HELIUM: Journal of Science and Applied Chemistry
https://journal.unpak.ac.id/index.php/he_jsac
Volume 02, Number 01, June 2022, Page 13 - 19
e-ISSN: 2776-4508; p-ISSN: 2776-1711
Publisher : Universitas Pakuan

Development of A Hybrid Method by Integrating Electrocoagulation Countinous System with Activated Charcoal Adsorption to Treat Sago Wastewater

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Article history: *received 10 April 2022; revised 24 May 2022; accepted 29 June 2022; published 30 June 2022*

Abstrct

Wastewater from the sago industry can pose a risk of pollution because it contains chemicals such as high organic matter content, fiber, 4% unextracted starch, nitrogen solution, and cyanoglucoside which are difficult to degrade in the environment. Sago wastewater is generally acidic, has foul smells, and has a high solids concentration. In this study, a hybrid method using electrocoagulation continuous system and activated carbon adsorption was developed to decrease organic and inorganic contaminates which considered importand environmental concern. These two methods have the advantages of simple equipment, easy operation, low efficiency, and short reaction time. The flow rate will affect the pause time of the sample in the reactor. The longer the pause time, feeding will lead to an increase in the level of turbidity that can be lowered. Industrial wastewater variations in the initial analysis then carried out the electrocoagulation process in a continuous system with a container capacity of 1680 mL with an electrode distance of 2 cm, a voltage of 18 volts, and wastewater into a container with a flow rate of 1 mL/, 2 mL/second, 3 mL/ sec, 4 mL/sec, and 5 mL/sec. In addition, the waste is directly into a container containing charcoal. The parameters studied were COD, BOD, TSS, TDS, KMnO4, DO, pH, DHL, and Al concentration analysis with Atomic Absorption Spectrophotometer, odor, and color analysis. The maximum removal efficiency for TDS, TSS. COD, BOD, TDS, $KMnO_4$ were 81.16%, 87.58%, 91.32%, 48.66%, 99.07%, for a flow rate of 1 mL/second, also other parameters also met have met the quality standards set by PerMenLH No. 5.

Keywords: Adsorption; Sago Waste; Electrocoagulation.

1. INTRODUCTION

In recent years, industrial development in Indonesia has increased, one of which is the sago industry. This can have both positive and negative impacts on society. The positive impact that occurs from the increase in this industry is increasing the economy and state tax revenues and open new vacancy. Meanwhile the negative impact arising from the existence of this sago industry is to produce of waste that can increase the risk of pollution and damage to the environment. This is because the increase in the amount of sago production is directly proportional to the amount of waste produced. Based on Indonesian plantation statistics for 2016-2018 and referring to the 2018 national sago production projection of 281,898 tons, nationally the total sago liquid waste produced is 7,800,000 m^3. The waste still contains high organic matter, fiber, 4% unextracted starch, nitrogenous solution, and cyanoglucoside [1]. Sago liquid waste is generally acidic, has foul smells, and has a high solids concentration. The smell is an indication of putrefaction in wastewater. The odor in wastewater is caused by volatile materials, dissolved gases, and by-products of decaying organic matter [2].

In general, sago wastewater treatment is carried out microbiologically by adding bacteria to the liquid waste, but this process requires a longer time [3][4]. electrocoagulation uses the principle of electrolysis with electrodes made of aluminum or iron which produces coagulant compounds to precipitate various pollutants in wastewater [5][6]. While adsorption is the process of clumping dissolved substances in solution by the surface of the adsorbent which makes the material enter and collects in an absorbent substance. Based on the results of research by Sutanto, et al (2018), showed that this electrocoagulation method could reduce TSS levels from 410 mg/L to 44.4 mg/L (89.17% efficiency) and COD levels decreased from 1335.05 mg/L to 557 mg/L (58.28% efficiency) at 18 volts and 60 minutes [7]. Furthermore, in 2020 the research was developed again using the electrocoagulation method and reducing H_2S (odor) levels using UV light. As a result, COD levels decreased from 891.67 mg/L to 462.67 mg/L (48.11% efficiency) and decreased TSS levels from 505.5 mg/L to 12.25 mg/L (97.58% efficiency). at a voltage of 30 volts and a time of 100 minutes. H2S levels dropped from 0.02 mg/L to <0.001 mg/L. UV rays have been effective in reducing H_2S levels but have not been able to eliminate odors in sago waste. In both studies, it has not been able to reduce COD levels to below the quality standard required by PerMenLH No. 5 of 2014 concerning waste quality standards for the tapioca industry and/or activities/businesses, where the TSS and COD values of waste are 100 mg/L and 300 mg/L, respectively [8].

