size of POFA. The lowest and the highest percentages which are (d 0.1) and (d 0.9) of the POFA particle size are 5.307 μm and 239.117 μm, respectively. The average particle (d 0.5) size is 52.430 μm. The results reveal that, the mean particle size of POFA was 97.674 μm and the specific surface area was 0.487 m^2/g. In this study, the POFA had a smaller particle size. Some researchers identifies that for effective adsorption, small particle sizes and large surface areas are required for high adsorbate removal at equilibrium[11]. It is confirmed that the larger the specific surface area and the finer the particle size distribution of adsorbent, the greater its adsorption capacity and interaction with an adsorbate.
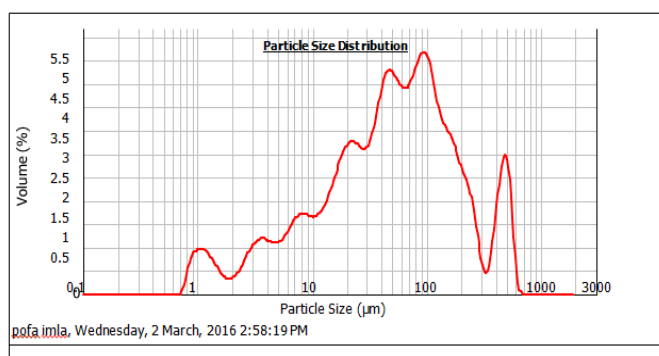
Figure 1: Particle size distribution profile for POFA
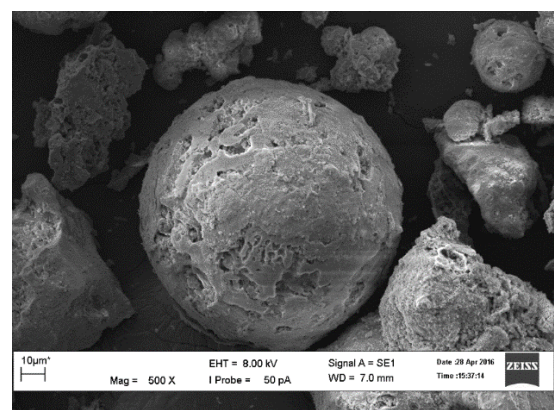

Figure 2: SEM image of raw POFA

Figure 2 shows the SEM results of the raw POFA particles. The image was captured at 500x magnification. The particles were irregular in shape and having porous texture. In addition, there was no agglomeration of POFA particles after the heat treatment. Previous studies have reported by other researcher where the ash consists of a series spherical vitreous particle of different size[12]. It has been seen clearly that the ash particles were mainly made up of irregular spherical, semi-spherical, and highly porous in nature which is a desirable characteristic of any adsorbents.

3.2   Factor affecting mercury removal efficiency

The experimental design using half level factorial analysis was carried out to determine the factors affecting mercury removal efficiency during adsorption process. These five factors include pH, contact time, initial concentration of the Hg^{2+} solution, adsorbent dosage, and agitation speed. Table 1 shows that the highest percentage of mercury removal was obtained at 98.03 % at 100 rpm of agitation speed with pH of 2 °C, 5 mg/L of initial concentration of the Hg^{2+} solution, and 0.25 g of adsorbent dosages for 4 hours in contact time.

In factorial analysis, the contribution of the main factors gives an important effect on the optimization part later. Several factors that highly contributed to mercury removal will be selected from this factorial analysis. From Table 2 and Figure 1, factor B gives the highest contributing factor of 22.76% in the mercury removal. Similar results are reported by some group researchers where the adsorption of metals and removal of mercury increases as a function of time [13,14]. Factor C recorded as the second highest contribution factor of 5.50 %. Meanwhile, factor D also contributed in the mercury removal about 3.55 % as its lies above of the t-values line.

Table 2: Percentage contribution of factors on response variables

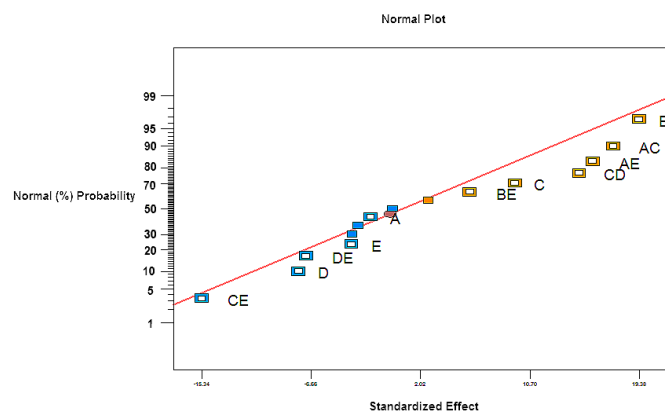| Factors	Percentage contribution (%)
	Mercury removal efficiency
A - pH	0.23
B - Contact time	22.76
C - Initial concentration of the Hg^{2+} solution	5.50
D - Adsorbent dosage	3.55
E - Agitation speed	0.73
AB	0.53
AC	18.22
AD	0.43
AE	14.94
BC	0.70
BD	2.075×10^{-3}
BE	2.14
CD	12.97
CE	14.28
DE	3.02

