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The Study of Improving Acid Mine Water Quality by Manganese Greensand and Activated Carbon Filtration

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Abstract
Bangka Belitung is one of the provinces in Indonesia known as a tin-producing area. Illegal tin mining results in excavated holes. The characteristics of the water in the ex-mining pits are acidic and contain heavy metals such as iron. This journal aims to determine the effect of filtration in improving the quality of water samples and determining the best doses of Na_2CO_3 and $KMnO_4$ as pre-treatment. Also, to find out which experiment resulted in higher Fe removal efficiency and whether the quality of the treated water complies with the Regulation of the Minister of Health Number 492 of 2010. This study used primary data of pH and iron content as dependent variables. The population of this research is Alang Lake water taken by random sampling method. The experiment was conducted on a lab scale with two independent variables, sampled with replicates every 30 minutes. In this experiment, the optimum doses of Na_2CO_3 and $KMnO_4$ were 300 mg/L and 0.75 mg/L, respectively. The treated water has a pH value of 7.4-8.1, iron concentration <0.25 mg/L with a removal efficiency of 93.56%, and turbidity of 0.05-0.75 NTU. Thus, it can be said the dependent variables are following the relevant regulations.
Keywords: *activated carbon, Fe, filtration, manganese greensand, water quality*

Abstrak
Bangka Belitung merupakan salah satu provinsi di Indonesia yang dikenal sebagai daerah penghasil timah. Aktivitas penambangan timah yang ilegal menghasilkan lubang galian. Karakteristik air di bekas lubang tambang bersifat asam dan mengandung logam berat seperti besi. Riset ini bertujuan untuk mengetahui pengaruh filtrasi dalam meningkatkan kualitas sampel air serta untuk mengetahui dosis Na_2CO_3 dan $KMnO_4$ yang terbaik sebagai *pre-treatment*. Juga untuk mengetahui eksperimen mana yang menghasilkan efisiensi penyisihan Fe yang lebih tinggi dan apakah kualitas air olahan sesuai dengan Peraturan Menteri Kesehatan Nomor 492 Tahun 2010. Penelitian ini menggunakan data primer pH dan kandungan besi sebagai variabel terikat. Populasi penelitian ini adalah air Danau Alang yang diambil dengan metode *random sampling*. Percobaan dilakukan pada skala lab dengan dua variabel bebas yang diambil sampelnya dengan ulangan setiap 30 menit. Pada percobaan ini dosis optimum Na_2CO_3 dan $KMnO_4$ masing-masing adalah 300 mg/L dan 0,75 mg/L. Pada air olahan nilai pH 7,4-8,1, kandungan besi <0,25 mg/L dengan efisiensi penurunan kandungan besi sebesar 93,56%, dan kekeruhan 0,05-0,75 NTU. Dengan begitu dapat dikatakan bahwa variabel terikat pada air olahan sudah sesuai dengan peraturan terkait.
Kata Kunci: *Fe, filtrasi, karbon aktif, kualitas air, manganese greensand*

1. Introduction

Water is one of the primary human needs so that the body can metabolize adequately. The human body requires 2-2.5 L of water to carry out daily activities in a day [1]. Earth has water availability as much as 71% of the earth's total area, but 97% of it is seawater; the remaining 3% is used as a water source for humans to carry out their lives [2]. Today, the annual need for clean water increases with population growth. Therefore, one-third of the world's population suffers from water scarcity. The increasing population also grows various human activities that harm the environment, such as mining [3].

As one of the mining areas in Indonesia, Bangka Belitung has many holes excavated from tin mining activities [4]. Illegal ex-pits tin mining becomes a source of water and the water is mixed with metal scraps from tin mining, thus affecting water quality [5]. The water stored in the hole is not suitable to be used as a water source for humans if proper and organized handling has not been carried out to measure the characterization of the water contained in the hole [3]. Water inspection has separate parts as physical, chemical, and biological.

Drinking water must meet physical, chemical, and biological needs. The requirements for clean water are clear, odorless, tasteless, average temperature, and do not contain solids [6]. The chemical requirements

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for water are neutral pH, contain no toxic chemical compounds, and have no salts or metal particles such as iron (Fe) [6]. However, the body needs iron content; below 0.3 mg/L, according to Minister of Health Regulation Number 492 of 2010. If the Fe concentration exceeds the quality standard, it will cause problems, for example, causing odor, yellow color on clothes, blockage in pipes, damage to the digestive tract, and death [7]. Moreover, organic content such as *E. coli* microorganisms should not be in drinking water.

