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Submission date: 15-Sep-2021 02:43PM (UTC+0700)
Submission ID: 1648926053
File name: er_Liquid_Waste_Treatment_Using_Electro_Coagulation_Membrane.pdf (239.17K)
Word count: 3544
Character count: 18927

Pulp and Paper Liquid Waste Treatment Using Electro Coagulation Membrane

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Abstract

The pulp and paper industry wastewater contains organic compounds such as lignins, resins, tannins, polysaccharides, alcohols and carboxylic acids. Pulp and paper liquid waste cannot be disposed of immediately because it can affect drinking water sources, endanger human health and cause unpleasant odors so that the waste must be treated first before being disposed of into the environment. The methods that can be used in the treatment of oily wastewater are ceramic membrane technology and electrocoagulation. The purpose of this study was to determine the effectiveness of the electrocoagulation-membrane combination method in treating pulp and paper wastewater in terms of the values of BOD, COD, TSS, pH and color. The application of this combined membrane and electrocoagulation method uses activated carbon from avocado seed waste and iron as electrodes at various variations, namely voltage (5 and 10 V), flow rate (1 and 3 L / Min), pressure (5 and 10 Psig) and processing time (30, 60 and 90 minutes). The results showed that the electrocoagulation-membrane method was effective in treating pulp and paper wastewater. After processing, the best operating conditions for each membrane were obtained. A pressure of 5 psig with a flow rate of 1 L / minute for membrane 1, a pressure of 5 psig with a flow rate of 1 L / minute for membrane 2 and a pressure of 5 psig with a flow rate of 3 L / minute for membrane 3.Based on the best operating conditions for each membrane , combined with the best operating conditions of the electrocoagulation method, namely a voltage of 10 V and an operating time of 90 minutes. The lowest COD and TSS values were obtained on membrane 2, namely 21 mg / L and 29 mg / L. The lowest BOD and color values were obtained on membrane 1, namely 27 mg / L and 42 TCU. The lowest pH value obtained on membrane 3 is 6.09.

Key words : electrocoagulation, fine membranes, activated carbon, pulp and paper wastewater.

1. Introduction

Pollution is the presence of harmful substances that enter the environment. Pollution is caused by pollutants. These pollutants can be solids, liquids, or harmful gases (Manisalidis et al., 2020). This pollution is a result of industrial activities and inappropriate vehicle emissions (Ghorani-Azam et al., 2016). Severe air pollution causes major health problems and consequences (Peng et al., 2019). One of the industries that causes environmental pollution is the pulp and paper industry (Singh & Arya, 2019). The pulp and paper (P&P) industry occupies a challenging position in relation to the natural environment. The wastewater generated is up to 70 m3 per metric ton of paper. Pulp and paper production is carried out in aqueous media and the addition of chemical additives can greatly change the nature of the waste generated. The waste is harmful to the surrounding environment (Hubbe et al., 2016).

Therefore, wastewater treatment from the pulp and paper industry is very necessary. Several methods can be used such as adsorption, advanced oxidation, membrane filtration, coagulation and flocculation, solar photo-catalysis, electro-coagulation, catalyzed ozonation, and solar photo-Fenton process (Simonič & Vnučec, 2012). In connection with the development of the pulp and paper industry in Indonesia, an integrated treatment is needed by the government, industry and the community in an effort to control waste so that it is always below the established quality standards. Therefore, an alternative technology is needed for proper waste treatment and easy operation, one of which is by using ceramic membrane technology.

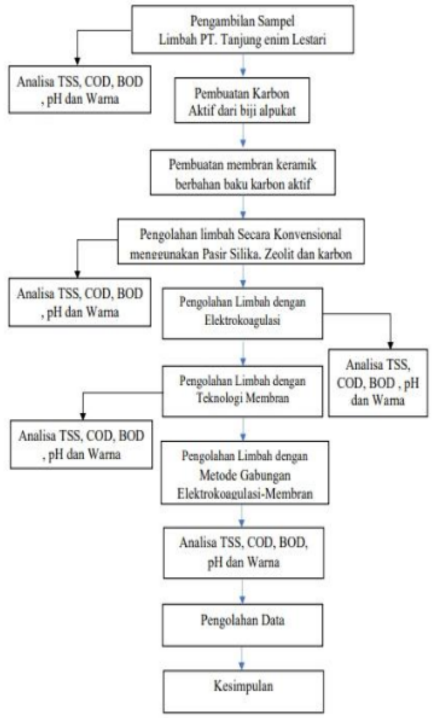
Membrane technology was chosen because it can separate various impurities in water without the need for the addition of chemicals, so that membrane technology is better than conventional technology (Dahlan, 2014).

