



The Treatment of Hospital Wastewater Using Iron Electrode Electrocoagulation: Analysis by Response Surface Methodology

Darmadi, Mirna Rahmah Lubis, Adisalamun*

Chemical Engineering Department, Universitas Syiah Kuala, Banda Aceh, INA

*E-mail: adisalamun@che.unsyiah.ac.id

Article History

Received: 26 May 2019; Received in Revision: 15 July 2019; Accepted: 16 July 2019

Abstract

Hospital wastewater basically contains organic and inorganic compounds. Levels of these compounds can be determined by testing of Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Dissolved Solid (TDS), and Total Suspended Solid (TSS). In order to treat the hospital wastewater, the electrocoagulation method using Fe-Fe electrodes is proposed in this research and response surface method is used for optimizing the response variable. This research shows that the relationship between the process variables and the TSS is significantly influential. As the contact time (15, 30, and 45 minutes) is longer and the voltage (6, 9, and 12 volts) is higher, the reduction percentage of TSS increases. However, the electrolyte solution (0–1 M) has little influence/significance to a response variable of TSS. The percentage of TSS reduction declines at contact time 45 minutes. The model recommended is a quadratic form with an error less than 1.6%. In such a way, the optimum condition is at contact time 36 minutes, voltage 12 volts, and the sodium chloride concentration 0.1 M. The highest reduction percentage of TSS is at 30 minute contact time, 12 volt, and 0.1 M sodium chloride concentration with TSS reduction percentage of 72.45%.

Keywords: electrocoagulation, electrolyte, hospital wastewater, response surface

1. Introduction

The hospital wastewater (HWW) is all hospital liquid waste including infectious waste from medical and non-medical activities. The wastewater contains high number of contaminants affecting the environment (Rudin, 2018), such as iodine (Rahmi *et al.*, 2018). The HWW treatment could be carried out by various ways, such as membrane bioreactor (Alrhoun, 2014).

Other methods includes ozonation (Hansen, 2016) and electrocoagulation. Operating procedures affect the amount of the formed waste (Sabeen, 2018). Based on ways of the HWW treatment, electrocoagulation is selected to treat hospital wastewater with the physical process because the process is more advantageous than conventional methods of waste treatment. It could reduce more quickly the smallest colloid/particle content or form finely colloidal particles.

The reduction is not capable in conventional treatment (Wardhani, 2012). Electrocoagulation is one of combined methods of waste treatment by electrolysis and coagulation (Lubis *et al.*, 2019). It treats textile wastewater (Bazrafshan, 2014) and municipal wastewater (Sharma and Chopra, 2014). Electrocoagulation using iron electrode (Fe-

Fe) takes place if the direct current is applied in electrocoagulation cell (Pirkarami and Ebrahim, 2014). The iron anode will oxidize water to produce oxygen gas (O_2). Fe^{2+} ions from the anode dissolving is reduced by OH^- ions to form $Fe(OH)_2$. The anode will produce $Fe(OH)_2$ floc. Bubbles of O_2 and H_2 gases produced during the process will help pollutant floated to the surface. The hospital wastewater treatment aims to prevent the environmental pollution and to prevent health disorder. However, previous research did not investigate TSS in the hospital wastewater using Response Surface Methodology (RSM). RSM is selected because the method can optimize a process by analyzing the effect of variables on the efficiency (Lubis, 2019).

This research aims to treat the total suspended solid and optimize it statistically by RSM using Design Expert 10.0.1 software through Box-Behnken Design (BBD). BBD has not been used to determine the TSS performance yet. Three important factors (contact time, voltage, and electrolyte concentration) are considered as variables; TSS removal is defined as a response. The variables are varied according to RSM with the BBD help. The BBD and RSM are used for modelling the effect of variables on TSS performance. The variable interactions are studied to obtain the optimum response.

2. Methodology

2.1. Material

Chemicals used in this research included NaCl (100%, IBI Scientific), hospital wastewater, distilled water, and iron electrode plate E 6013 (TianQiao, China). Electrolyte solution was made of the NaCl. Then, the electrolyte solution was put into a batch reactor and iron plates was installed vertically. It was set with power supply at maximum 12 Volt for maximum 45 minutes. The wastewater was analyzed before and after the treatment.

