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The Effect of Flow Rate Discharge on TDS, pH, TSS, and Cu in Electrocoagulation with Continuous Reactors

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Abstract

The gold amalgamated wastewater flow can seep into the soil and partly lead to ditches and end into seasonal rivers. The waste contains several heavy metals, including Hg and Cu. In the rainy season, runoff can occur which dissolves pollutants in wastewater at the ground surface so that water can flow into sewers or will seep into the ground and heavy metals contained in a waste can contaminate groundwater. Thus, it is possible to contaminate the quality of surface water (rivers) and groundwater (wells) around gold processing and wastewater disposal sites. The purpose of this study was to determine the effect of flow rate as an independent variable in continuous electrocoagulation. This research was a laboratory scale and used gold processing wastewater from gold washing activities. Electrocoagulation in this study using a continuous reactor. Based on the results of the processing carried out by the continuous system electrocoagulation method. The discharge of Q1 (0.0156 m^3/s) reduces the levels of TDS, pH, TSS, and Cu in wastewater greater than the discharge of Q2 (0.018144 m^3/s). These results have also exceeded the TSS parameter quality standard which refers to the Decree of the Minister of the Environment No. 202 of 2004 about Quality Standards for Wastewater for Businesses and/or Mining of Gold and/or Copper Ore, so it is safe to dispose of into the environment.

Keywords: Electrocoagulation, Effect, Flow rate

I. INTRODUCTION

Water scarcity can occur due to climate change and population changes, resulting in an imbalance between demand and availability. Reuse of wastewater, especially in the mining industry, can be minimized as an alternative. My wastewater quality varies greatly from each mine, based on their type of mine and local conditions. General characteristics of wastewater, mines can be identified as follows: hardness such as $CaCO_3$, high suspended solids (turbidity), and low conductivity, Chemical Oxygen Demand (COD), and Biological Oxygen Demand (BOD), the moderate concentration of minerals (K, Ca, P2O5, etc.), and heavy metals (Lottermoser, 2010). This wastewater must be managed and treated properly, to prevent pollution on the water or solid. Various wastewater treatment methods are available applied to mine wastewater treatment, such as coagulation, flocculation, sedimentation, exchange, and membrane technology, which can produce better water quality. The choice of processing method is affected by various criteria: economy, facilities, and speed.

Koka Kulonprogo is an area that has gold mining business activities. The mining method applied using underground mining or mining methods simple underground. The mining results are processed in a simple and traditional method. The processing of gold ore creates entropy in the form of wastewater and tailings. Mine processing materials using simple technology, still using hazardous materials such as mercury (Hg), without adequate processing knowledge and technicality, so it can create serious environmental problems (Keraf, 2001). The handling of wastewater and tailings is still simple. Waste yield amalgamation of gold is dumped directly into the ground around the site amalgamation processing. The gold amalgamation wastewater stream seeps inside the land and partly leading into ditches and ending into seasonal ditches and rivers that are around. Solid waste or tailings will settle on the surface soil and will accumulate constantly covering the soil layer if activity waste disposal is always carried out. The waste contains several heavy metals other than Hg include Cu, Au, Cd (KepMenLH, 2004). In the rainy season, runoff can occur which dissolves pollutants in wastewater at the surface of the soil so that it can flow into sewers or will seep into the ground and can contaminate groundwater below the surface by heavy metals. Thus, it is possible to contaminate surface water quality (river) and groundwater (wells) around the gold processing and water disposal sites the waste.

Reduction of heavy metals in wastewater can be using treatment with chemistry (coagulation, absorption, and ion-exchanged), physical chemistry (electrocoagulation and solidification), and biologically (phytoremediation and bioremediation). Wastewater treatment which will be used physically and chemically (electrocoagulation and solidification). Electrocoagulation is a method of electrochemical water treatment where the anode releases an active coagulant in the form of metal ions (usually aluminum or iron) into the solution, while at the cathode an electrolytic reaction occurs in the form of releasing hydrogen gas (Ni'am & Othman, 2014; Nouri et al, 2010; Touahria et al., 2016; Zongo et al., 2012). This electron-releasing reaction will help treat the wastewater(Butler et al., 2011). The electrocoagulation method can use with 2 types of reactors namely batch and continue. The two processes will produce different water flows, chemical interactions, concentration gradients, different floc formation, and precipitation during the process (Mamelkina et al., 2017).