2. Material and Methods

This research activity was carried out in the Chemical Reaction Engineering Laboratory, Catalysts, and Bioprocesses, Department of Chemical Engineering, Faculty of Engineering, Sriwijaya University. The time of the research is estimated to be from December 6, 2020 to February 30, 2021. The sample used is taken directly from the waste disposal of PT. Tel Pulp and Paper. The tools used in this study were filter paper,Furnace,Erlenmeyer, funnel, glass beaker, measuring cup, vacuum pump, oven, analytical balance, pH meter, grinder, stirring rod, 7mm plastic hose, porcelain cup, scissors, plastic bucket, ceramic membrane, cutting mils, fluw meter, dropper pipette, 1.5 L mineral drink bottle, housing membrane. The materials used are pulp and paper industrial wastewater, avocado seeds, clay (clay), aquadest, 30% KOH solution, bentonite, zeolite.

The stages of this research began with sampling at PT. Tanjung enim lestari pulp and paper. The next step is to make activated carbon from avocado seeds and manufacture ceramic membranes made from activated carbon as raw material. Next, the design of a conventional membrane filtration device is carried out using silica sand, zeolite, and activated carbon. Furthermore, using the filtration method using a ceramic membrane where the manufacture of ceramic membrane fillers has a variety of typesfirst consists of 70% clay, 10% activated carbon avocado seeds and 20% zeolite. The second membrane composition consisted of 70% clay, 15% activated carbon from avocado seeds and 15% zeolite. The third membrane composition was 70% clay, 20% activated carbon from avocado seeds and 10% zeolite.

The flow chart illustrates the work steps that will be carried out in this study and will simply display the stages of work that will be carried out. The research flow diagram can be seen in the following image:



Gambar. 3.1. Diagram Alir Penelitian

3. Results and discussion

The electrocoagulation method in this study uses electrodes that are powered by a voltage with a power supply. The variations used are voltages of 5, 10 and 15 volts, as well as processing times of 30, 60 and 90 minutes. These times and stresses have been tested previously by comparing several variables to determine the most effective variables in reducing the parameters of TSS, BOD, COD, Color and pH of pulp and paper wastewater. The most effective variables were obtained, namely a voltage of 15 volts and a time of 90 minutes, then three types of membranes with the best operating conditions were combined with the best operating conditions of electrocoagulation. A pressure of 5 psig with a flow rate of 1 L/min for membrane 1, a pressure of 5 psig with a flow rate of 1 L/min for membrane 2 and a pressure of 5 psig with a flow rate of 3 L/min for membrane 3.

The following table shows the comparison of the use of electrocoagulation against the best operating conditions of membrane 1 without electrocoagulation, namely a flow rate of 1 L/min and a pressure of 5 psig.

Tabel 3.1. Results of Pulp and Paper Waste Analysis with Electrocoagulation at the Best Conditions from Membrane 1 (Pressure 5 psig and Flow Rate 1 L/Min)

Jenis Perlakuan	Waktu (jam)	BOD	COD	TSS	Warna	pH	Fluks
Dengan Elektrokoagulasi	0,5	59	72	42	67	6,90	0,7407
	1	41	55	37	41	6,11	0,4814
	1,5	27	49	46	42	6,11	0,3086
Tanpa Elektrokoagulasi	0,5	106	97	121	175	7,30	0,6444
	1	53	72	61	110	6,21	0,2778
	1,5	41	52	32	45	6,11	0,1432

The following table shows the comparison between the use of electrocoagulation and the best operating conditions of membrane 2 without electrocoagulation, namely a flow rate of 1 L/min and a pressure of 5 psig.

Tabel 3.2. Results of Pulp and Paper Waste Analysis with Electrocoagulation at the Best Conditions from Membrane 2 (5 psig Pressure and 1 L/Minute Flow Rate)

Jenis Perlakuan	Waktu (jam)	BOD	COD	TSS	Warna	pH	Fluks
Dengan Elektrokoagulasi	0,5	63	70	51	101	6,91	0,8444
	1	32	49	32	76	6,12	0,4629
	1,5	39	21	29	43	6,09	0,3012
Tanpa Elektrokoagulasi	0,5	70	87	105	195	7,25	0,7259
	1	52	68	54	140	6,27	0,4444
	1,5	34	49	41	66	6,27	0,2099

The following table shows the comparison between the use of electrocoagulation and the best operating conditions of membrane 3 without electrocoagulation, namely a flow rate of 1 L/min and a pressure of 5 psig.