2.2. Wastewater Sampling

In Aceh Province, Indonesia, there were 69 hospitals. Out of these, 22 were private and the rest were public. To study wastewater TSS, it was necessary to choose major hospitals with a range of services, plenty of instrumentation, and large of patients. For this investigation, hospitals having 200 beds were considered as major. Twelve public hospitals in Aceh met these criteria. Then, two hospitals were selected randomly making a size of 16.7%. At least ten percent samples was considered as a good sample for small population. The hospitals included Sigli General Hospital having 274 beds and Banda Aceh Meuraxa Hospital with 205 beds. There were some wards in the hospitals. Each ward resulted in wastewater having various characteristics. All wastewaters joined at a disposal tank and were homogenized.

2.3. Electrocoagulation Procedure

To take a sample, the wastewater was collected from the tank. The HWW 150 ml was taken for initial analysis of TSS. A sample 1.5 liters was poured into a reactor, the sodium chloride of 11.3 ml was added and stirred using a magnetic stirrer. The iron plate was arranged into four batch reactors. The cable equipment was connected to DC power supply in parallel. The electrical voltage was set 12 Volts, and the stirrer rate of two rpm kept the feed concentration homogeneous. A stopwatch was turned on for 45 minutes during the operating time.

After finishing the investigation, the reactor of the treated liquid waste was let for 30 minutes to precipitate impurities in the liquid waste. The TSS in the wastewater was analyzed by gravimetry (glass-fiber filter VWR brand grade 161). This investigation was repeated in the same way using liquid waste electrodes of Fe-Fe at the same precipitation time with various contact times

(15, 30, and 45 minutes), voltages (6, 9, and 12 Volt), and solution concentrations (0.0; 0.5; and 1.0 M).

2.4. Data Analysis

Furthermore, the data of the research result were processed using Response Surface Methodology (RSM) with Design Expert 10.0.1 software through BBD. The data processing was carried out using RSM with BBD. The method aims to reduce the number of investigations carried out in this research, to investigate the effect of the independent variable on the response, and to optimize the response. The RSM was a mathematical model processed using the optimum design theory. An initial step of the investigation was to determine both factors and response observed in the investigation.

The analyzed response was the effect of the contact time, voltage, and electrolyte solutions on the reduction percentage of TSS with the level ranges of high (+), low (-) and moderate (0) as appeared in Table 1. After determining independent variables, furthermore BBD offered the rule of variable interaction of 17 investigations randomly with five center points as observed in Table 2. The mathematical equation used was determined based on Analysis of Variance (ANOVA) and model (in this case the significant result of the significance or insignificance test of the model). Besides, it was to predict data accuracy obtained. The BBD was one of the optimization processes in RSM where its investigation design had a combination of 2^k designs with incomplete block design by adding a centre run in its design.

Table 1 Level of Box – Behnken design with parameters for TSS

Variable	Parameter	Low	Medium	High
A	Contact time (minute)	15	30	45
B	Voltage (Volt)	6	9	12
C	Electrolyte	0.0	0.5	1.0

3. Results and Discussions

3.1. Wastewater Characteristic

Wastewater used in this research is from the hospital wastewater. Hospitals investigated contained no wastewater plant. The results of parameters are presented in Table 2.

Table 2. Physicochemical characterisations of wastewater

Parameters	Environmental Ministry Regulation	Hospital Wastewater
pH	6-9	6.5
BOD (ppm)	50	50
COD (ppm)	80	108
TDS (ppm)	2000	3020
TSS (ppm)	30	37
Oil and fat (ppm)	10	10
Ammonia nitrogen (ppm)	10	10
Temperature (°C)	38	35.2

The initial pH is 6.5 (neutral). This value is within the permissible value of Decree of the Environmental Minister. COD, TDS, and TSS values are 108 ppm, 3,020 ppm, and 37 ppm, respectively. These characteristics in the HWW are far from the standard of wastewater from health care facilities in Indonesia (Decree of the Environmental Minister, 2014). The highest concentration of TDS and TSS are from Sigli General Hospital. BOD and oil (fat) are 50 ppm and 10 ppm, respectively. These values are within the permissible values of Decree of the Environmental Minister.