Therefore, this study aims to determine the optimum flow time for the success of sago industrial wastewater treatment by electrocoagulation with a continuous system and deodorizing using activated charcoal. The flow rate will affect the pause time of the sample in the reactor. The longer the pause time, the greater the level of turbidity that can be lowered.

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2. METHODS

The equipment used in this study was a reactor of electrocoagulation devices, a filter with a pore size of 0.45 m, a heater, a stopwatch, a DC Power Supply, a desiccator, a pH meter Lutron PH-207HA, DO meter Lutron DO -5509, Lutron YK-22CTA conductometer, Atomic Absorption Spectrophotometer (SSA) Agilent 240 FS, and Gas Chromatography (GC) Thermo Fisher Scientific. While the materials used in this research are activated charcoal, organic free water, digestion solution ($K_2Cr_2O_7$) 0.1 N, sulfamic acid (NH_2SO_3H), potassium hydrogen phthalate/KHP ($C_8H_5KO_4$) COD 500 mg O_2/L raw solution, acid sulfate (H_2SO_4) 8 N free of organic substances, sodium oxalate ($(COONa)_2.2H_2O$), potassium permanganate ($KMnO_4$) 0.01 N, oxalic acid ($(COOH)_2.2H_2O$) 0.01 N, hydrochloric acid (HCl) concentrated water, mineral-free water, and sago industrial wastewater.

2.1. Sago Liquid Waste Treatment

Sago wastewater treatment uses a continuous system electrocoagulation method which is a method of coagulation and deposition of fine particles in wastewater using direct electric current to a pair of electrodes (two sheets of Al plate) 2 cm apart with variations in flow rates 1, 2, 3, 4 and 5 mL/sec and an electrolysis voltage of 18 volts. Then proceed with the adsorption process using activated charcoal. The reactor of equipment used for processing sago liquid waste can be seen in Figure 1.

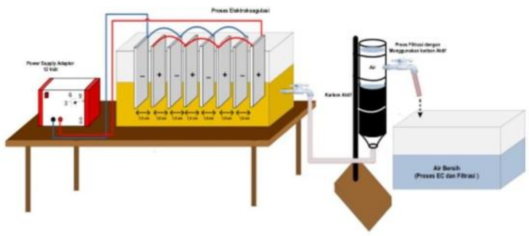

Figure 1. Reactor Model of Continuous System Electrocoagulation Unit and Activated Charcoal Adsorption

2.2. Analysis of Sago Liquid Waste

Parameters analyzed before and after processing include BOD, COD, TSS, DO, pH, DHL, TDS, permanganate number, Al content as well as odor and color analysis. Furthermore, the quality change data for each parameter is plotted against the flow rate to find the optimum flow rate. COD measurements were carried out titrimetrically using the closed reflux method, TSS measurements were carried out using the gravimetric method, DO used the electrochemical method, pH measurements were using the potentiometric method, DHL and TDS were using the conductometer method, the permanganate number was measured using the titrimetric method, odor and color analysis was carried out by organoeptic and Al levels were measured using Atomic Absorption Spectrophotometry (AAS).

3. RESULT AND DISCUSSION

Characteristics of sago processing wastewater and compared with wastewater quality standards according to PerMenLH No. 5, concerning waste quality standards for tapioca industry and/or activities/businesses, are shown in Table 1.

