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Original Research Article

The Effect of Contact Time and Oil Palm Frond Activated Carbon Dose as an Adsorbent in Decreasing Iron (Fe) in Groundwater

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Abstract
Groundwater well is the alternative well providing water source in the city of Balikpapan, especially in its sub-district of Karang Joang, North Balikpapan, where the location has not been fully integrated with PDAM water pipes. Normally, the groundwater has minerals including iron (Fe). For the drinking water, the concentration standard of Fe follows the national law of Permenkes No. 493/Menkes/Per/IV/2010 which is 0,3 mg/L as the threshold. Oil palm frond is an organic matter that is found easily in East Kalimantan, containing high levels of lignin and cellulose which used to produce activated carbon. The activated carbon was evaluated with five variant doses of 5; 10; 15; 20; and 25 g/L over groundwater and the contact time of 30; 45; 60; 75; and 90 minutes. The time contact was measured by the rotation of the stirrer with the speed of 100 rpm. The results showed the optimum time contact of 30 minutes with the adsorbent dose of 5 g/L with the elimination efficiency of 99,56%. The time contact and the adsorbent dose variants have insignificant effect because of the least time contact as well as the least dose are the optimal condition in decreasing Fe.

Keywords: Activated carbon; Fe; Groundwater; oil palm frond

1. Introduction

Groundwater wells are the common alternatives for providing clean water in Balikpapan City, especially in Karang Joang sub-district, North Balikpapan. The location is a sub-urban area without full access to clean water from the Municipal Waterworks (PDAM). Artesian or groundwater well is one way of exploiting groundwater, where the infiltrated water will reach a saturation point so that it cannot penetrate the ground. High Fe levels in the water have a negative impact on people who consume this water without further processing. Water that contains iron levels exceeding quality standards can be harmful to the human body because it causes genetic abnormalities in chromosomes, failures in iron metabolism, which eventually accumulates the iron in the brain and causing Alzheimer's and Parkinson's disease (Belaidi & Bush, 2016). The quality of drinking water has been regulated by the quality standards set in Permenkes No. 493/Menkes/Per/IV/2010 concerning Drinking Water Quality Requirements with a threshold of 0.3 mg/L.

Adsorption is a method that can be used to reduce the Fe content. It is conducted by adding adsorbent to the water. Commonly, the adsorbent uses activated carbon. Activated carbon can be produced by carbonizing the organic materials. Activated carbon is considered effective in reducing Fe levels. Several studies show that activated carbon has proven effective in reducing Fe content. Such as research conducted by (Syauqiah et al., 2011), they succeeded in reducing the Fe content of waste water according to the target (below water quality standards). It's made the waste water safe for discharge into water bodies. With a contact time of 80 minutes and 700 rpm of stirring, activated carbon made from oil palm shells reduces 92.7803% of the Fe content in groundwater so that the quality of the groundwater meets the quality standards (Sirajuddin et al., 2022).

For activated carbon, there are three of activation process: physical, chemical and combined physical-chemical processes. The physical activation process is carried out by burning material in a furnace. The chemical activation process is carried out by adding certain chemical compounds to the carbon, there are NaOH, KOH, H_3PO_4, Na_2CO_3, and others. The properties of activated carbon itself are not only influenced by the type of raw material, surface area, distribution of pores and surface chemical properties of activated carbon, but are also influenced by the activation method used. H_3PO_4 has preferences compared to others since H_3PO_4 as a catalyst not as it were advances bond cleavage responses but moreover encourages crosslinking through cyclization, condensation, and shaping a layer of linkage such as phosphate and polyphosphate esters which seem secure the inside pore structure and, in this way, avoid over the top burn-off in carbon actuation. With the contemplations of natural affect, vitality fetched, and carbon abdicate, H_3PO_4 is profoundly appealing and has been progressively utilized in large-scale AC generation in later decades (Yang et al., 2020).

Oil palm fronds can be used as ingredients in the production of activated carbon due to its high content of lignin (14%), cellulose (31.5%), hemicellulose (19.2%), and lignin (14%) (Maulina & Iriansyah, 2018). In addition, In East Kalimantan, the quickest advancement of oil palm plantations is in Kutai Kartanegara district, which position for the area of oil palm plantations after the East Kutai district. In 2017, Kutai Kartanegara district had an oil palm plantation area of up to 217,000 ha (Damanik et al., 2020). Therefore, oil palm fronds are easy to find in the study area.