Amonia nitrogen is 10 ppm as in Decree of the Environmental Minister. The TSS after electrocogulation is 10.193–26.976 ppm

(Table 3) and it fulfills standard of Ministry Regulation No. 5 in 2014 (Decree of the Environmental Minister, 2014). Here, the experimental and predicted TSS reduction appears in Table 3. A higher electrolyte concentration (1 M) gives higher levels of electrical conduction to help the colloidal floc adhere to the electrode. Reaction in both combined electrodes of Fe-Fe is $2\text{H}_2\text{O} + \text{Fe} \rightarrow \text{H}_2 + 2\text{Fe}(\text{OH})_2$.

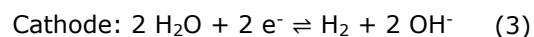
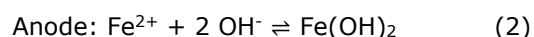
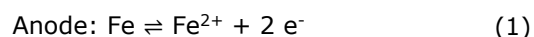
Besides, at a certain time, the electrocoagulation temperature of sample increases. It makes suspended solids collide actively and break before the floc formation to cause relatively large TSS (the removal decreases). Both of the reductions are better. Sodium chloride could make the electrocoagulation inhibited in degrading the HWW. The solution as supporting electrolyte will be decomposed into sodium and chloride ions. As a reaction occurs in the iron anode, it results in iron ion to bind chloride ion and forms FeCl_2 .

The other cause is the electrocoagulation principle based on electrolysis cell. Every electrolysis cell consists of two electrodes, the anode and the cathode. In the cell, the anode functions as a coagulant in a coagulation-flocculation process. At the cathode, the reaction produces hydrogen gas to lift flocs that do not go down.

Table 3. Data of experimental and prediction models on the percentage of TSS reduction

Run	Contact Time (minute)	Voltage (Volt)	Electrolyte (M)	Experiment TSS (%)	Prediction TSS (%)	Percentage of Error (%)
1	45.00	12.00	0.50	72.45	71.79	0.92
2	45.00	6.00	0.50	67.98	69.29	1.92
3	15.00	9.00	1.00	43.11	43.15	0.09
4	30.00	6.00	1.00	55.56	54.86	1.26
5	15.00	12.00	0.50	54.63	53.33	2.39
6	30.00	9.00	0.50	63.89	62.60	2.02
7	30.00	12.00	0.00	66.33	67.03	1.06
8	45.00	9.00	1.00	71.62	71.02	0.84
9	15.00	6.00	0.50	27.09	27.76	2.45
10	30.00	9.00	0.50	61.87	62.60	1.18
11	30.00	9.00	0.50	60.56	62.60	3.37
12	45.00	9.00	0.00	71.32	71.28	0.05
13	30.00	9.00	0.50	64.56	62.60	1.04
14	30.00	9.00	0.50	62.11	62.60	0.79
15	30.00	6.00	0.00	53.56	52.30	2.37
16	15.00	9.00	0.00	38.56	39.16	1.56
17	30.00	12.00	1.00	66.92	68.19	1.89

In combined Fe-Fe plates, the simplicity of the reduction-oxidation mechanism is as follows.



Hospitals are among the main locations in terms of discharged characteristics and volume. Medicinal treatment contributes to the total effluent treatment with a rather hard task. Because of their color content, hospital effluents, in particular, may interfere with the bodies of water to disturb biological process. HWW contains various toxic chemicals and salts to create an ecotoxicological impact in water bodies. HWW is treated conventionally through different chemical methods. Biological treatment process is ineffective in removing dyes with structured and branched macromolecules. Physicochemical treatment, such as coagulation-flocculation, adsorption, reverse osmosis, and ultrafiltration is used for dye removal. Nevertheless, these processes have limited applicability in the decolorization. Therefore, electrocoagulation is proposed to treat the HWW (Kabdash 2012). Many researchers investigate the electrocoagulation of prepared samples containing dyes and sodium chloride as the electrolyte. In this research, color caused by dyes can be removed effectively during electrocoagulation using iron electrodes.