Tabel 1. Sago Wastewater Characteristics Before Processing

Testing Parameter	SNI Methods	Unit	Standard	Result
BOD_5	06 6989.73-2004	mg/L	150	297,00
COD	06 6989.73-2004	mg/L	300	948,64
TSS	06 6989.3-2004	mg/L	100	481
pH	06 6989.11-2004	-	6,0-9,0	4,52
DO	06 6989.73-2004	mg/L	-	1,35
DHL	06 6989.1-2004	µS	-	728,5
$KMnO_4$	06 6989.22-2004	mg/L	-	385,68
TDS	06 6989.3-2004	mg/L	-	617

According to Table 1, shows that the standardized test parameters which include the degree of acidity (pH), total dissolved solids (TSS), chemical oxygen demand (COD) and biological oxygen demand (BOD) do not meet the quality standards. The very low dissolved oxygen (DO) parameter of 1.35 mg/L was caused by the high content of organic pollutants in the wastewater which was represented by the COD value. The high value of COD is due to the processing of liquid waste sago contains a lot of organic substances as a by-product of the sago extraction process. The high value of TSS is due to the organic matter being insoluble and forming a suspension. The TSS and COD values of palm flour industrial wastewater can reach 720 mg/L and 4231 mg/L. This difference can be caused by the amount of sago waste produced differently [9].

3.1. Study of Flowrate to Increase in DO

Dissolved oxygen (DO) although not required by wastewater quality standards, it is very important to know related to the number of organic pollutants in wastewater. The more the number of organic pollutants, the less dissolved oxygen because oxygen is used by microbes to degrade waste. Clear water can contain dissolved oxygen up to 8 mg/L depending on water temperature [10]. Unpolluted public waters contain DO > 5 mg/L. The sago wastewater before the electrocoagulation process had a low DO value of 1.35 mg/L. A low DO value indicates that the water quality is poor.

The process of electrocoagulation and activated charcoal adsorption is effective to increase the DO value. In Figure 1, the DO value increased at each flow rate. The smaller the applied flow rate, the the longer the contact time.

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At a flow rate of 5 mL/second (dwelling time of 5.6 minutes) it can increase the DO value to 1.75 mg/L. At a flow rate of 1 mL/second (28 minutes pause time) it can increase the DO value to 5.2 mg/L. So that the highest increase in DO occurs at the application of a flow rate of 1 mL/s.

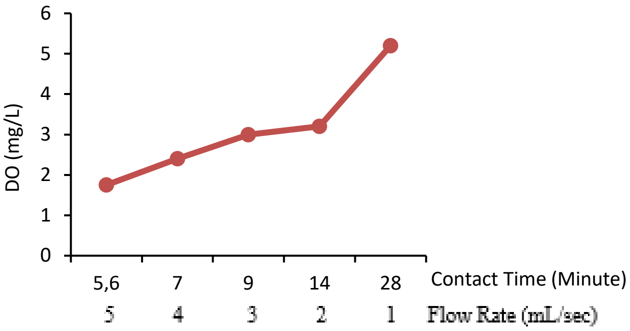

Figure 2. Increasing of DO Wastewater Level

Dissolved oxygen is needed by all living organisms for respiration, metabolic processes or the exchange of substances which then produce energy for growth and reproduction. In addition, oxygen is also needed for the oxidation of organic and inorganic materials in aerobic processes.

3.2. Study of Flowrate to Increase in pH

During the electrocoagulation process, an oxidation reaction will occur at the anode of the metal making up the electrode, in this study aluminum is oxidized.

$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3\ e-$

$2H_2O_{(l)} + 2e^- \rightarrow H_{2\ (g)}\uparrow + 2OH^-_{(aq)}$

Figure 3 shows the relationship between increasing pH and the applied flow rate, the smaller the applied flow rate, the longer the contact time, so the increase in pH is also greater. At a flow rate of 5 mL/second (the pause time of the waste in the reactor is 5.6 minutes) it can increase the pH to 6.08. A flow rate of 1 mL/second (28 minutes pause time) can increase the pH to 7.73. These results have met the quality standards of PerMenLH No. 5 2014, where the highest increase in pH was obtained at the application of a flow rate of 1 mL/s. The small flow rate is proportional to the longer the contact time which will produce more and more OH-. The amount of OH- produced will increase the pH. The pH of the solution slightly increased during the electrocoagulation process due to the formation of OH- and H_2 gas [11].

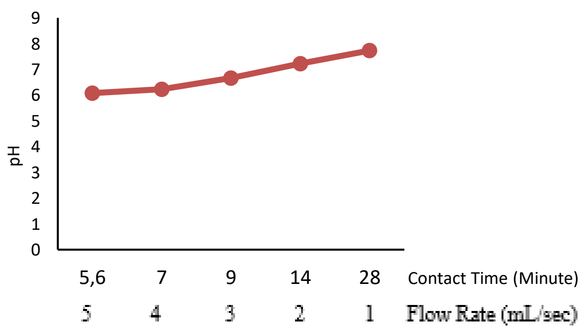

Figure 3. Increasing of pH Sago Wastewater

3.3. Study of Flowrate to Deacrease in DHL

Electrical conductivity or conductivity in water is a value that states the ability of a liquid solution to conduct an electric current. The electrical conductivity of water depends on the concentration of ions and water temperature, therefore the increase in dissolved solids will affect the increase in electrical conductivity [12].