This research aims to determine the effect of contact time and oil palm frond activated carbon dose as an adsorbent in decreasing iron (Fe) in groundwater. The variations of contact time are 30; 45; 60; 75 and 90 minutes and the doses of oil palm fronds activated carbon are 5; 10; 15; 20; and 25 g/L. This research use groundwater in Karang Joang (around KM. 8 area), North Balikpapan. The effect of variation can be seen from the value of the effectiveness of Fe removal from activated carbon oil palm fronds.

2. Methods

2.1. Ground Water Collection

The groundwater used in this study obtained from Al-Hijrah Mosque, located on Jl. Kesatriaan, KM 08, Karang Joang sub-district, North Balikpapan. The groundwater was collected from the groundwater well using sampling techniques in accordance with Indonesian national standards (Standar Nasional Indonesia Metode Pengambilan Contoh Air Tanah, 2008). The guideline standard contains a method for extracting wells using a faucet. Therefore, the groundwater from the well was collected by flowing the groundwater for 2 minutes before being taken, and then the groundwater is collected in jerry cans

2.2. Activated Carbon Sample Preparation

The oil palm fronds come from privately owned plantations in Karang Joang sub-district, Balikpapan. Then it washed and cut into 1 cm sizes and then dried conventionally under the sun. After that, drying process was continued using the oven at 105ºC for 2 hours. The oil palm fronds were then carbonized in the furnace with the temperature of 500 ºC for 15 minutes. Carbon was crushed using a grinding machine and then sieved with a 100 mesh siever. The carbon was activated using 0.1 M H3PO4 solution for 22 hours with a mass ratio 3:1 of carbon and solution. The activated carbon was washed using distilled water and NaOH solutions until the activated carbon pH reached between 6-7. The carbon was dried repeatedly until reaching an equilibrium weight.

2.3. Moisture content (SNI 06-3730-1995)

Following the Indonesian standard of SNI 06-3730-1995, the activated carbon sample was tested for its water content by evaporating water on the carbon by heating it using an oven for 3 hours at 110˚C. The activated carbon is weighed at 2 g, and then it was heated for 3 hours (W1). After that, it is weighed again to determine the difference in weight. The calculation of water content (dry basis) is carried out using the equation (1):

$$Water\ content\ (\%) = \frac{W_1}{W_2} \times 100\% \quad (1)$$

2.4. Ash content (SNI 06-3730-1995)

The activated carbon is weighed at 2 g (W1) and put into a porcelain cup and then burnt in a furnace at 800˚C for 2 hours. The ash resulted from the process was weighed (W2) to calculate the mass difference before and after the process. The ash content was calculated using the equation (2):

$$Ash\ content\ (\%) = \frac{W_1}{W_2} \times 100\% \quad (2)$$

2.5. Efficiency adsorption analysis

The data analysis measured from the laboratory was used to calculate the effectiveness of iron content removal in the groundwater. The iron content was measured before the adsorption process (Co). After that, the Fe content was re-measured (Ce) so that the removal effectiveness can be calculated with the following equation (3):

$$Fe\ removal\ (\%) = \frac{(Co - Ce)}{Co} \times 100\% \quad (3)$$

Variations made in this study were the stirring time of 30; 45; 60; 75; and 90 minutes, and the activated carbon doses of 5; 10; 15; 20; and 25 g/L. After calculating the effectiveness value, the optimum contact time and the adsorbent dose was analyzed using the SPSS software.