3.2. Statistical Model of TSS

A BBD with RSM can determine the effect of contact time, voltage, and sodium chloride concentration on the efficiency of the TSS reduction. RSM eases to determine the statistical model. It proposes a model to explain their correlation using the interaction effect and adjusting the reduction efficiency at various conditions. The tendency to change every condition reduction is investigated using the RSM. The optimum condition is significant for technical implementation. Based on the analysis, quadratic model indicates conformity with the result. The model conformity depends on the value of determination coefficient (R-Squared) near 1.00 and the small deviation standard.

It means the model conformity used is appropriate because the R-Squared and adjusted-R² are higher than coefficients of a

cubical model. The value shows a variance/total variance ratio detected. Cubical and linear models are not appropriate for the research result. A good model accuracy has R-Squared above 0.8. It means that the response model in this research can describe well the reduction. The model validation provides a mathematical model obtained to predict the percentage value of TSS reduction in the HWW treatment by electrocoagulation indicated in Equation 5. It results in a relationship between TSS reduction and three variables selected.

$$\begin{aligned} \text{TSS} = & - 62.65550 + 3.75757 \times A \\ & + 8.92217 \times B + 11.00600 \times C \\ & - 0.025557 \times A^2 - 0.14558 \times B^2 \quad (5) \\ & - 2.780 \times C^2 - 0.12817 \times A \times B \\ & - 0.14167 \times A \times C - 0.23500 \times B \times C \end{aligned}$$

The equation in terms of actual factors can be used to make predictions about the TSS for given levels of each factor. Here, the model accuracy also could be identified from R-Squared value of 0.9922 that states that 99.22% of the data could be represented by the model, and the rest 0.78% could not be represented. The largeness of the coefficients is an indicator for model uniformity. Other than each effect of individual major process parameter, the TSS is also affected by the interaction variables. The interaction between voltage and electrolyte concentration, parameter BC, is the most influential factor and the second largest interaction influencing the TSS.

Equation 5 shows that if parameter AC is increased by 1 coded unit, the TSS decreases by 0.235 units. This effect is lower than contact time and voltage. The interaction between contact time and electrolyte concentration, parameter AC, is a third significant factor and the second highest interaction influencing the TSS. Equation 5 shows that if parameter AC is increased by 1 code unit, the TSS decreases by 0.14167 units. This effect is also lower than contact time and voltage. The interpretation of Equation 5 is similar to the one discussed in (Kholisa, 2018).

3.3. Statistical Analysis

Table 3 indicates that the highest reduction of TSS obtained is 72.45% by electrocoagulation in run 1 at contact time of 45 minutes, voltage 12 Volts and electrolyte 0.5 M. However, the lowest percentage of TSS reduction is 27.09% that occurs in run 9 at contact time 15 minutes, voltage 6 Volts and electrolyte 0.5 M.