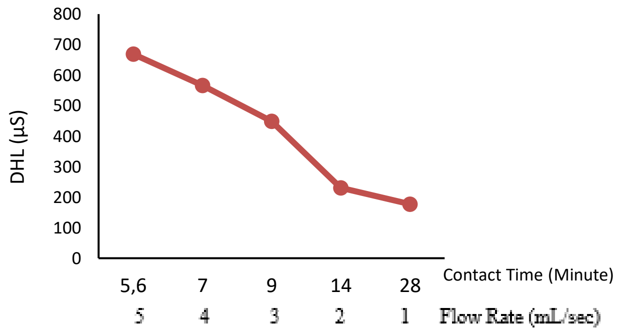

Figure 4. Decreasing of DHL Level

Figure 4. shows the relationship between the decrease in DHL and the applied flow rate, the smaller the flow rate applied, the longer the contact time, so the decrease in DHL is also greater. At a flow rate of 5 mL/sec (dwelling time of 5.6 minutes) it can reduce the DHL to 669 S. At a flow rate of 1 mL/second (28 minutes pause time) it can reduce the DHL to 177.25 S. DHL that continues to decrease can be caused by most of the flocculated ions so that the electrical conductivity ions in the solution are decreasing. The decrease in electrical conductivity occurs because when measuring the value of electrical conductivity there are still flocculants in the form of clumps of water impurities which are insulating materials, thereby reducing the ability of water to conduct electric current [13].

3.4. Study of Flowrate to Increase Al Concentration

Dissolution of aluminum occurs at the anode electrode. In the aluminum dissolution process, different chemical reactions occur on the surface of the two electrodes. The diode process results in the dissolution of aluminum metal into Al^{3+}.

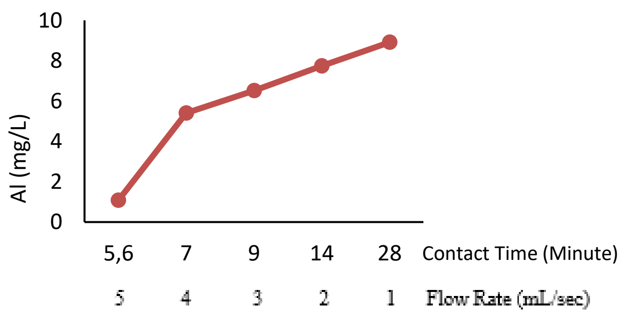

Figure 5. Increasing of Al Sago Wastewater Level

Figure 5 shows the relationship between the increase in Al content and the applied flow rate, the smaller the applied flow rate, the longer the contact time, and the greater the increase in Al content. At a flow rate of 5 mL/second (dwelling time of 5.6 minutes) the Al content was 1.08 mg/L. At a flow rate of 1 mL/second (dwelling time 28 minutes) the Al content was 8.91 mg/L. The results of the largest increase in Al levels occurred at the application of a flow rate of 1 mL/second. Although the levels of aluminum are not listed in the Regulation of the Minister of the Environment of the Republic of Indonesia Number 5 of 2014 Appendix V, the measurement of dissolved aluminum levels is still carried out to find out how much aluminum is released in the anode rod.

3.5. Study of Flowrate to Decrease Total Suspended Solid (TSS) Level

Suspended solids affect the turbidity and color of the water. If there is deposition and decomposition of these substances in the receiving water body, the wastewater will reduce the use-value of these waters because it reduces water quality due to reduced penetration of the sun into the water body, water turbidity increases which causes growth disorders for producer organisms. [14].