Based on the research before, electrocoagulation has a treatment time of 20 to 60 minutes. With a voltage of 40 V to remove chromium ions using an iron electrode (Nouri et al., 2010). It can be concluded that the density and time of electrolysis, along with the initial concentration were able to determine the success of the removal rate (Shafaei et al., 2010). Gold processing wastewater treatment in the village Jendi, Wonogiri Regency using electrocoagulation with aluminum electrodes in batch reactors can reduce TSS, Hg, and Cu. Research result indicates that at a contact time of 30 minutes with a voltage of 9 volts has high effectiveness in reducing TSS, Hg, and Cu (Asrifah, 2019).

The electric field in the electrolytic cell will make colloid particles migrate to the anode so that the coagulation-flocculation process occurs. Process electrolysis of water also causes the formation of tiny bubbles of oxygen and hydrogen to the anode and cathode, respectively. These microbubbles are absorbed by the flocculating material and will rise to the surface (Zongo et al., 2012). Batch reactors have process drawbacks only run once in each processing so it's on a certain time the process will stop, the processing time is long, this process model only can be done for industries with a small production capacity. During the electrocoagulation process, sludge will form. This mud contains metal oxides and hydroxides (Mamelkina et al., 2017). The contaminants contained in the sludge will be removed by electrocoagulation. Heavy metals that contain in the wastewater will be attached to the plates and the sludges will be deposited. The purpose of this research is to know the effect of flow rate discharge as an independent variable in continuous electrocoagulation. The independent variables whose effects were observed on these parameters were TSS, TDS, Cu, and pH.

II. LITERATURE REVIEW

Electrocoagulation is a method of treating many types of wastewater which usually combines electrochemical, coagulation, and flocculation processes (Bazrafshan et al., 2015; Rusdianasari et al., 2019). This method uses a simple mechanism, namely by cathodic deposition with several types of reactors, one of which is a tank cell (Chen, 2004). The advantages of this method include high efficiency, simple operation, high settling ability, does not produce sludge after treatment, and relatively adequate cost (Abbas & Ali, 2018; Naje et al., 2017). Several studies have been conducted using the electrocoagulation method to minimize wastewater, starting from wastewater from herbicides, medical, water hardness, and heavy metals (Danial et al., 2017; Malakootian and Yousefi, 2009; Naje et al., 2017; Muharam et al., 2017; Bazrafshan et al., 2015). Several types of heavy metals that can be effectively removed using electrocoagulation are Nickel (Ni), Chromium (Cr), Cadmium (Cd), Copper (Cu), Iron (Fe), Zinc (Zn), and Mercury (Hg) (Bazrafshan et al., 2015; Cataldo Hernández et al., 2012; Chaturvedi, 2013; Un and Ocal, 2015; Wulan et al., 2017). Copper (Cu) removal significantly reached 98.90%, influenced by current density and pH (Un and Ocal, 2015). Metal removal is also influenced by the input voltage, operating time, and high and low metal concentrations. Based on a study conducted by Wulan et al. (2017), the optimum conditions for achieving 79-98% metal removal are with a voltage of 30V, and an operational time of 30 minutes for batch or continuous. While Petsriprasit et al. (2010) in their research showed that with the same operating time (30 minutes) Copper (Cu) can be removed as much as 99% in the initial pH condition of 5.

III. RESEARCH METHODOLOGY

This research is a laboratory scale. The wastewater samples used were taken from gold processing, particularly the gold washing process. There are three parameters analyzed, namely the quality of well water, river water, and gold processing wastewater. Well, water (groundwater) and river water quality parameters will be tested based on Minister of Environment Decree No. 202 of 2004 concerning Wastewater Quality Standards for Gold and/or Copper Mining Business and/or Activities and additions to several parameters. The parameters tested before and after the wastewater treatment process consists of: Total Dissolve Solid (TDS), Total Suspended Solid (TSS), pH, and Copper (Cu). The quality standard for each parameter is in accordance with Table 1. The research variables can be seen in Table 2.