Water containing iron in appearance is clear and shiny. However, with exposure to air, the water becomes cloudy due to the oxidation of iron to the Fe^{3+} state, which accelerates the colloidal structure. The oxidation rate is not fast, and therefore, the degraded structure can persist for a long time in the air circulating through the water [8].

Iron (Fe) is one of the essential components of surface water and groundwater. Iron (Fe) is one of the microelements needed by the body; Iron (Fe) plays a vital role in the body's metabolic cycle. Hazards for health from heavy metal Fe only arise if Fe content >1 mg/l, damaging the digestive tract, digestive disorders, nausea, and motion sickness in humans [9]. Water containing iron (Fe) is not entirely in demand by families because it can cause yellow stains on clothes, porcelain, and rust on various other equipment and reduce the desire to drink water.

Based on the results of lab tests in May 2021, Alang Lake's raw water has several parameters that do not comply with the Regulation of the Minister of Health Number 492 of 2010. These parameters are pH (2.54) and Iron (3.63 mg/L). This paper aims to find a solution; thus, the quality of the treated water complies with the relevant guidelines. The proposed treatment uses filtration with the medium of silica sand, manganese greensand, activated carbon, and a pre-treatment of addition Na_2CO_3 and $KMnO_4$ to significantly improve water quality.

2. Material and Methods

This research uses both primary and secondary data. The secondary data was reviewed to determine the treatment that will be used in this research. The primary data takes through the experimental method by performing laboratory experiments. This experiment conduct at laboratory scale in the Laboratory of Perumda Tirta Sejiran Setason, Muntok, Bangka Barat, Bangka Belitung. The experiment was conducted in August 2021.

The water sample was collected from Alang Lake which located in Tempilang, Bangka Barat, Bangka Belitung. The water sample took 80 Litres on August 12th, 2021, using a random sampling method. The water sample was taken at (2°06’36” S 105°39’11” E).

Materials and Tools

The tools needed for this experiment were 1 L beaker glass, 100 mL beaker glass, 1 mL volumetric pipette, 100 mL measuring glass for chemical addition—a basin with a capacity of 20 L, agitator, and drill tool for mixing the water sample with chemicals. The water intake to flow the water sample into the filtration unit uses a gallon with a capacity of 12 L. The filtration unit in this experiment uses an acrylic block with the dimension of 25 cm x 10 cm x 10 cm with a total volume of 2.5 L.

Figure 1. Batch reactors for experiments
Source: Research documentation (2021)

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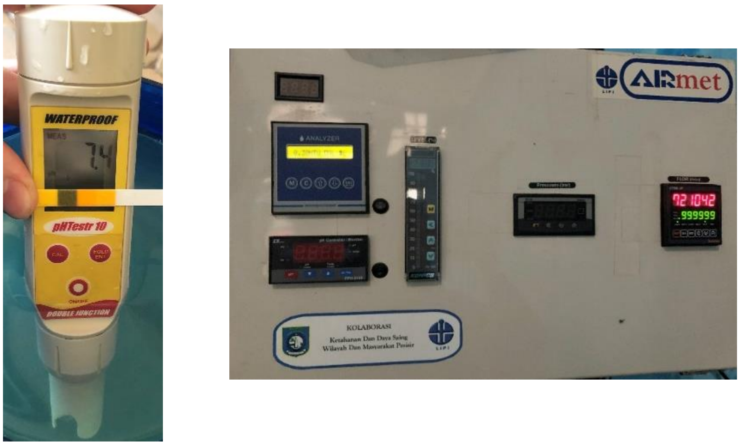

(a) (b)

Figure 2. (a) pH Meter (b) Turbidity Meter
Source: Research documentation (2021)

Experimental Methods

Determining Optimum Dosage of Na_2CO_3 and $KMnO_4$

In preparing a water sample before it goes to the filtration process, it needs to adjust the pH of the water sample into neutral to base condition, in this research uses Na_2CO_3 as water neutralization, determining the optimum dosage Na_2CO_3 it is running by jar test. The pH of raw water is 2.4; thus, it needs to be neutralized to comply with Regulation of Ministry of Health Number 492 of 2010 that stated pH must be in the value 6.5-8.5. Moreover, the filtration medium in this experiment using manganese greensand require the pH of the water samples in the range of 7.5-9.0 to achieve the maximum performance.[10]