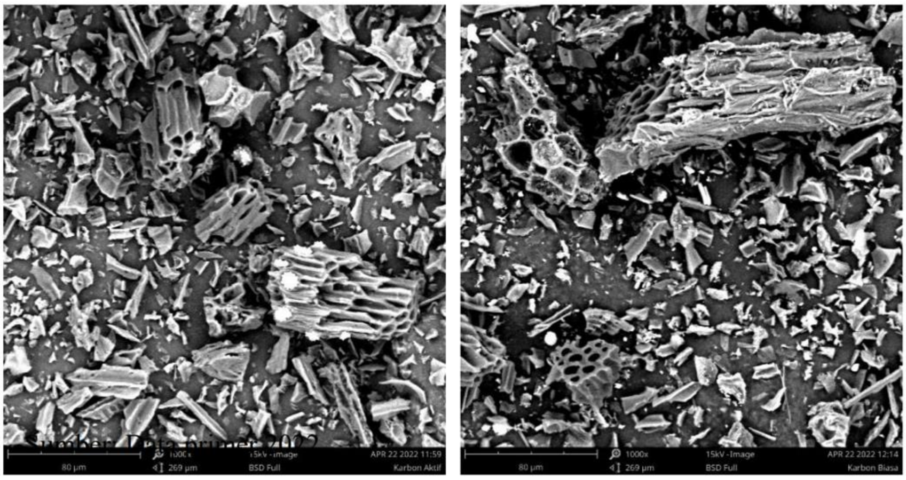
2.6. ANOVA one-way test

Statistical analysis testing in this study was to determine the effect of contact time and adsorbent dosage used during processing on Fe removal in groundwater. The ANOVA one-way test was applied from the SPSS 25.0. The confidence interval used is 95% with a significance level of 0.05.

3. Result and Discussion

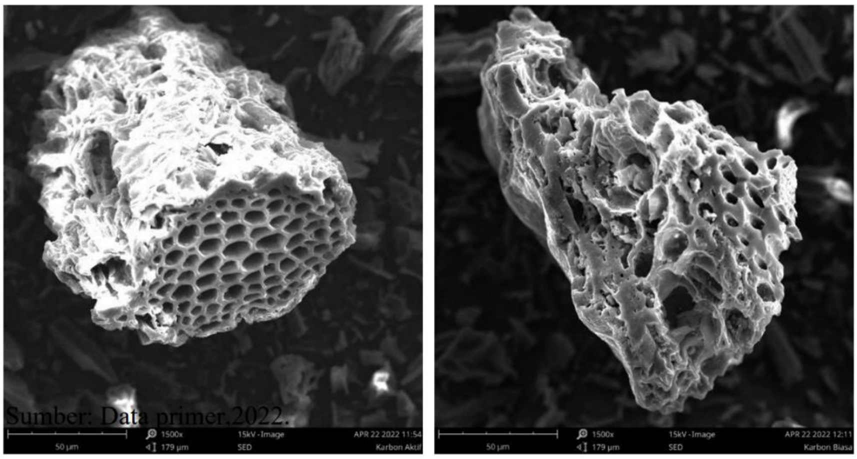
3.1. Characteristics of activated carbon

Activated carbon is chemically activated using a H_3PO_4 solution with the aim of degrading organic molecules, limiting the formation of tar, decomposing organic compounds, and dehydrating water trapped in carbon cavities so that it can help remove hydrocarbon deposits produced during carbonization. The result of visual observation shows in Figure 1 (1000x SEM magnification) dan figure 2 (1500x SEM magnification).



(A) (B)

Figure 1. Carbon surface shape from SEM 1000X magnification; (a) Activated carbon and (B) Carbon



(A) (B)

Figure 2. Carbon surface shape from SEM 1000X magnification; (A) Activated carbon and (B) Carbon

Figure 1 dan 2 shows the difference between carbon and activated carbon. Carbon still contains impurity particles and have smaller pore sizes, whereas activated carbon tends to be cleaner with larger pores. This is because activated carbon has been activated with H_3PO_4 solution. In the manufacture of activated carbon needs to be activated. Activation will affect the increase in carbon absorption. The activation process will cause the impurities covering the carbon pores to also evaporate (detach) with increasing activation temperature (Idrus et al., 2013). H_3PO_4 has several advantages compared to other activators, which are more friendly to the environment, requires low energy in the activation process, and is more economical. Another advantage of using H_3PO_4 is that it can produce activated carbon material that has larger pores than some other chemical activators (Riyanto et al., 2021).

Evaluation on the characteristics of oil palm frond activated carbon on this research was conducted to ensure that the activated carbon produced meets the standard of SNI 06-3730-1995. The results of analyzation for water and ash contents are written in the Table 1.