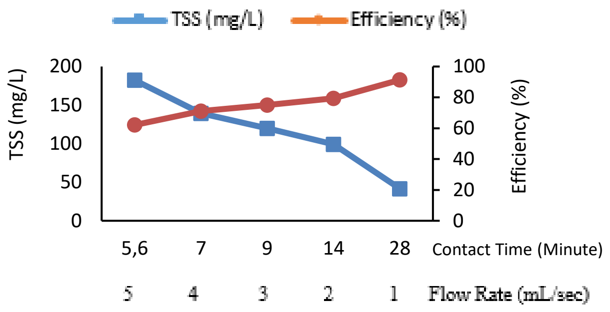

Figure 6. Deacreasing of TSS Sago Wastewater Level

Figure 6. shows the relationship between the decrease in TSS concentration and the applied flow rate, it can be seen that the smaller the applied flow rate, the longer the contact time, so that the decomposition potential of the Al electrode to release Al^{3+} is greater so that the $Al(OH)_3$ floc also increases. the greater it is. The longer the contact time with activated charcoal, the better the adsorption so that the efficiency of TSS concentration reduction is also greater. At a flow rate of 5 mL/second (dwelling time of 5.6 minutes) it can reduce TSS levels by 62.11% and at a flow rate of 1 mL/second (dwelling time of 28 minutes) it can reduce TSS levels by 91.32%. The highest decrease in TSS levels was obtained at the application of a flow rate of 1 mL/second where the initial concentration was 481 mg/L to 41.75 mg/L with a reduction efficiency of 91.32%. TSS is a pollutant that is in suspended form. When a material is suspended, the material is solid with a certain size. This solid material can be easily adsorbed into the coagulant or adsorbed into the air bubbles. The results of this adsorption will be separated upwards (flotation) resulting in a decrease in the concentration of TSS [15].

3.6. Study of Flowrate to Decrease Total Dissolved Solid (TDS) Level

Total dissolved solids represent the salts in the solution, including mineral salts from the water supply. Dissolved solids are important especially if the liquid waste will be reused after treatment, dissolved solids are a parameter of the amount of material dissolved in water.

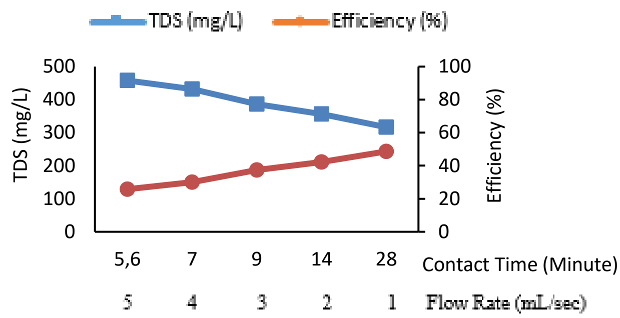

Figure 7. Decreasing of TDS Sago Wastewater Level

TDS levels during this research process decreased at each predetermined flow rate (Figure 7). The smaller the water flow rate, the longer the contact time will cause a decrease in the efficiency of TDS removal. The decomposition of TDS is carried out by autotrophic and heterotrophic microorganisms to synthesize cells. At a flow rate of 5 mL/second (contact time of 5.6 minutes), it can reduce TDS levels by 25.77% and at a flow rate of 1 mL/second (contact time of 28 minutes), it can reduce TDS levels by 48.66 %. The results of the highest reduction in TDS levels in this study were obtained at the application of a flow rate of 1 mL/second, where the initial sample contained TDS of 617 mg/L to 316.75 mg/L with a reduction efficiency of 48.66%. Although it is not required in the quality standard, TDS content analysis is still carried out to determine the number of dissolved solids in the wastewater sample.

3.7. Analysis of BOD and COD Level

COD (Chemical Oxygen Demand) is the need for chemical oxygen to break down all organic matter contained in water. BOD (Biological Oxygen Demand) is the amount of dissolved oxygen needed by aerobic bacteria to decompose organic matter. The results of the BOD and COD testing of sago wastewater before and after the electrocoagulation process and activated charcoal adsorption are presented in Table 2.

Table 2. BOD and COD Testing Result, Before and After Treatment

Testing Parameter	Sample (mL/S)	Conc. (mg/L)	Average (mg/L)	Standard
BOD	Before	297,00	**297,00**	**150 mg/L**
	After 1 (1)	37,80	**36,90**	
	1 (2)	36,00		
COD	Before	948,64	**948,64**	

	After 1 (1)	180,32	**178,75**	**300 mg/L**
	1 (2)	177,18		

Based on Table 2, the levels of these two parameters exceed the quality standards set by PerMenLH No. 5 of 2014 Appendix V with COD levels higher than BOD levels. This shows that there are many organic compounds in the water. COD value is always higher than BOD, this is because many organic substances are oxidized chemically but cannot be oxidized biologically [16].