Table 1. Water Quality Standard

Parameters	Units	Quality Standard*)	Analysis Method
TDS	mg/L		SNI 6989.3-2019
pH	-	6-9	SNI 06-6989-11-2004
TSS	mg/L	200	SNI 6989.3-2019
Cu	mg/L	2	SNI 6989.6 -2009

*) Minister of Environment Decree No. 202 of 2004 about Wastewater Quality Standards for Businesses and, or Gold and, or Copper Ore Mining Activities

Table 2. Variables Used In The Study

No	Independent Variable	Dependent variable
1.	Flow rate discharge	TDS, TSS, Cu, Hg, Efficiency

Electrocoagulation in this study using a continuous reactor. This reactor is divided into 3 bulkheads, each of which has 2 electrodes consisting of 1 pair of anode and cathode. The operation process runs for 360 minutes. Wastewater flows from the reservoir with a volume of 2,592 L. This system is provided by gravity control from the reservoir to the reactor batch. In this study, wastewater is generated with a constant flow rate. The flow rate variations used were 0.2592 L / minute and 15.552 L / minute. The experiment was carried out with the Al electrode plate using a strong current of 5 amperes which is equal to a voltage of 12 volts. The time used to observe changes in each parameter is every 10, 20, 30, 40, and 50 minutes. At that time, the TDS, pH, TSS, and Cu parameters will be tested. This aims to determine at what time the waste quality decreases effectively. The experimental reactor of electrocoagulation using the continuous method shows in Figure 1.

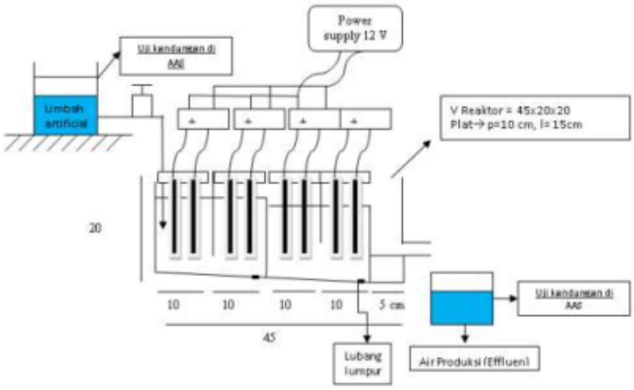

Figure 1. Experimental Electrocoagulation Setup

The analysis method used is statistical regression analysis. This method is used to determine the effect of a parameter on other parameters. The calculation of the removal efficiency of TSS, Cu, and Hg after EC treatment was carried out using the formula 1 :

$$CR(\%) = \frac{C_0 - C}{C_0} x\, 100 \quad (1)$$

Determination of the optimal treatment of the two discharge variations is obtained by calculating the average of R of wastewater parameters.

IV. RESULT AND DISCUSSION

Wastewater that used in this research is gold mining washing wastewater which is produced after the gold processing process. The waste is then taken for initial testing and continued processing with the continuous system electrocoagulation method. The gold washing wastewater is subjected to laboratory tests for parameters TDS, pH, TSS, and Cu which will then be compared with quality standards. Based on laboratory results, testing of the four parameters obtained results which can be seen in Table 3. Based on the Table, the TSS parameter has a value that is far above the quality standard threshold. Meanwhile, the TDS, pH, and Cu parameters were still within the permitted quality standard threshold.

Wastewater from the gold processing process is in **Table 3**. The test results of the initial waste before electrocoagulation processing in a continuous reactor were compared to raw gold wastewater processing, namely the Minister of Environment Decree No. 202 of 2004 concerning Wastewater Quality Standards for Gold and or Copper Ore Mining Business and or Activities.

Table 3. Parameter value in gold processing treatment.