Tabel 3.3. Results of Pulp and Paper Waste Analysis with Electrocoagulation at the Best Conditions from Membrane 3 (Pressure 5 psig and Flow Rate 3 L/Min)

Jenis Perlakuan	Waktu (jam)	BOD	COD	TSS	Warna	pH	Fluks
Dengan Elektrokoagulasi	0,5	61	75	52	56	6,81	1,1259
	1	33	51	36	65	6,07	0,5296
	1,5	29	46	27	66	6,05	0,2667
Tanpa Elektrokoagulasi	0,5	62	73	112	153	7,32	1,0074
	1	59	62	92	87	6,92	0,5185
	1,5	32	63	54	76	6,88	0,2716

The best change in TSS was obtained 90 minutes of operation on membrane 2 with a flow rate of 1L/minute and a pressure of 5 Psig of 29 mg/L, compared to 41 mg/L without electrocoagulation. The highest TSS value was obtained on type 1 membrane with an operating time of 30 minutes, a flow rate of 1L/minute and a pressure of 5 Psig of 52 mg/L. It can be concluded that the longer the membrane-electrocoagulation process, the more effective the process, this is in accordance with the initial hypothesis. The longer the permeate taking time, the greater the decrease in TSS, COD, BOD and pH levels.

The pH value indicates the level of acid-base balance in the solution, waste that is very acidic or has a low pH is very dangerous for the environment due to its corrosive nature, while waste that is too alkaline or has a very high pH value is also not good for the environment. The best decrease in pH was found in type 3 membranes with an operating time of 90 minutes, flow rate of 3 L/M and pressure of 5 Psig. The highest pH value was found in type 2 membrane with an operating time of 30 minutes, a flow rate of 1 L/M and a pressure of 5 Psig.

The decrease in pH that occurs in the electrocoagulation method is because the electrocoagulation process produces more H+ ions produced through the water reduction reaction at the cathode causing the acidity level to decrease, while in the membrane method a decrease in pH can occur because activated carbon has a role in lowering the pH.

The results of the pulp and paper wastewater treatment using electrocoagulation appear to have an effect in decreasing the BOD value faster than without the use of electrocoagulation. It can be seen from the graph, that at 30-90 minutes of operation, BOD decreased. This shows that the longer the process, the more electrocoagulation decomposes waste organic compounds.

The results of pulp and paper wastewater treatment with the addition of the electrocoagulation method gave a higher COD value reduction. From the graph above, it can be seen that the use of the electrocoagulation method resulted in a decrease in COD along with increasing operating time. Pulp and paper liquid waste can reduce its COD content from the original high COD content of pulp and paper waste to below the waste water quality standard. The use of electrocoagulation also reduces high COD levels. This proves that the use of electrocoagulation is considered feasible for use in liquid waste, pulp and paper.

The addition of the electrocoagulation method to the pulp and paper wastewater treatment process tends to show a smaller flux quantity than without using electrocoagulation. The cause of this is because in the electrocoagulation process there will be floc formation due to the applied voltage. The floc that is formed will make the concentration of the liquid waste higher, because the floc is a lump of particles in the waste. The flux obtained is lower than without using electrocoagulation because the volume that comes out of the membrane is also reduced, because the filtration process in the membrane will tend to be longer, this is in accordance with research conducted by Dahlan (2018) where the higher the inlet concentration, the output volume obtained also decreased.

Coagulation is the change of the sol phase into a gel phase assisted by a coagulant. Latex will become a coagulum if the electrical charge is lowered (dehydration), the pH of the latex is reduced (addition of acid H+), and the addition of electrolytes. Decrease in latex pH can occur naturally and the addition of coagulants (Oktriyedi et al., 2021). In the EC process, the coagulant is produced in situ by electrolytic oxidation of a suitable anode material. This technology removes metals, colloidal and particulate solids, as well as dissolved inorganic pollutants from aqueous media by introducing highly charged polymeric metal hydroxide species (Mollah et al., 2001).

Research by Zaied & Bellakhal (2009) has shown the application of the electrocoagulation method in the processing of black liquid from the paper industry. The effect of variables such as electrolysis time, current density, type of electrode material and initial pH on polyphenol removal, COD and color intensity was determined. The final pH of the treated black liquid is nearly neutral, which allows it to be directly discharged into the natural watercourse. Consequently, electrocoagulation can be considered as a suitable alternative to existing methods or applied as a pre-treatment step of biological processes used for black liquor treatment (Zaied & Bellakhal, 2009).

The EC method provides inspiring results from pulp and paper industrial wastewater treatment. Investigations show that the EC method is a versatile process and has strong removal efficiency against BOD (Biochemical Oxygen Demand), COD (Chemical Oxygen Demand), Color, TOC (Total Organic Carbon) and TDS (Total solved solid) from pulp and paper industry waste. . All physiochemical parameters of EC treated water were found to be below the permissible limits set by WHO (World Health Organization) and CPCB (Central Pollution Control Agency); Thus, it can be used as an alternative to reduce the input of clean water in agriculture. The pH of the treated water after EC treatment was found to be within the permissible limits of the environmental standard, it was obtained close to a neutral pH. This is a significant feature of the EC method and does not require a neutralization process compared to conventional treatments (Kumar & Sharma, 2019).

