Jurnal Kejuruteraan SI 1(6) 2018: 81-87
http://dx.doi.org/10.17576/jkukm-2018-si1(6)-10

Comparative Study for Lake Water Remediation: Chemical Coagulation and Electrocoagulation

(Kajian Perbandingan Pemulihan Air Tasik: Pengentalan Kimia dan Elektrokoagulasi)

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ABSTRACT

Water scarcity worldwide has reached the alarming rate and received the greatest attention. The increasing of pressure attributed by world water demand had inspired researchers on use of alternative water sources such as wastewater, a new norm. However, it is a critical need to find out the proper treatment technique to treat the wastewater. In this study, the reclamation and reuse of lake water (wastewater source) was conducted through chemical coagulation and electrocoagulation processes. Aluminium sulphate $Al_2(SO_4)_3$ was utilized in this study as the coagulant for chemical coagulation process. The dosage of $Al_2(SO_4)_3$ was manipulated in the range of 2 mg/L – 8 mg/L to investigate the optimal $Al_2(SO_4)_3$ dosage which brings to greatest removal of chemical oxygen demand (COD), colour, and turbidity from the collected wastewater sample. Whereas, electrocoagulation process was conducted in a lab-scale electrocoagulation reactor with two parallel monopolar aluminium electrodes. The electrocoagulation operation duration and DC power supply to the electrocoagulation reactor were varied in studying the performance of electrocoagulation process. Results obtained from this study depicted that both chemical coagulation and electrocoagulation processes were attractive for the treatment of lake water. However, chemical coagulation presented greater removal efficiency compared to electrocoagulation where it managed to remove COD, colour, and turbidity up to 93.04%, 93.86%, and 93.98%, respectively at the optimal $Al_2(SO_4)_3$ dosage of 6 mg/L. Sweep coagulation which comprises the adsorption and entrapment of pollutant particles onto aluminium hydroxides was the major mechanism for flocs formation in chemical coagulation process. Besides, the percentage of removal for electrocoagulation process is due to the applied voltage and operation duration. The percentage removal of COD, colour, and turbidity were increased with the increment of DC power supply voltage and pro-longed the operation duration. Results shown that the highest percent of removal is achieved by electrocoagulation reactor operated at 10 V for 45 min in this study, where the percentage of removal for COD, colour, and turbidity was 73.91%, 92.98%, and 71.44%, respectively.

Keywords: Lake water; wastewater remediation; chemical coagulation; electrocoagulation; comparative study

ABSTRAK

Kekurangan air di seluruh dunia telah mencapai kadar yang membimbangkan dan mendapat perhatian yang paling besar. Tekanan terhadap permintaan air global menggunakan sumber air yang tidak konvensional seperti air kumbahan, norma baru. Walau bagaimanapun, adalah keperluan kritikal untuk mengetahui teknik rawatan yang sempurna untuk merawat air kumbahan. Dalam kajian ini, pemulihan air tasik (sumber air kumbahan) dijalankan menggunakan proses pengentalan kimia dan proses elektrokoagulasi. Aluminium sulfat $Al_2(SO_4)_3$ digunakan sebagai koagulan untuk proses pembekuan kimia dalam kajian ini. Dos $Al_2(SO_4)_3$ yang berbeza digunakan dalam julat 2 mg/L – 8 mg/L untuk mendapatkan dos optimum yang menghasilkan kadar penyingkiran tertinggi keperluan oksigen kimia (KOK), warna, dan kekeruhan dari sampel air sisa terkumpul. Manakala proses elektrokoagulan dilakukan dalam reaktor elektrokoagulasi berskala makmal dengan dua elektrod aluminium monopolar selari. Tempoh operasi elektrokoagulasi dan bekalan kuasa DC ke reaktor elektrokoagulasi berubah-ubah dalam mengkaji prestasi proses elektrokoagulasi. Keputusan menunjukkan bahawa kedua-dua proses rawatan yang dikaji adalah berkesan dalam merawat air tasik. Walau bagaimanapun, pengentalan kimia menunjukkan kecekapan penyingkiran yang lebih efektif berbanding dengan elektrokoagulasi di mana penyingkiran COD, warna, dan kekeruhan sampel air sisa mencapai 93.04%, 93.86%, dan 93.98%, pada dos $Al_2(SO_4)_3$ optimum 6 mg/L. Pembekuan menyapu yang melibatkan penjerapan dan perangkap zarah pencemar ke aluminium hidroksida merupakan mekanisme utama untuk pembentukan flocs dalam proses pembekuan kimia ini. Selain itu, difahami bahawa kecekapan penyingkiran proses electrocoagulation mempunyai hubungan langsung dengan voltan dan tempoh operasi yang digunakan. Peratusan penyingkiran COD, warna, dan kekeruhan meningkat dengan kenaikan voltan bekalan kuasa DC dan memanjangkan tempoh operasi. Didapati bahawa kecekapan penyingkiran memuncak untuk reaktor elektrokoagulasi yang dikendalikan pada 10 V selama 45 minit dalam kajian ini, di mana peratus penyingkiran COD, warna, dan kekeruhan adalah 73.91%, 92.98%, dan 71.44%.

Kata kunci: Air tasik; pemulihan air kumbahan; pengentelan kimia; elektrokoagulasi; kajian perbandingan

INTRODUCTION

In recent decades, the general population of the world has been subjected to a number of environmental issues such as global warming, natural disasters, pollution, etc which are triggered by human activities. Among these issues, a global water crisis known as "water scarcity" has gaining the most attention.