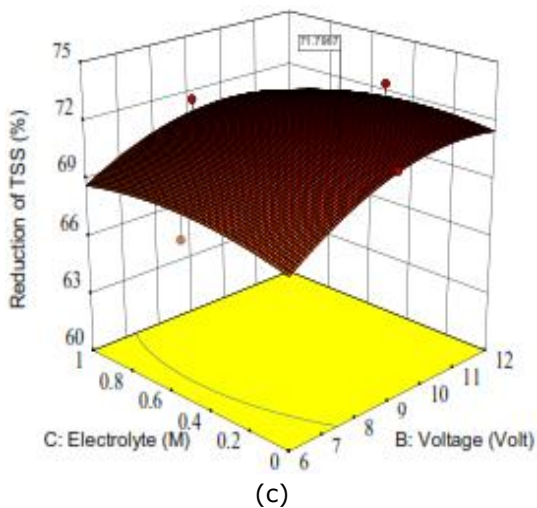
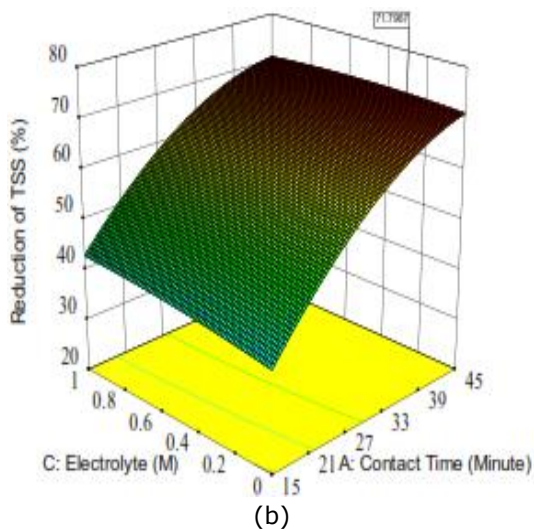
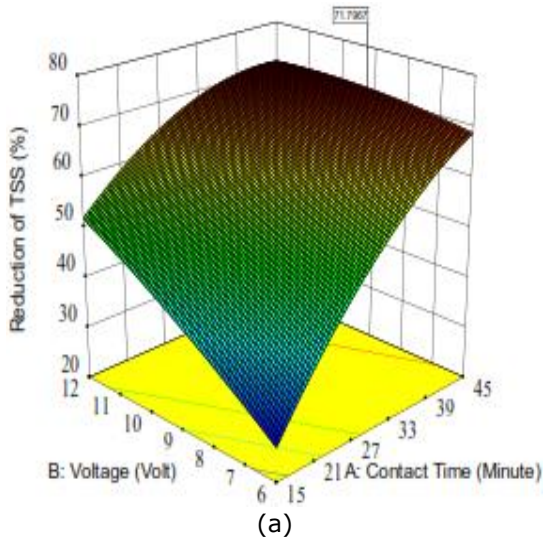


Figure 1. The effect of process variables on the percentage of TSS reduction: (a) contact time and voltage; (b) contact time and electrolyte concentration; (c) voltage and electrolyte of concentration.

It occurs because the contact time and voltage applied are relatively higher. Based on the result, as the contact time and the voltage applied is higher, the TSS removal is clearly also higher. The effect of process variable could be seen in the contour plot and three-dimensional graphs presented in Figure 1.

Figure 1 indicates the relationship between contact time and voltage on the percentage of TSS reduction. The graph shows that as the time and voltage are larger, the percentage of TSS reduction is larger. On the other hand, as the time and voltage are low, the percentage of TSS reduction is also lower. As mentioned by (Rusdianasari *et al.*, 2017), the longer contact time and the largeness of voltage will reduce the TSS value because more floc binding particles causes waste clearer. Figures of 1b and 1c indicate the relationship between contact time and electrolyte on the percentage of TSS reduction and the relationship between voltage and electrolyte on the percentage of TSS reduction. The three graphs show that as the contact time (15–45 minutes) and electrolyte concentration (NaCl 0–1 M) are higher, the percentage of TSS reduction increases more. However, the percentage increase of TSS reduction appears not too significant. Similarly, as the voltage and the electrolyte concentration are higher, the percentage of TSS reduction also increases more, but the result is not so significant.

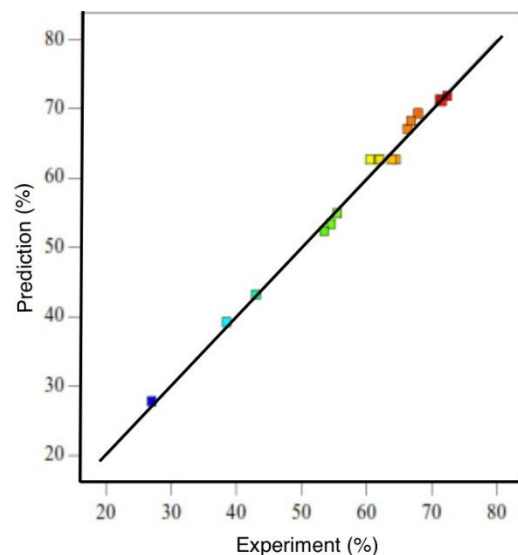


Figure 2. The relationship between the value of research result and the prediction value of TSS reduction.