Parameters	Units	Analysis Method	Quality Standard *)	Result
TDS	mg/L	SNI 6989.3-2019	-	239,5
pH	-	SNI 06-6989-11-2004	6-9	7,45
TSS	mg/L	SNI 6989.3-2019	200	5863
Hg	mg/L	Mercury Analyzer	0,005	4,47
Cu	mg/L	SNI 6989.6 -2009	2	0,08

*) Minister of Environment Decree No. 202 of 2004 about Wastewater Quality Standards for Businesses and, or Gold and, or Copper Ore Mining Activities

These quality standards provide a standard of whether the wastewater must be treated or not. If the level of parameters present in the wastewater exceeds the quality standard, the wastewater must be treated. The test results of the wastewater samples are as follows:

- The TDS level in wastewater is 160 mg/L. TDS levels are not required in standard gold processing wastewater quality. TDS can be used to determine the level of dissolved particles in the wastewater. One of the dissolved solids is salt.
- The TSS level is 5863 mg/L and exceeds the water quality standard of gold processing waste by 200 mg/L. High TSS levels will affect the aesthetics of water and cloudy colored water. The high TSS in wastewater indicates the high suspended particles in the wastewater. The suspended particles in wastewater are dissolved metals, dissolved hash particles, clay, and others.
- Cu level in wastewater is 0.13 mg/L, where the level has not exceeded the quality standard of wastewater, which is 2 mg/L.
- Hg level in wastewater is 4,47 mg/L, where the level has bigger than the quality standard of wastewater. the quality standard of Hg in wastewater is mg/L. hg is one of the heavy metals that are harmful to living things and the environment.

The physical condition of wastewater can be seen in Figure 2. Wastewater is cloudy in color and contains dissolved particles and suspended solids. There is low in esthetic. By taking into account these conditions, the TSS and Hg levels are above the environmental quality standard. This wastewater can endanger the environment if it is disposed of directly into the environment. High levels will be a burden on the environment for self-purification. The ability of the environment to carry out heavy self-purification has made the pollutant known to the environment. The spread of Hg in the environment will take place through the hydrological cycle and the food flow cycle in an ecosystem in the environment. The highest level of tropical food is humans. So that the accumulation of Hg in the human body can result in carcinogens (cancer-causing). Heavy metal in the ecosystem will experience bioaccumulation and biomagnification. Low levels of Cu can become dangerous if bioaccumulated. In this process, Cu will accumulate in the human body and other living things. So that the Cu still has to be processed so that it is not harmful to the environment.

Figure 2. The physical condition of wastewater

The gold washing wastewater treatment will be treated with a continuous electrocoagulation tank. The fixed variable used in this study is the flow rate of water. The discharge used in this research is 0.0156 and 0.0181 m^3/s. Then each processing was carried out with a contact time of 0 minutes, 10 minutes, 20 minutes, 30 minutes, 40 minutes, and 50 minutes. Based on the processing that has been done, a decrease in the level of waste varies with each parameter. The processing efficiency for each parameter obtained also has varying values. The result shows in Table 4.

The TDS parameter has an initial waste content of 239.5 mg/L and after processing the lowest level is obtained, with content 143 mg/L at 0 minutes processing time and discharge of 0.0156 m^3/s. The content in TDS consists of dissolved solids in water in the form of fine sand. The 0 minutes time on the continuous treatment unit is calculated when the first water exits through the continuous unit reactor. The solid will be deposited at the bottom of the tank so that the level will decrease when the water comes out of the tank. The highest reduction efficiency in TDS levels was 40.2923%.

The pH parameter before processing had a value of 7.45 and decreased to 7.05 after processing. Processing efficiency in pH parameters is 5.4326%. The highest effectiveness value occurs at a discharge of 0.0156 m^3/s with a contact time of 10 minutes. The TSS parameter has a quite drastic decrease from before processing to after processing, namely 5,863 to 178 mg/L. The processing efficiency that occurs in the TSS parameter is 96.9640%. This causes the TSS parameters to meet the existing quality standards after processing using the batch system electrocoagulation method. The highest effectiveness occurs at a discharge of 0.0156 m^3/s and a contact time of 20 minutes. The processing result on the Cu parameter has an effectiveness of 87.5%. This value was obtained due to a decrease in Cu levels in wastewater from 0.08 mg/L to 0.01 mg/L. The decrease occurred at a discharge of 0.0181 m3/s with a contact time of 30 minutes.