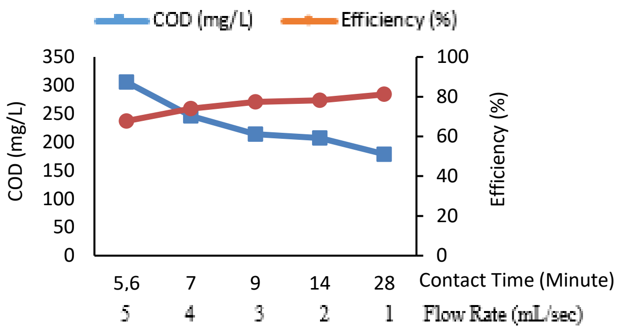

Figure 8. Decreasing of COD Sago Wastewater Level

Figure 8 shows the relationship of decreasing COD concentration to the applied flow rate. A flow rate of 5 mL/second (dwelling time of 5.6 minutes) can reduce COD levels by 67.77%. A flow rate of 1 mL/second (dwelling time 28 minutes) can reduce COD levels by 81.16%. In this study, the highest reduction in COD levels was at the application of a flow rate of 1 mL/second. Where the initial COD content was 948.64 mg/L to 178.75 mg/L with a decreasing efficiency of 81.15%.

The smaller the applied flow rate, the longer the contact time in the electrocoagulation container, and the more precipitate is obtained so that the electrocoagulation process is more intensive. This is in accordance with Faraday's law, namely the amount of substance obtained from the electrode is directly proportional to the amount of current flowing in the electrolysis cell. According to Faraday, the longer the contact time and current, the more charge is generated so that more coagulant is formed so that more organic pollutants are coagulated. Thus, the COD value is decreasing [6].

The decrease in COD levels was caused by the oxidation and reduction processes in the electrocoagulation reactor. At the electrodes, oxygen and hydrogen gas are formed which will affect the reduction of COD. The longer the electrocoagulation process lasts, the more colloids that are bound to form large-sized flocs or that float up. Due to the large number of flocculants formed and the flotation of light particles, the COD concentration decreased [17]. In addition, the adsorption process uses activated charcoal, the longer the contact time with activated charcoal, the better the adsorption. Same as COD, the optimum flow rate to reduce the highest BOD level is 1 mL/second (contact time 28 minutes). Where the initial BOD levels were 297.00 mg/L to 36.90 mg/L with a decreasing efficiency of 87.58%. The COD value indicates the total content of organic substances, both biodegradable and non-biodegradable, while the BOD value only measures the biodegradable part [18].

3.8. Study of Flowrate for Decreasing Permanganate Number

The presence of organic matter in water is closely related to the occurrence of physical changes in water, namely the emergence of unwanted odors, tastes, colors, and turbidity. Organic substances can be identified by determining the number of permanganates. Although KMnO4 as an oxidizing agent does not oxidize all existing organic substances, this method is very practical and fast to work. The organic substances that can be oxidized are carbohydrates, phenol, and the rest of the fermentation.

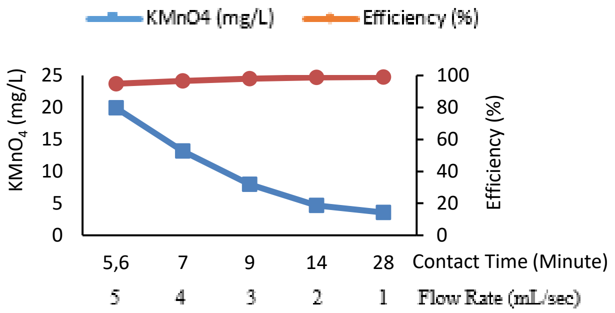

Figure 9. Decreasing of Permanganate Number

Based on Figure 9, the method used in this study with various flow rates can reduce the permanganate rate, where at a flow rate of 5 mL/second (5.6 minutes pause time) it can reduce the permanganate rate by 94.83%. At a flow rate of 1 mL/second (28 minutes pause time) it can reduce the permanganate number by 99.07%. The highest reduction results occurred at the application of a flow rate of 1 mL/second, where the initial sample contained a permanganate value of 385.68 mg/L to 3.57 mg/L.