Table 4. Analysis of variance on the effect of every variable on the percentage of TSS reduction

Source	Sum of Squares	DF	Mean Square	F Value	P-value Prob > F	Remark
Model	2493.57	9	277.06	98.65	< 0.0001	Significant
A – Time	1799.40	1	1799.40	640.69	< 0.0001	Significant
B – Voltage	393.96	1	393.96	140.27	< 0.0001	Significant
C – Elec. Conc.	6.92	1	6.92	2.46	0.1605	
A²	139.22	1	139.22	49.57	0.0002	Significant
B ²	7.23	1	7.23	2.57	0.1527	Not Significant
C ²	2.04	1	2.04	0.72	0.4228	Not Significant
AB	133.06	1	133.06	47.38	0.0002	Significant
AC	4.52	1	4.52	1.61	0.2453	Not Significant
BC	0.50	1	0.50	0.18	0.6866	Not Significant

Results of experimental and prediction TSS using the BBD appear in Figure 2. The average percentage of error obtained is 1.60%. It indicates that the model used is correct and could be relied on because the error percentage obtained has small value. It also appears from the graph obtained, the value is extremely near the linear line in Figure 2. It shows the relationship between the actual and predicted value of TSS reduction. The effect of independent variable could be observed in Table 4. The technique finds a model and the significance using Student's t-test. Analysis of Variance (ANOVA) shows the interaction significance between factors and response variables. It differentiates the average value of data groups by the variance comparison. The anova model used as provided by the program is the model with the highest level and producing the significant anova value. The anova model in this design is linear, quadratic, special cubic, and cubic. A model with significance in anova and non-significance in lack of fit is chosen to analyze variable (Nurmiah *et al.*, 2013). The model F-value of 98.65 implies the model is significant. There is only a 0.01% chance that this large F-value could occur because of noise. Table 4 of anova on the TSS reduction indicates the significant figure of A². It could determine a more significant regression coefficient using P-value. Values of "Prob > F" less than 0.05 indicate model terms are

significant. In this case, Time (A), voltage (B), A², and AB are significant model terms because their P-values less than 0.05.

Such variables are important, and they influence TSS reduction. A value of 0.05 is appropriate because most analysis refers to 0.05 as the limit for significance. As the value is less than the limit, a significant difference occurs. As it is larger than 0.05, an important difference does not occur. Time and voltage variables have the same effect on the TSS reduction. The interaction of contact time and voltage variables influences the percentage of TSS reduction; P-value 0.0002 indicates the value under 0.05, but the electrolyte concentration has P-value 0.1605. The value does not much influence the TSS reduction. Another interaction such as B², C², AC, and BC indicates the P-value above 0.05 that the interaction among variables does not influence the percentage of TSS reduction.

The first thing to analyze the result is to detect the model conformity. Based on Table 4, the model is said statistically significant. It can be applied to treat HWW to obtain optimum TSS reduction. This can be shown by the model P-value (< 0.0001) and the model F-value (98.65). P-value less than 0.05 shows the significant model. The P-value shows a good interaction and a significant test for individual variables. The P-value higher than 0.05 shows that the model is significant. A significant model is applicable in

wastewater treatment to get the optimum reduction (Murdani *et al.*). Based on Table 4, contact time and voltage appears significantly influence the TSS reduction percentage. TSS is the dry-weight of insoluble solids in a water sample trapped by a filter.

4. Conclusions

The research shows that electrocoagulation method was effective to reduce TSS in the hospital wastewater. It applied various process conditions. such as contact time 15, 30 and 45 minutes, voltages 6, 9, and 12 Volts, electrolyte concentrations 0.0; 0.5; and 1.0 M. As a longer time and a larger voltage were applied, the percentage of TSS reduction was 72.45% at the time 45 minutes and voltage 12 Volts. The percentage of TSS reduction was the highest. The electrolyte solution was less influential (not significant) on the TSS. Analysis of variance indicated that the interaction among variables influencing a response was dominated by contact time and voltage. In order to reduce TSS, contact time, voltage, and the interaction between contact time and voltage variables were more significant with P-value less than 0.05. Therefore, electrocoagulation using iron electrodes was applicable for TSS removal in hospital wastewater with the optimum condition. The resulted quadratic model can be used to obtain more appropriate value of contact time, voltage, and electrolyte concentration. Electrocoagulation with RSM method is also recommended to be further investigated using other types of wastewater, such as industrial wastewater.