Table 1. Water and ash content results of the oil palm activated carbon

Parameter	Maximum content of SNI (%)	Carbon (%)	The activated carbon (%)
Water Content	15	8%	0.31
Ash Content	10	11%	4.87

The results from the research experiment (Table 1) showed the activated carbon produced in this study meets the requirements standards of SNI No.06-3730-1995 for the parameters of water content and ash content, so that the activated carbon can be used as an adsorbent. The water content in carbon is higher than activated carbon. The amount of water content after activation was tested on activated carbon to determine the hygroscopic properties of activated palm frond carbon. The decrease in water content is also closely related to the hygroscopic nature of the H_3PO_4 activator. H_3PO_4 very strong dehydrating agent to a constant weight of activated carbon. The binding power of water by H_3PO_4 is stronger than HCl and H_2SO_4. The binding of water molecules in the activated carbon by the activator causes the pores in the activated carbon is bigger. The larger the pores, so surface area of active carbon will increase. This increase in surface area results in an increase in adsorption capacity of activated carbon. Increasing the adsorption ability of activated carbon, the better the quality of the activated carbon. The decrease in the ash content occurs because the metals and metal oxides bound to the carbon dissolve in the activator thereby opening the pores of the carbon. Carbon activation with H_3PO_4 activator ash content in activated carbon is very small. This happens because H_3PO_4 is able to dissolve metals and metal oxidants better than H_2SO_4 and HCl (Pujiono & Mulyati, 2017).

3.2. Effect of adsorbent dosage on Fe removal

Figure 1 presents the percentage of Fe removal with various doses of the activated carbon on 5; 10; 15; 20; 25g/L with different contact time (a) 30, (b) 45, (c) 60, (d) 75, (e) 90 minutes. It can be seen that the activated carbon of 5 g/L has the highest rate of Fe removal in various time. Once the dose of the activated carbon is increased, the percentage of Fe removal trend decreases. Therefore, the addition of a larger dose is not optimal in absorbing iron levels. At the dose of 5 g/L, the adsorption ability at a longer contact time has a decreasing trend of adsorption ability due to desorption. Meanwhile at a dose of 10 g/L, the absorption ability at various times showed that the absorption ability at 60 minutes contact time was better than that of 45 minutes contact time. At contact time 60, the removal of Fe was 97.6% whereas at the contact time of 45 minutes the remaining Fe content was 97.2%. At a dose of 15 g/L, the most optimum entrapment was found at the contact time of 30 minutes with a removal percentage of 99%. Optimum adsorption at a dose of 20 g/L occurred at the contact time of 30 minutes, with the residual iron content elimination of 99.1%. Optimum absorption at a dose of 25 g/L was found at a contact time of 30 minutes with the Fe removal content of 98.9%.

From the results of the research conducted, it can be seen that the larger the dose, the smaller the adsorption capacity. The addition of the dose that is done actually reduces the effectiveness of absorption. This is of course different from the several studies that have been conducted, that increasing the dose should increase the effectiveness of absorption. This can be caused by the large number of adsorbents that can saturate the surface of activated carbon, so that the adsorption capacity decreases (Abdi, 2016). In addition, interactions between adsorbents and other adsorbents can occur due to the large amount of adsorbent mass which results in absorption by the adsorbent itself (Jubilate et al., 2016). Of course, further research needs to be carried out to prove this phenomenon, because in the lowest dose of activated carbon from palm fronds (5 g/L) with a contact time of 30 minutes, and a stirring speed of 100 rpm, optimal absorption conditions of 99.53% have been obtained.