Following preparation is to oxidize dissolved Fe in the water sample by $KMnO_4$ because the water samples contain 3.885 mg/L of Fe^{2+}. Determining the optimum dosage of $KMnO_4$ is by running the jar test. The changing color of raw water indicates the optimum dosage from clear transparent into yellow. Yellow indicates that Fe^{2+} has already oxidized, becoming Fe^{3+}. If the water becomes light purple, excess $KMnO_4$ was added.[11]

Steps of the Pre-Treatment of The Water Sample by Adding Na_2CO_3:

Preparation of making 2% Na_2CO_3 solution:

1. Prepare 1000 mL beaker glass, fill it with the water sample.
2. Add 20 grams of Na_2CO_3 to the beaker glass.
3. Mix it by static mixer until the Na_2CO_3 is dissolved in the water.

Jar Test in Determining Optimum Dosage of Na_2CO_3

1. Prepare five beaker glasses containing the water sample with a volume of 1 L.
2. Add various dosages to each beaker glass, as shown in **Table 1.**

Table 1. The Dosages of natrium carbonate in water samples

	The Dosages
August 12th, 2021	Na_2CO_3 (2%)
Sample 0	0 mL
Sample 1	10 mL
Sample 2	15 mL
Sample 3	20 mL
Sample 4	25 L

Sources: Research documentation (2021)

3. Mix the water sample with 2% solution of Na_2CO_3 120 rpm for 5 minutes then continue for the next 20 minutes with 20 rpm. After that wait for 15 minutes, then check the pH value of the water sample on each beaker glass (SNI 19-6449-2000).

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Steps of the Pre-Treatment of The Water Sample by Adding $KMnO_4$:

Preparation of making 1% $KMnO_4$ solution:

1. Prepare 100 mL beaker glass, fill it with the water sample.
2. Add 1 gram of $KMnO_4$ to the beaker glass.
3. Mix it by static mixer until the $KMnO_4$ is dissolved in the water.

Jar Test in Determining Optimum Dosage of $KMnO_4$:

1. Prepare four beaker glasses containing the water sample with a volume of 1 L.
2. Add various dosages to each beaker glass, as shown in **Table 2**:

Table 2. The Dosages of potassium permanganate in water samples

	Replicate	
August 12th, 2021	$KMnO_4$ (1%)	$KMnO_4$ (1%)
Sample 0	0 mL	0 mL
Sample 1	0.050 mL	0.050 mL
Sample 2	0.075 mL	0.075 mL
Sample 3	0.100 mL	0.100 mL

Sources: Research documentation (2021)

3. Mix the water sample with 1% solution of $KMnO_4$ 100 rpm for a minute then continue for the next 30 minutes by 20 rpm. After that wait for 15 minutes, then check the color of the water sample on each beaker glass [12].

Preparation of Filtration Unit

Three filter mediums that were used in this experiment are silica sand, manganese greensand, and activated carbon. In experiment 1 the filter medium only contains silica sand and manganese greensand. The second experiment use all three filter mediums.

Steps of Preparation of Filtration Unit as follows:

1. Set up Materials and Tools
2. Prepare the filtration unit
3. Fill both the filtration unit with silica sand at the height of 5 cm, then continue by adding 8.4 cm of manganese greensand. (Variable A)
4. Fill both the filtration unit with silica sand at the height of 5 cm, then continue by adding 4.6 cm manganese greensand, and add activated carbon 3.8 cm. (Variable B)
5. Run the water into the filter until the effluent is clear.

The picture shows the position of the gallon and the filtration unit:

Figure 3. Position of Gallon and Filtration Basin
Sources: Research documentation (2021)

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Analysis Method & pH Measurement

The parameters measured in this experiment are pH, turbidity, and Fe. The pH was measured using the pH testr10 digital pH meter. The pH value is a common and important parameter in determining the quality of water. It is also known as the hydronium ion index scale. pH measures the concentration of hydronium ion concentration in a solution. A neutral solution has a pH of 7 and a concentration of 10-7 mole/L of hydroxide ions. To know the pH levels in the water is necessary to measure the pH value of the water. pH is a parameter that measures the acidity or basicity of a solution [13]. A pH value less than 7 indicates an acid level; on the other hand, greater than 7 indicates no acid content. Water, an ideal drink consumed, has a neutral level, pH 7.[14]

Device Calibration:

1. Prepare buffer solution that indicates the pH 4
2. Clean the electrode of the pH meter with distilled water and put it in the buffer solution until the value of pH 4 appears.