The level of Hg contained in gold washing wastewater treatment has high content. High levels of mercury may have originated from the use of high doses of mercury in the gold leaching process. This content is far above the predetermined quality standard threshold of 4.47 mg/L. The continuous electrocoagulation system treatment process shows a decrease in Hg levels in wastewater until the efficiency reaches 97%. The highest efficiency is found in processing with a discharge of 0.0156 m3/s at 50 minutes. However, this reduction still doesn't reach the existing quality standard, with a value of 0.0005 mg/L.

Table 4. Result of Experimental Process

No.	Flow Discharge (m^3/s)	Times (minutes)	TDS (mg/L)	pH	TSS (mg/L)	Hg (mg/L)	Cu (mg/L)	% TDS Removal	% pH Removal	% TSS Removal	% Hg Removal	% Cu Removal
1	0,0156	0	143	7,19	3990	6,77	0,06	40,2923	3,5547	31,9461	-51,4541	25
2	0,0156	10	160	7,05	2658	0,64	0,04	33,1942	5,4326	54,6648	85,6823	50
3	0,0156	20	151	7,13	178	65,08	0,04	36,9520	4,3595	96,9640	-1355,9284	50
4	0,0156	30	148	7,09	391	0,26	0,04	38,2046	4,8960	93,3311	94,1834	50
5	0,0156	40	148	7,11	300	0,2	0,04	38,2046	4,6278	94,8832	95,5257	50
6	0,0156	50	147	7,12	183	0,11	0,07	38,6221	4,4936	96,8787	97,5391	12,5
7	0,0181	0	156	7,37	165	1,00	0,05	34,8643	1,1402	97,1857	77,6286	37,5
8	0,0181	10	151	7,39	534	0,59	0,04	36,9520	0,8719	90,8920	86,8009	50
9	0,0181	20	152	7,39	444	0,25	0,02	36,5344	0,8719	92,4271	94,4072	75
10	0,0181	30	152	7,35	228	0,43	0,01	36,5344	1,4085	96,1112	90,3803	87,5
11	0,0181	40	150	7,39	215	62,37	0,05	37,3695	0,8719	96,3329	-1295,3020	37,5
12	0,0181	50	149	7,32	421	0,19	0,08	37,7871	1,8109	92,8194	95,7494	0