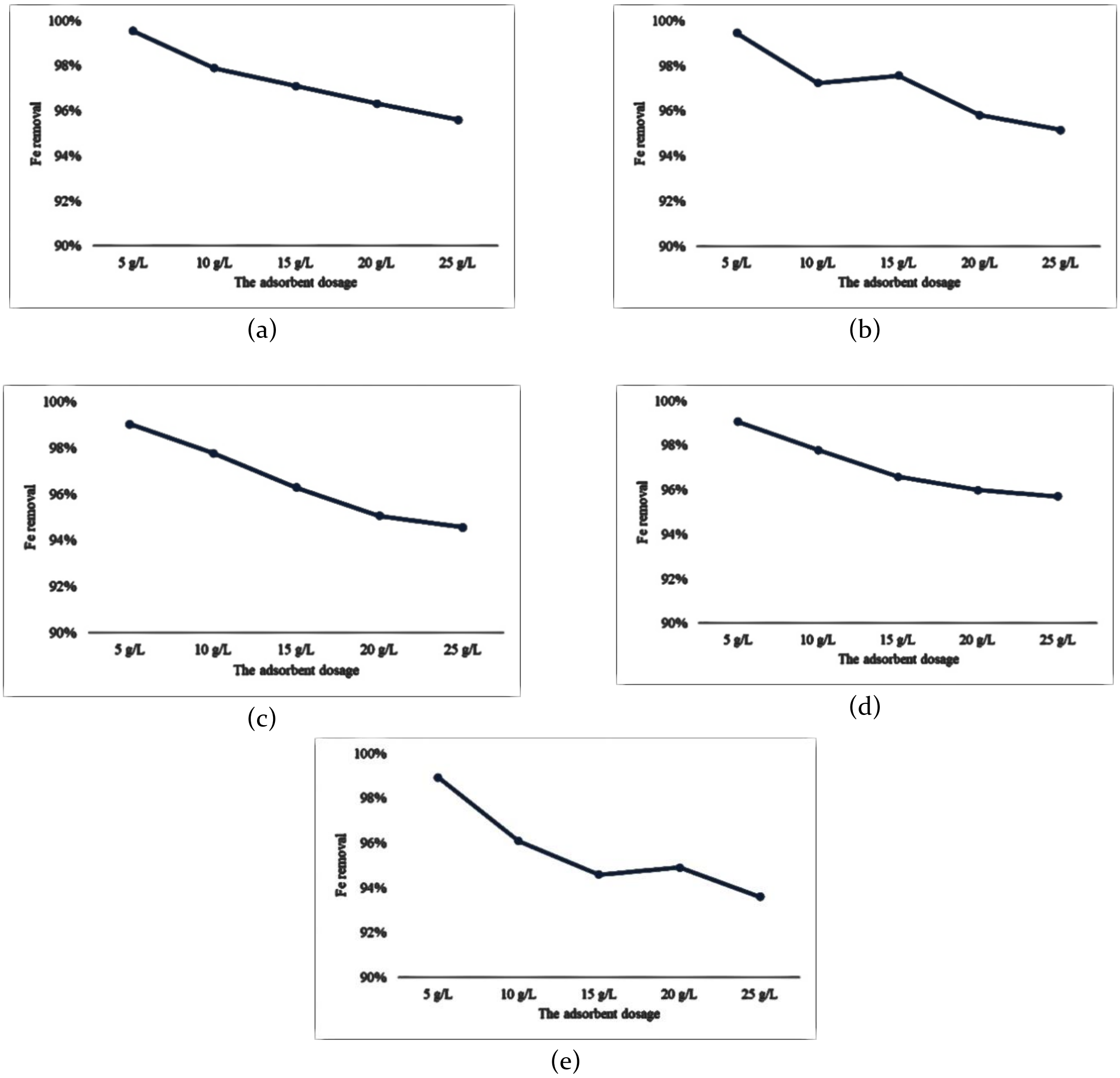


Figure 2. Presents the percentage of Fe removal with various doses of the activated carbon on 5; 10; 15; 20; 25g/L with different contact time (a) 30, (b) 45, (c) 60, (d) 75, (e) 90 minutes

3.3. Effect of adsorbent dosage on Fe removal

The Figure 2 the percentage of Fe removal with various contact time on 30; 45; 60; 75; 90 minutes with different dose of activated carbon (a) 5, (b) 10, (c) 15, (d) 20, (e) 25 g/L. It was found that the most effective contact time in eliminating Fe levels was 30 minutes at the dosage of 5 g/L. The contact time of 90 minutes is the least optimum variation for all doses. This can be resulted from the saturated adsorbent produced from a too long contact time, effecting the released adsorbate so that the addition of time has no effect on the Fe adsorption. The adsorption ability decreases over the time because saturation in the adsorption process has been reached resulted from active groups components filled the adsorbent. The decrease in adsorption efficiency is possible due to the desorption process or the release of the adsorbate again during stirring. Desorption occurs due to a saturated adsorbent surface. In the saturated state, the adsorption rate decreases so that the contact time is no longer affected (Ariansyah Lubis & Ali Hasimi Pane, 2021).

With the rising time, the number of empty sites decreases so that the adsorption became thinner, resulting in repulsion between the adsorbate on the surface of the adsorbent and the adsorbate (Reza Pahlepi et al., 2019). Eventually, the pores on the remaining free surface are almost not able to be filled due to the repulsive forces between the adsorbed pollutants and the pollutants in the liquid phase

(Khaniabadi et al., 2016). Hence, the most optimal time in this test is the fastest time, which is 30 minutes with an absorbed dose of 5 g/L.