Measurement:

1. Remove protective cap and clean electrode with distilled water.
2. Turn on the device and immerse the pH meter until the immersion level.
3. Stir gently and wait until the reading is stabilized.
4. Switch off the device, clean the electrode with distilled water and replace the protective cap.

Turbidity Measurement

In testing the turbidity value in water using a method based on SNI-06-6989-25 2009.

Device Calibration:

1. Turn on the turbidity meter
2. Calibrate the instrument using a turbidity standard solution (800 NTU, 100 NTU, 20 NTU, and 0.02 NTU)
3. Immerse the turbidity meter with a standard solution.

Measurement:

1. Put the water sample in a beaker glass
2. Put the turbidity meter on beaker glass
3. Wait for a couple's minutes until the value that appears on the screen is already stable.

Fe Measurement & Removal Efficiencies

In measuring the Fe value, using three parties' (MEDIALAB) services. Fe was analyzed by method SM 23rd 3120B – 2017. The removal of Fe throughout observation can be calculated as the removal efficiency. The formula for removal efficiency is shown below [15].

$$R = \frac{I - F}{I} \; x \; 100\%$$

Where:
R = Removal efficiency (%),
I = Initial concentration of the parameter
F = Final concentration of parameter

3. Results and Discussion

Filter Design

In this experiment, filtration is the primary process to decrease the concentration of Fe. Filtration in this experiment uses three filter mediums: silica sand, manganese greensand, and activated carbon. The filtration basin is acrylic in a block form with a length x width x height (10 x 10 x 25 cm).

Table 3. The filtration unit design

Parameters:		
Flow rate	0.08	$m^3/(m^2.min)$
	80	$dm^3/(m^2.min)$
	0.8	$dm^3/dm^2.min$
	0.008	$liter/(cm^2.min)$

Parameters:		
Depth of greensand	460	Mm
	46	Cm
Depth of anthracite	380	Mm
	38	Cm
Lab-scale 10%		
Unit Flow rate specification	0.0008	liter/(cm^2.min)
	0.8	cm^3/(cm^2.min)
Depth of greensand	4.6	Cm
Depth of anthracite	3.8	Cm
Length of square	10	Cm
Area	100	cm^2
Flow rate	80	cm^3/min
	0.08	liter/min
	4.8	liter/hour
Silica Sand	5	Cm

Sources: Research data (2021)

Figure 4. Filtration basin
Sources: Research data (2021)

Water Sample Preparation

The water sample in this research was taken from Alang Lake on August 12th, 2021. Alang Lake is an ex-pit tin mining. The initial pH value and Fe concentration of the water in the Alang Lake do not comply with Regulation of Ministry of Health Number 492 of 2010. Even though the appearance of the water sample is clear, the pH and the Fe concentration of both experiments are 2.4 and 3.885 mg/L, respectively. Regarding the regulation of the standard for drinking water in Indonesia, the pH value must be 6.5-8.5, and the maximum concentration of Fe is 0.3 mg/L.

In improving the water quality by parameters pH and Fe concentration, it needs to increase the pH value by adding Na_2CO_3 and decreasing the Fe concentration with oxidation by $KMnO_4$ and filtration by silica sand, manganese greensand, and activated carbon. Before the filtration, the water samples are pre-treatment first by adding Na_2CO_3 and $KMnO_4$ to the water. It needs to know the optimum dosage for the water sample. Determining the optimum dosage was obtained by jar test.

The indicator in determining the optimum dosage of Na_2CO_3 by jar test is the pH value of the water

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samples is already in the range of 6.5-8.5 according to Regulation of Ministry of Health Number 492 of 2010. Also, due to the usage of manganese greensand as filter media filtration process that requires pH value of water samples must be in 7.5-9.0. **Table 4** shows the result of the addition of various dosages of Na_2CO_3. It shows that the optimum dosage of Na_2CO_3 that complies with the pH value requirement is 15 mL of 2% solution.

Referring to [16], the pH value of the water sample increased from 3.5 to 6.67 after the addition of 150 mg/L of Na_2CO_3. In this research, the water samples need higher dosages of Na_2CO_3 to achieve the optimum pH to run the experiment.