Adsorption by activated charcoal by taking organic compounds from liquids in contact with activated charcoal. In the adsorption process, organic molecules that are in the liquid phase will be attracted and bound to the surface of the activated charcoal pores when the liquid passes through the activated charcoal. Therefore, the longer the contact time with activated charcoal, the better the adsorption.

In general, potassium permanganate is the same as BOD and COD which represents the content of organic matter in water. The measurement principle is the same, namely stating the consumption of oxygen used to oxidize organic matter, but potassium permanganate states the overall organic content or called Total Organic Matter (TOM). The difference lies in the oxidizing ability of potassium permanganate, where the oxidizing agent KMnO4 is not as good as the oxidizing agent used in COD measurements, so the concentration of potassium permanganate tends to be lower than that of COD.

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e-ISSN: 2776-4508; p-ISSN: 2776-1711
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3.9. Odor and Color Analysis of Sago Wastewater

Odor is an indication of the presence of spoilage of wastewater. The cause of the odor in wastewater is due to the presence of volatile materials, dissolved gases, and by-products of decaying organic matter. Sago liquid waste before being treated has a distinctive odor of sago from the decomposition process of organic compounds. However, after the electrocoagulation process and activated charcoal adsorption were carried out, the sago liquid waste became odorless. Odor removal is thought to occur because the filtration media using activated charcoal can reduce suspended and dissolved solids that give sago a distinctive odor.

The high content of organic matter in tapioca industrial wastewater makes the liquid waste very easy to decompose. So that even without treatment there will be decomposition of waste by microbes in the waste and in nature to produce simple organic compounds that cause odors such as methane (CH_4) and carbon dioxide (CO_2). The process of decomposition of organic compounds using microbes that produce methane (CH_4) and carbon dioxide (CO_2) is known as the methanogenesis process. The process of methanogenesis is the process of decomposition by methanogenic microbial species. This process occurs through two phases in which volatile fatty acids (VFA = Volatile Fatty Acid) and the final product of fermentation because of bacterial hydrolysis of fermentation are converted directly into methane and carbon dioxide [2].

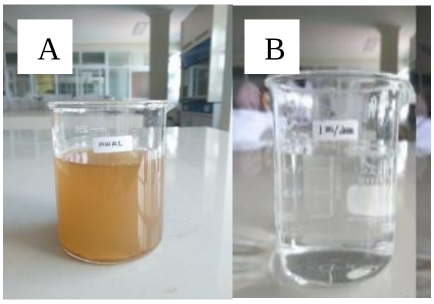

Figure 10. Sago Wastewater: Before Treatment (A) and After Treatment (B)

Figure 10 shows sago liquid waste before and after the electrocoagulation and adsorption process using activated charcoal. The liquid waste generated from the sago starch extraction process is brown in color and has a distinctive odor that will stink over time. The sago liquid waste before being processed is brown in color, presumably due to a browning reaction. The browning reaction occurred because the sago extraction wastewater was thought to contain oxidized phenolic compounds. The browning process occurs because of the phenol oxidase enzyme and oxygen associated with the phenolic compound substrate [19].

Figure 10 shows sago wastewater after electrocoagulation and adsorption using activated charcoal. Where after the electrocoagulation process and activated charcoal adsorption was carried out, the sago wastewater which was originally brown in color and smelled became colorless and odorless. The color change after processing is caused by the decrease in the value of dissolved solids and suspended solids. The smaller the applied flow rate also affects the color changes that occur in sago wastewater, where the wastewater at a flow rate of 1 mL/second (contact time 28 minutes) is clearer than the results at a flow rate of 5 mL/second (contact time 5.6 minutes).

4. CONCLUSION

Based on the results of the research that has been carried out, it can be concluded that the maximum removal efficiency for TDS, TSS. COD, BOD, TDS, $KMnO_4$ were 81.16%, 87.58%, 91.32%, 48.66%, 99.07%, for a flow rate of 1 mL/second, also other parameters also met have met the quality standards set by PerMenLH No. 5. Therefore, activated charcoal is effective in eliminating odors in sago liquid waste.

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