A simple EC apparatus has been made using iron and aluminum as sacrificial electrodes. The EC equipment is then used for local paper mill wastewater treatment under various operating condition. However, in order to completely remove COD and BOD from wastewater, possible secondary treatment using adsorption techniques is still required (Yuliani et al., 2017).

This study assessed the trend of dye removal from synthetic textile wastewater at low voltages (ie, 1 V and 2 V) and high voltages (ie, 2 V, 4 V, 6 V, 8 V and 10 V) at different NaCl concentrations. use standalone. EC, self-contained membrane filtration and EC integrated membrane systems. For standalone membrane filtration, the dye removal efficiency is so low that it is not possible. Nanofiber membranes can be reused at least twice. The problem can be overcome by the development of advanced nanofiber membranes with enhanced mechanical strength which will be a follow-up study. In addition, the EC-integrated membrane system increases the dye removal efficiency by 10-30% and the sludge formed during EC is also separated at the same time. Therefore, the treatment of dye wastewater using an integrated system can be concluded to be quite effective, even at lower voltages exceeding the efficiency of the EC and membrane methods alone (Saad et al., 2020).

The proposed technology with the use of electrocoagulation/membrane filtration requires about 15% less chemicals compared to conventional processes. Submerged microfiltration using ceramic membranes exhibits a very stable filtration operation with easy removal of the filtration cake with back-pulsed air. The proposed new hybrid process of membrane electrocoagulation/filtration shows very promising results for solving the problem of removing Se as well as other metals (especially As, Cu, Pb) in industrial wastewater treatment (Mavrov et al., 2006).

The efficiency of the electro-coagulation treatment method for the treatment of wastewater from the paper industry is considered in this study. Three treatment methods were considered. Wastewater is first treated using iron salts or carbonate chemical coagulant only. Then the wastewater is treated by electro-coagulation method only. Finally electro-coagulation with the addition of chemical salts was studied. Cheap iron plates are used for sacrificial electrodes in the electro-coagulation method. The effect of operating conditions on maintenance efficiency was investigated. Finally, combining a chemical salt coagulant with an electrocoagulation technique results in an increase in the removal efficiency. The addition of iron salts was found to be more suitable for EC than the addition of Ca. The findings of this study confirm the findings of previous studies as described in the background section of this paper. Based on these findings, it can be concluded that the best option for treating wastewater from the paper industry is the electro-coagulation technique, at optimal operating conditions, with the addition of iron salts (Lafi, 2011).

The treatment of pulp and paper mills' wastewater using PACl coagulant coupled with PAMs enhanced the reduction/removal of turbidity, TSS, and COD when compared to the results obtained when the coagulants and flocculants were used alone. However, any increase in the PAMs dosage does not have a significant effect on PACl coagulation. PACl, coupled with Organopol WPB20 was the best system from among all the systems studied and showed the highest efficiency in terms of reduction in turbidity, the removal of TSS, and the reduction in COD. The additions of PAM improved the treatment performances. The calculations of Langelier's saturation index (LSI) showed values from -0.2 to - 0.5 in the acidic samples. In this case the water was in carbonate equilibrium and the minimum dosage of calcium carbonate, that clogs the membrane, precipitated (Simonič & Vnučec, 2012).

The pulp and paper (P&P) industry worldwide has made substantial progress in treating process water and wastewater, thereby limiting the discharge of pollutants into receiving water. The review covers a variety of wastewater treatment methods, providing P&P companies with cost-effective ways to limit the release of biological or chemical oxygen demand, toxicity, solids, color, and other indicators of pollutant load. Conventional wastewater treatment systems, often consisting of primary clarification followed by an activated sludge process, have been widely applied in the P&P industry. Higher pollutant removal rates can be achieved with additional treatments, which may include anaerobic biological stages, advanced oxidation processes, bioreactors and membrane filtration technologies. Improved performance of wastewater treatment operations can often be achieved with effective measurement technologies and with the addition of strategic agents including coagulants, flocculants, filter aids, and optimized fungal or bacterial cultures. In addition, the P&P plant can implement upstream process changes, including dissolved-air release (DAF) systems, filtration storage, and kidney-like operations to purify process water, thereby reducing the pollutant load and the volume of effluent discharged to the end-of-pipe wastewater treatment plant (Hubbe et al., 2016).

4. Conclusions

The results of pulp and paper wastewater treatment using electrocoagulation appear to have an effect in decreasing BOD values faster than without the use of electrocoagulation.

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