Table 4. The optimum dosage of natrium carbonate for water neutralization

	Optimum Dosage	Replicate	
August 12th, 2021	Na_2CO_3 (2%)	pH (1)	pH (2)
Sample 0	0 mL	2.4	2.4
Sample 1	10 mL	6.2	6.4
Sample 2	15 mL	7.4	7.9
Sample 3	20 mL	8.2	8.5
Sample 4	25 mL	8.5	9

Sources: Research data (2021)

In determining the optimum dosage of $KMnO_4$, a jar test was performed with various dosages to get the color changes of water sample from transparent become yellow that indicate the $KMnO_4$ already oxidize the Fe^{2+} become Fe^{3+}. The water sample is purple if the dosage of $KMnO_4$ is exceeded. In stoichiometry, the dosage of $KMnO_4$ is higher than in the experiment. The usage of $KMnO_4$ dosage is less in the experiment due to the formation of excess MnO_2, which can function as an oxidizing agent. **Table 5** shows color changes of the water sample from transparent become yellow by adding 0.075 mL of 1% $KMnO_4$ solution.

Table 5. The optimum dosage of potassium permanganate in oxidizing Ferrous Ion

	Replicate		
August 12th, 2021	$KMnO_4$ (1%)	$KMnO_4$ (1%)	Color Indicator
Sample 0	0 mL	0 mL	Clear Transparent
Sample 1	0.050 mL	0.050 mL	Light yellow
Sample 2	0.075 mL	0.075 mL	Yellow
Sample 3	0.100 mL	0.100 mL	Purple

Sources: Research data (2021)

Experimental Result

In the first and second experiments, the pH was checked six times, the raw water, effluent at 0, 30, 60, 90, and 120 minutes. The first experiment uses silica sand and manganese greensand as a media filter. The second experiment uses silica sand, manganese greensand combined with activated carbon as a media filter. The result of the pH during this study are shown in **Figure 5** The pH of the raw water was 2.4 that categorical as acid. Thus, it needs to be added by alkaline solution. After adding Na_2CO_3, the pH of all samples complies with Regulation of Ministry of Health Number 492 of 2010.

The measurement of iron content in the water samples was carried out in the raw water and the treated water at 30, 60, 90, and 120 minutes. The decreasing of Fe concentration is shown in Figure 6. The initial concentration of Fe in raw water is 3.885 mg/L, respectively. The treated water samples for both experiments had Fe concentration <0.25 mg/L, which means already safe to be consumed; regarding Regulation of Ministry of Health Number 492 of 2010, the maximum concentration of Fe is 0.3 mg/L.

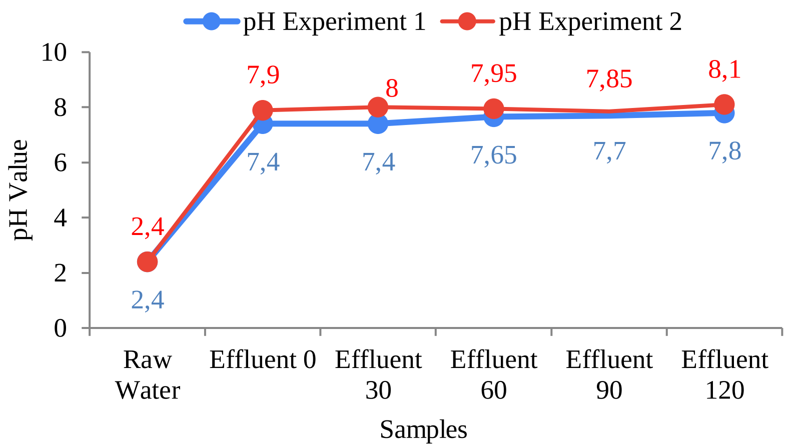

Figure 5. The pH performance of raw and treated water
Sources: Research data (2021)

The removal efficiencies of Fe concentration in water samples are:
Removal Efficiencies of Experiment 1:

$$R1 = \frac{(3.885 - 0.25)}{3.885} x\ 100\%$$

$$R1 = 93.56\%$$

Removal Efficiencies of Experiment 2:

$$R2 = \frac{(3.885 - 0.25)}{3.885} x\ 100\%$$

$$R2 = 93.56\%$$

Refers to the experiment of [17], it gives the removal efficiencies of Fe concentration by filtration with silica sand and manganese greensand combined with activated carbon is 78.36%. Thus, this research achieves higher removal efficiencies as shown in **Figure 7**.