Figure 3: Normal plot

3.3   Normal plot of residuals

From normal plot as shown in Figure 3, factors B and C gave a positive effect (refer to orange colour) to the mercury removal. This suggested that the highest values will favoured the response. Thus, an increase in B increases the adsorption of mercury solution. For main effects, an effect is said to be positive when an increase to its high level will cause an increase in the response, while negative effect is when an increase to its high level will result a decrease in the response. The experimental data for the mercury removal efficiency from the empirical model is in good agreement with the observed ones in the range of the operating factors.

4.   CONCLUSION

Two level half factorial design screening was successfully employed to determine the significant factors, which are pH, contact time, initial concentration of the Hg^{2+} solution, adsorbent dosage and agitation speed in mercury removal. The results obtained from this study showed that factors contact time, initial concentration of the Hg^{2+} solution and adsorbent dosage were among the highest contributionon mercury removal.In summary, this demonstrated that POFA could be potential to be used as low-cost adsorbent for the removal of mercury.

REFERENCES

[1] Khanday, W., Marrakchi, F., Asif, M., Hameed, B.H. 2016. Mesoporous zeolite–activated carbon composite from oil palm ash as an effective adsorbent for methylene blue. Journal of the Taiwan Institute of Chemical Engineers, 70 (2), 32-41. doi: 10.1016/j.jtice.2016.10.029

[2] Elhami, S., Shafizadeh, S. 2016. Removal of mercury (II) using modified Nanoclay. Materials Today: Proceedings, 3(8), 2623–2627. doi: 10.1016/j.matpr.2016.06.005

[3] Ahmad, A.A., Hameed, B.H., Aziz, N. 2007. Adsorption of direct dye on palm ash: Kinetic and equilibrium modelling. Journal of Hazardous Materials, 141(1), 70-76. doi: 10.1016/j.jhazmat.2006.06.094

[4] Zhang, X., Shen, B., Zhu, S., Xu, H., Tian, L. 2016. UiO-66 and its Br-modified derivates for elemental mercury removal. Journal of Hazardous Materials, 320, 556–563. doi: 10.1016/j.jhazmat.2016.08.039.

[5] Liu, Y., Wang, X., Wu, H. 2017. Reusable DNA-functionalized-graphene for ultrasensitive mercury (II) detection and removal. Biosensors and Bioelectronics, 87 (1), 129-135. doi: 10.1016/j.bios.2016.07.059

[6] Lu, X., Huangfu, X., Ma, J. 2014. Removal of trace mercury (II) from aqueous solution by in situ formed Mn–Fe (hydr)oxides. Journal of Hazardous Materials, 280, 71-78. doi: 10.1016/j.jhazmat.2014.07.056

[7] Sibanda, W., Pretorius, P. 2011. Application of two-level fractional factorial design to determine and optimize the effect of demographic characteristics on HIV prevalence using the 2006 South African Annual Antenatal HIV and Syphilis Seroprevalence data.International Journal of Computer Application, 35(12), 15-20.

[8] Samad, K.A., Zainol, N. 2017. The use of factorial design for ferulic acid production by co-culture. Industrial Crops and Products, 95 (1), 202-206. doi: 10.1016/j.indcrop.2016.10.028

[9] Ahmad, T., Rafatullah, M., Ghazali, A., Sulaiman,O., Hashim, R.2011. Oil palm biomass–based adsorbents for the removal of water pollutants: A review. Journal of Environmental Science and Health, Part C, 29(3), 177-222. doi: 10.1080/10590501.2011.601847

[10] Attari, M.,Bukhari, S.S., Kazemian, H., Rohani,S. 2017. A low-cost adsorbent from coal fly ash for mercury removal from industrial wastewater. Journal of Environmental Chemical Engineering, 5(1), 391-399. doi: 10.1016/j.jece.2016.12.014

[11] Bada, S.O., Potgieter-Vermaak, S. 2008. Evaluation and treatment of coal fly ash for adsorption application. Leonardo Electronic Journal of Practices and Technologies, 1 (12), 37-48.

[12] Al Bakri, A.M.M., Kamarudin, H., Bnhussain, M., Nizar, K.I., Rafiza, A.R., Zarina, Y. 2012. The processing, characterization, and properties of fly ash based geopolymer concrete. Reviews on Advanced Materials Science, 30, 90-97.

[13] Kuncoro, E.P., Fahmi, M.Z. 2014. Kinetics of Hg and Pb removal in aqueous solution using coal fly ash adsorbent. Journal of Proceeding Series, 1, 1-4.

[14] Verma, V.K., Tripathi, I.N. 2014. Kinetic study on the removal of mercury by fly ash. Global Nest Journal, 16(2), 385–392.