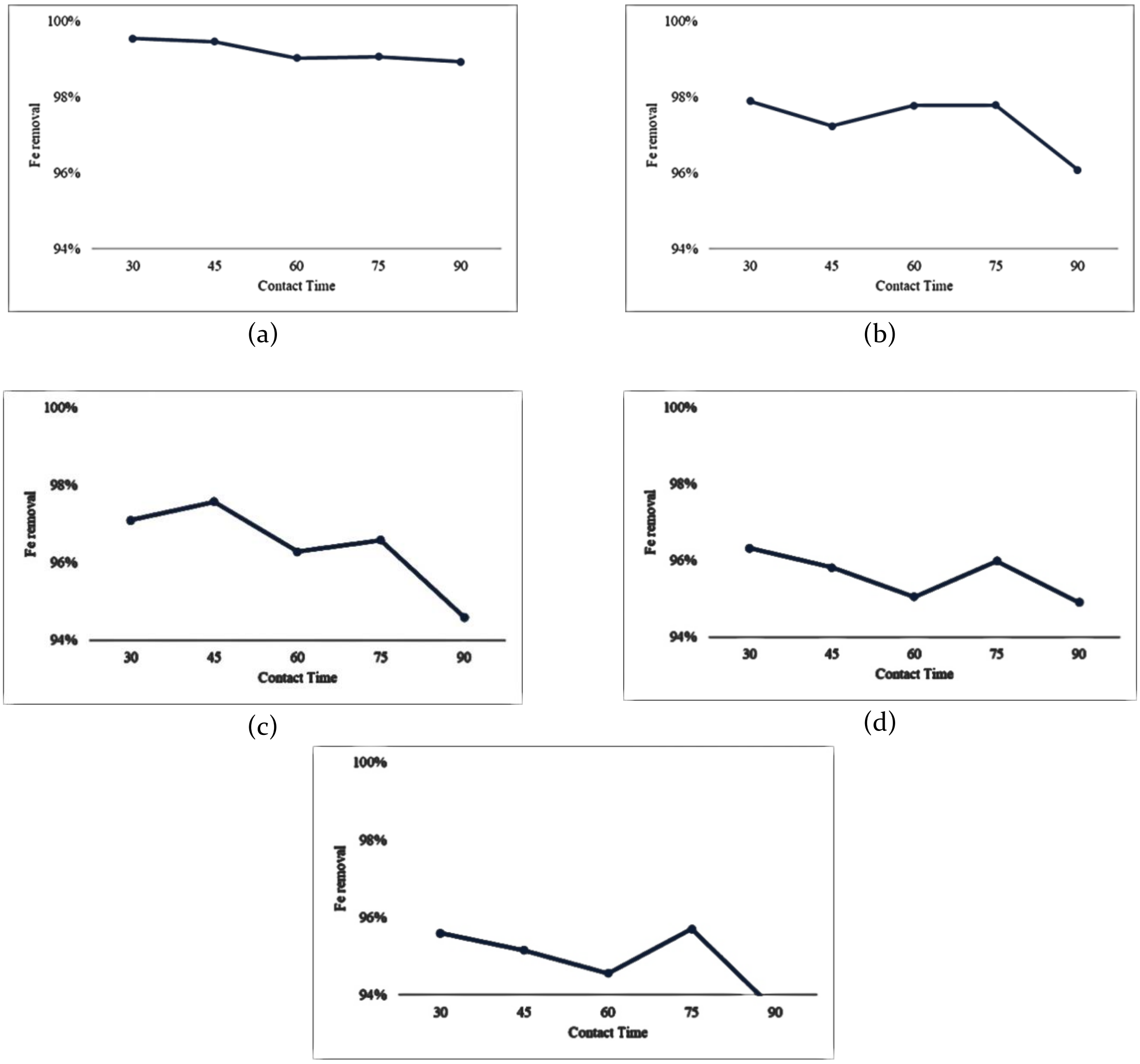


Figure 3. Presents the percentage of Fe removal with various contact time on 30; 45; 60; 75; 90 minutes with different dose of activated carbon (a) 5, (b) 10, (c) 15, (d) 20, (e) 25 g/L

3.4. ANOVA One-Way Test

This ANOVA test was carried out on the contact time and the adsorbent dosage variables. ANOVA One-Way test was used to analyze the data. The test used 5% of the significance level to measure the normality and homogeneity as presented in Table 2 and Table 3.