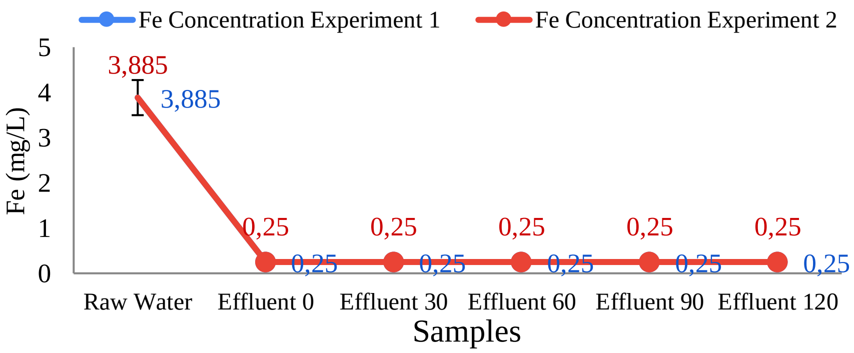

Figure 6. The Fe concentration of raw and treated water
Sources: Research data (2021)

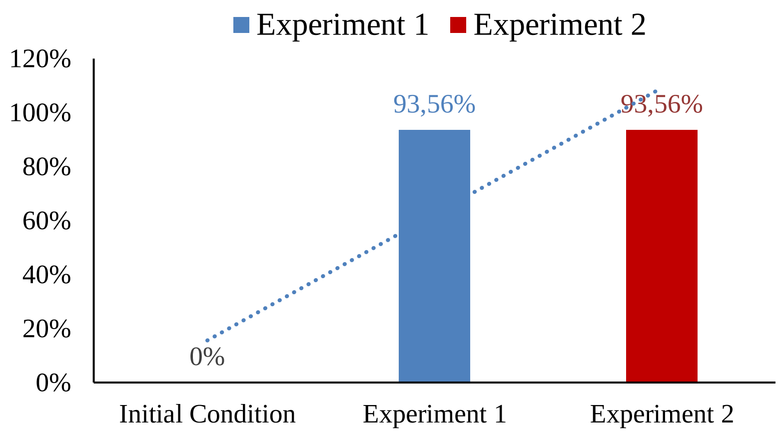

Figure 7. The Removal efficiency of Fe concentration in the treated water
Sources: Research data (2021)

The last measurement is turbidity; this measurement aims to ensure that the process during this experiment does not make turbidity of the water exceed the standards. The result of the turbidity check is shown in **Figure 8**. The initial turbidity of experiments 1 and 2 are 0.94 and 0.96 NTU, respectively. After the treatment, the turbidity of treated water from experiment 1 is around 0.07-0.77 NTU. The second experiment resulted in a turbidity value of 0.055-0.665 NTU.

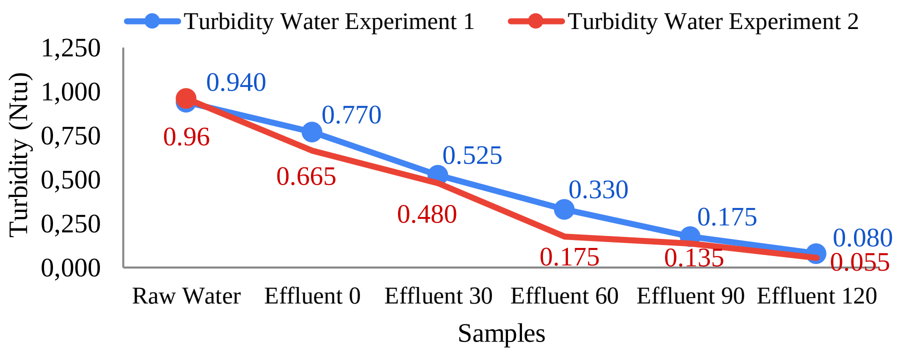

Figure 8. The turbidity value of raw and treated water
Sources: Research data (2021)

4. Conclusion

The water of the acid mine is utilized by the resident as one of their water sources. Unfortunately, the characteristic of raw water does not comply with the Regulation of Ministry of Health Number 492 of 2010. Thus, it needs proper treatment before it is consumed. This research proposed treatment of filtration by silica sand, manganese greensand, and activated carbon to reduce the Fe concentration of the water sample. Pre-treatment is needed before the filtration process. Pre-treatment in this experiment consists of water neutralization using the addition of Na_2CO_3 and water oxidation by $KMnO_4$. The result of this research are the optimum dosage of Na_2CO_3 and $KMnO_4$ is 300 mg/L and 0.75 mg/L, respectively. The filtration process significantly improves the treated water quality of pH and Fe concentration compared to raw water quality. The removal efficiencies of Fe concentration in experiments 1 and 2 resulted in the same value, which is 93.56%. The pH value and Fe concentration of the treated water from both experiments comply with the Regulation of Ministry of Health Number 492 of 2010.

5. Acknowledgment

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