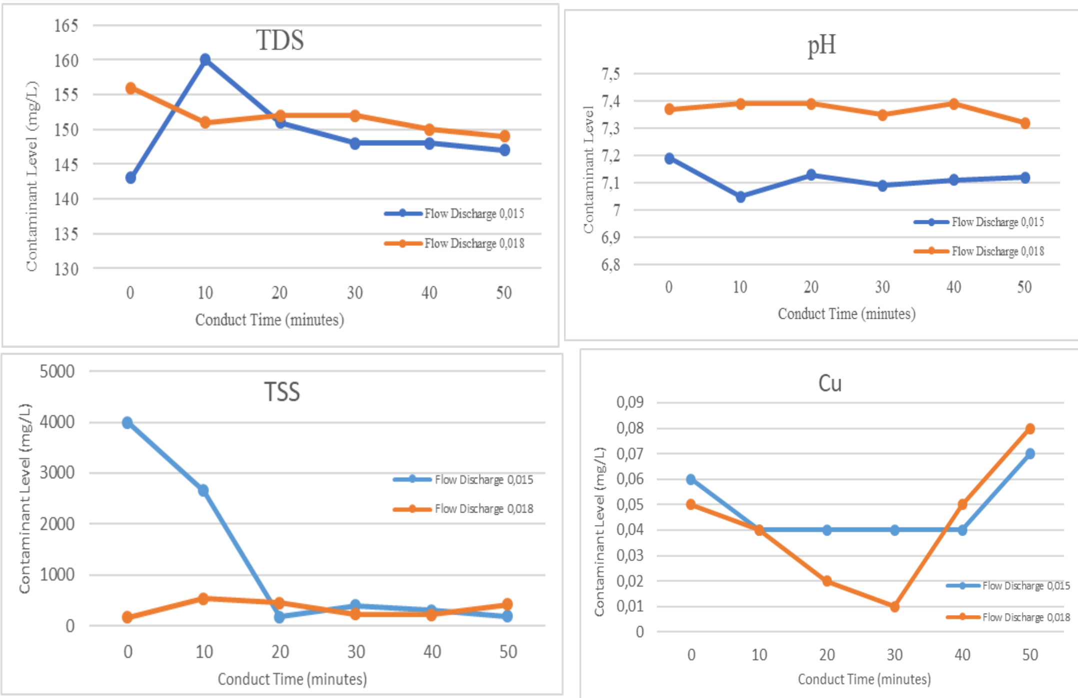

Figure 3. Graph of the relationship of variation of wastewater flow debit with parameter conditions in wastewater

Table 5. The value of correlation coefficient (R) and regression equation of each parameter in the variation of gold processing wastewater treatment.

Parameter (mg/L)	Q1 (Discharge) 0.015552 m³/s		Q2 (Discharge) 0.018144 m³/s	
	R	Regression Equation	R	Regression Equation
TDS	17.7 %	Y= 37.012 +0.023X	70.4 %	Y= 154.381 -0.109X
Cu	4.04 %	Y= 44.048 -0.179X	13.4 %	Y= 63.095 -0.607X
TSS	71.4 %	Y= 46.562+1.262X	0.14 %	Y=94.425-0.005X
Hg	4.16 %	Y=-331,309+6.356X	14.75 %	Y=148.258-11.599X
Average	24.58%		24.17%	

The results of the calculation of the value of R for the wastewater quality parameters in the table above can be seen:

- TDS parameters
  The discharge Q2 has a good effect on the efficiency of reducing TDS in gold processing wastewater treatment. The discharge Q2 has a bigger correlation coefficient (R) for the TDS parameter than Q1. The value of R in Q2 is 70.4% or 0.704 (positive and close to 1). While the value of R in Q2 has a value of 17.7% or 0.177 (positive and runs the number 0).
- Parameters Cu
  The discharge Q1 and Q2 have little effect on the efficiency of Cu reduction in gold processing wastewater. Both have positive R values and are close to 0. The correlation coefficient in Q1 is 4.04% or 0.404, while Q2 is 13.4% or 0.134. Q2 has a greater correlation coefficient than Q1.

- TSS parameters
  The discharge Q1 has a good effect on the efficiency of TSS reduction. Q1 has a greater correlation coefficient than Q2. The value of R in Q1 is 71.4% or 0.714, while it's positive and close to 1. The value of R in Q2 is 0.14% or 0.014 (positive and is close to 0).
- Hg parameters
  The discharge Q1 and Q2 have little effect on the efficiency of Cu reduction in gold processing wastewater. Both have positive R values and are close to 0. The correlation coefficient in Q1 is 4.16% or 0.416, while Q2 is 14.75% or 0.1475. Q2 has a greater correlation coefficient than Q1.

Determination of the optimal treatment of the two discharge variations is obtained by calculating the average of R in wastewater parameters. The largest average of R from two discharge treatments determines the optimal treatment. The mean value of R in Q1 (the average of R = 24.58%) is greater than the discharge treatment Q2 (the average of R = 24.17%). Q1 is the optimal treatment for gold processing wastewater treatment. Flow discharge affects the residence time of wastewater in the electrocoagulation process. The smaller the wastewater flow, the greater the residence time. With a longer residence time, the electrochemical reaction process in wastewater is more optimal.

V. CONCLUSION AND FURTHER RESEARCH

Based on the results of the processing carried out by the continuous system electrocoagulation method, the discharge is 0.0156 m3 / s to reduce the levels of TDS, pH, TSS, and Cu in wastewater. These results have also exceeded the quality standards for TSS parameters which refer to the Minister of Environment Decree No. 202 of 2004 about Wastewater Quality Standards for Businesses and, or Gold and, or Copper Ore Mining Activities, so it is safe when disposed of into the environment.

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