Acknowledgments

Authors thank to Murdani, Sri Rahmiyati Rezeki, and Roro Sita Prameswari for their help and the support from Magister Grant of Syiah Kuala University in 2016 (Project No. 267/UN11.2/PP/SP3/2016).

References

- Alrhoun M., Carrion C., Casellas M., Dagot C. (2014) Hospital wastewater treatment by membrane bioreactor: performance and impact on the biomasses, *International Conference on Biological, Civil and Environmental Engineering*, Dubai, March 17–18, 95–101.
- Bazrafshan E., Mahvi A.H., Zazouli M.A. (2014) Textile wastewater treatment by electrocoagulation process using aluminum electrodes, *Iranian Journal of Health Sciences*, 2 (1), 16–29.
- Decree of Environmental Ministry (2014) *Environmental Ministry Regulation No 5 Baku Mutu Air Limbah (Standard of Wastewater Quality)*, Jakarta.
- Hansen K.M.S., Spiliotopoulou A., Chhetri R.K., Casas M.E., Bester K., Andersen H.R. (2016) Ozonation for source treatment of pharmaceuticals in hospital wastewater–ozone lifetime and required ozone dose, *Chemical Engineering Journal*, 290, 507–514.
- Kabdash, I., Arslan-Alaton, I., Olmez-Hanci, T., Tunay, O. (2012) Electrocoagulation applications for industrial wastewaters: a critical review, *Environmental Technology Reviews*, 1 (1), 2–45.
- Kholisa, B., Fester V.G., Haldenwang, R. (2018) Prediction of filtrate suspended solids and solids captured based on operating parameters for belt filter press, *Chemical Engineering Research and Design*, 134, 268–276.
- Lubis, M.R. (2019) Optimising protein and total dissolved solid to synthesize soy sauce from soybean residue using box-behken design, *Journal of Engineering Science and Technology*, 14 (1), 122–137.
- Lubis, M.R., Fujianti, D.F., Zahara, R., Darmadi (2019) The optimization of the electrocoagulation of palm oil mill effluent with a Box-Behnken Design, *International Journal of Technology*, 10 (1), 137–146.
- Murdani, Jakfar, Ekawati, D., Nadira, R., Darmadi (2017) Application of response surface methodology (RSM) for wastewater of hospital by using electrocoagulation, *IOP Conf. Series: Materials Science and Engineering*, 345, 1–7.
- Nurmiah S., Rizal S., Sukarno, Rosmawanty P., Budi N. (2013) Application of response surface methodology in the optimization of process conditions of alkali treated cottonic (ATC) processing, *IPB Kelautan dan Perikanan*, 8, 9–22.
- Pirkarami A. M., Ebrahim O. (2013) Removal of dye from industrial wastewater with an emphasis on improving economic efficiency and degradation mechanism, *Journal of Saudi Chemical Society*, 12, 1–8.

- Rahmi, Fachruddin, S., Nurmalasari (2018) Pemanfaatan limbah serat sago (*Metroxylon sago*) sebagai adsorben iodin, *Jurnal Rekayasa Kimia dan Lingkungan*, 13 (1), 70–77.
- Rudin, S. N. F. M., Shabri H. A., Muis Z. A., Hashim H., Ho W. S. (2018) Value-added waste potential of wastewater sludge from pharmaceutical industry: a review, *Chemical Engineering Transactions*, 63, 493–498.
- Rusdianasari, Taqwa A., Jaksen, Syakdani A. (2017) Treatment optimization of electrocoagulation (EC) in purifying palm oil mill effluents (POMEs), *J. Engineering and Technological Science*, 49 (5), 604–617.
- Sabeen A.H., Ngadi N., Noor Z.Z., Raheem A.B., Agouillal F., Mohammed A.A., Abdulkarim B.I. (2018) Characteristics of the effluent wastewater in sewage treatment plants of Malaysian urban areas, *Chemical Engineering Transactions*, 63, 691–696.
- Sharma A.K. and Chopra A.K. (2014) Influence of operating conditions on the electrolytic treatment for the removal of color, TSS, hardness and alkalinity using Al-Al electrode combination, *Journal of Applied and Natural Science*, 6 (1), 279–285.