Table 2. The Normality test result according to the time function

	Time (minute)	Kolmogorov-Smirnov[a] Statistic	df	Sig.	Shapiro-Wilk Statistic	df	Sig.
Result	30	0.150	5	0.200*	0.972	5	0.885
	45	0.177	5	0.200*	0.961	5	0.818
	60	0.187	5	0.200*	0.946	5	0.708
	75	0.225	5	0.200*	0.916	5	0.502
	90	0.261	5	0.200*	0.876	5	0.292

	Time (minute)	Kolmogorov-Smirnov[a]			Shapiro-Wilk		
		Statistic	df	Sig.	Statistic	df	Sig.

Table 3. The Normality test result according to the dosage function

	Dose (g/L)	Kolmogorov-Smirnov[a]			Shapiro-Wilk		
		Statistic	df	Sig.	Statistic	df	Sig.
Result	5	0.294	5	0.183	0.858	5	0.220
	10	0.313	5	0.122	0.780	5	0.055
	15	0.251	5	0.200*	0.917	5	0.508
	20	0.257	5	0.200*	0.923	5	0.547
	25	0.207	5	0.200*	0.909	5	0.459

The results of the normality test used in the study were Shapiro-Wilk because the obtained data was less than 50 (Mohd Razali & Bee Wah, 2011). The data can be stated as normally distributed if the significance value is greater than the confidence level. In this normality test, the confidence level is 0.05 and the sig value in the normality calculation results is greater than 0.05, so the data is normally distributed.

Table 4. Homogeneity test results at time variations

		Levene Statistic	df1	df2	Sig.
Result	Based on Mean	0.309	4	20	0.868
	Based on Median	0.133	4	20	0.968
	Based on Median and with adjusted df	0.133	4	14.569	0.968
	Based on trimmed mean	0.288	4	20	0.882

Table 5. Homogeneity test results at various doses

		Levene Statistic	df1	df2	Sig.
Result	Based on Mean	1.089	4	20	0.388
	Based on Median	0.570	4	20	0.687
	Based on Median and with adjusted df	0.570	4	15.931	0.688
	Based on trimmed mean	1.016	4	20	0.423

The results of the homogeneity test are a prerequisite for ANOVA analysis because it is necessary to analyze that the variance of the population is the same (Usmadi, 2020). From the results of the calculation of the homogeneity test at the time variation it has a sig value greater than alpha 0.05 with a value of 0.868 so that the data has the same variance. The homogeneity test according to the dose variations has a Sig. value of 0.388, meaning the value is greater than the 0.05 confidence level. Therefore, the data was according to the same variance dose. From the test results in the Table 4 and Table 5, the data is homogeneous. The ANOVA One-Way test can be conducted when the data is proven to be normally distributed, and the result can be seen in Table 6.

Table 6. ANOVA One-Way Test based on the time

Time contact test	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	0.069	4	0.017	0.860	0.505
Within Groups	0.403	20	0.020		
Total	0.472	24			

Table 7. ANOVA One-Way Test based on the dosage

Dose test	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	0.381	4	0.095	20.961	0.000
Within Groups	0.091	20	0.005		
Total	0.472	24			

4. Discussion

The ANOVA test used in this study is the ANOVA One-Way in order to determine the effect of the variables having more than one variable. Table 6 and Table 7 show the results of the ANOVA One-Way test from variations in the contact time and the adsorbent dose, respectively. The influence of factors can be seen from the significance value which is greater or smaller than the confidence value of 0.05. If the significance value of the factor is greater than 0.05, the factor does not affect the experiment. In addition, when the significance value is smaller than the alpha value, then the factor influences the result (Sahliah et al., 2017). The significance value at contact time was 0.505 because the Sig. value was greater than 0.05 so that the contact time factor had no effect in this experiment. The significance value at the adsorbent dose is 0.00. Because the sig value is smaller than the confidence value of 0.05, the dose factor in this study has an effect. Thus, it can be concluded that the addition of the adsorbent doses did not significantly affect this study because the lowest dose of 5 gr/l was the most optimal dose for Fe content elimination. Then, the addition of contact time makes the removal effectiveness decrease

5. Conclusions

Based on the research that had been conducted, it can be concluded that the activated carbon produced from oil palm fronds is able to reduce iron (Fe) levels in groundwater. The optimal Fe removal efficiency is 99.53% with the optimum dose variation of 5 g/L and the contact time of 30 minutes. Variations of the contact time and the adsorbent dosage did not affect the effectiveness of Fe elimination because when the lowest variation used, the most optimum results were obtained.

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