Water scarcity is a wide definition of water-related issues such as water stress, water shortage, and a prevailing water crisis (EEA 2012). Although more than 70% of earth's surface consists of water, however majority of this water is not safe for our consumption. There is only around 3% of it is usable and most of it is difficult to reach. According to United Nations, as of December 2013, 783 million people are not able to get the freshwater. This is an alarming rate and is not an ideal amount of water to supply to global demand of 6.8 billion people (Hodgson 2016).

Water scarcity affects almost all aspects of day-to-day life from regular consumption, agriculture, education, and in some worse cases it is even the cause of death. However, not only day-to-day life water disrupted. The local economy also felt the strike by the lack of clean water. At larger perspective, 6 to 8 million people die annually worldwide is due to water-related diseases (The Water Project Organization 2014; UN Water Organization 2013). The negative health-related factors due to water scarcity include (although not limited to) dehydration, exposure to pathogenic microbes, diarrhoea schistosomiasis, trachoma, and intestinal helminths (Tarrass & Benjelloun 2012).

The increasing of pressure attributed by world water demand had inspired researchers on use of alternative water sources such as wastewater a new norm. However, highly polluted wastewater is not usable for human consumption. Moreover, discharge of wastewater into the waterbody will provide adverse impact on water quality. The aquatic life in water body will experience oxygen depletion and died whenever the water body is being continuously polluted. Therefore, there is a critical need to find out the proper treatment technique to treat the wastewater. Chemical coagulation has been conventionally applied to remove the natural organic matter (NOM), colour, and turbidity in water and wastewater (Renault et al. 2009; Wei et al. 2009). Conversely, many studies have reported the potential of electrocoagulation process in treating a variety of wastewater. However, each study started with different source of wastewater with different concentration of pollutants. Therefore, it is hard to compare and justify the effectiveness of these processes. In present work, the potential of chemical coagulation and electrocoagulation process for lake water (source of wastewater) remediation was explored. The effectiveness of each treatment was evaluated based on the parameter of chemical oxygen demand (COD), colour, and turbidity.

ELECTOCOAGULATION MECHANISM

Electrocoagulation involves the use of electricity, applied across the electrodes to generate metal ions in the wastewater sample by scarifying anode followed by coagulation-flocculation of pollutants from the wastewater sample. The active metal cations which produced at anode will react with hydroxide ions which generated at cathode to form metal hydroxides during the electrocoagulation process. These metal hydroxides are act as coagulants, where the pollutant particles are tend to attach on its surface and form larger aggregates which may either sink at the bottom of the electrocoagulation reactor or to be bring to the surface of water as flocs by the hydrogen bubbles generated at the cathode (Mitsika et al. 2013).

Aluminium is the most commonly used electrode material. During electrocoagulation process, Al^{3+} ions will generate in-situ in electrocoagulation reactor due to electrochemical dissolution of aluminium electrode at anode (Elnenay et al. 2017):

$$Al^{3+}(aq) + 3e^- \leftrightarrow Al(s) \quad (1)$$

Oxygen evolution is occurred at the anode (Elnenay et al. 2017):

$$4OH^- \rightarrow O_2 + 2H_2O + 4e^- \quad (2)$$

Simultaneously, the evolution of hydrogen is associated at cathode. The reaction occurs at cathode is dependents on the pH of wastewater sample. At neutral or alkaline pH, hydrogen is produced via Equation (3) (Elnenay et al. 2017):

$$2H_2O + 2e^- \rightarrow 2OH^-(aq) + H_2(g) \quad (3)$$

While under acidic condition, Equation (4) is used to explain the hydrogen evolution at cathode (Elnenay et al. 2017).

$$2H^+(aq) + 2e^- \rightarrow H_2(g) \quad (4)$$

Al^{3+} and OH^- ions generated by electrode reactions react to form different monomeric and polymeric species, which transform finally into $Al(OH)_3$ (s) depend upon total metal concentration and pH (Elnenay et al. 2017).

$$Al^{3+} + H_2O + e^- \rightarrow Al(OH)^{2+} + \frac{1}{2}H_2 \quad (5)$$

$$Al(OH)^{2+} + H_2O \rightarrow Al(OH)_2^+H^+ \quad (6)$$

$$Al(OH)_2^+ + H_2O \rightarrow Al(OH)_3 + H^+ \quad (7)$$

Then

$$Al^{3+}(aq) + 3H_2O \rightarrow Al(OH)_3(s) + 3H^+(aq) \quad (8)$$

Both aluminium cathode and anode may be chemically attached by OH^- ions in view of the amphoteric nature of aluminium (Elnenay et al. 2017).

$$2Al(s) + 6H_2O + 2(OH)(aq) \rightarrow 2Al(OH)_4^-(aq) + 3H_2(g) \quad (9)$$

METHODOLOGY

MATERIALS AND CHEMICALS

Aluminium sulphate, $Al_2(SO_4)_3$ used in this study was in analytical grade, purchased from Sigma Aldrich (Malaysia). Ultrapure (UP) water with a quality of 18 $M\Omega cm^{-1}$ was used to dissolve $Al_2(SO_4)_3$. Two aluminium plates in shape of rectangular with the dimension of 3 cm (width) × 10 cm (length) × 0.3 cm (thick) were employed as the anode and cathode for electrocoagulation process.

SAMPLE COLLECTION POINT

Wastewater in this study was collected from the catchment lake adjacent to Faculty of Engineering and Built Environment, University Kebangsaan Malaysia, Malaysia with GPS coordinate of 2.924325º, 101.770912º. The satellite map for the catchment lake is shown in Figure 1, while the characteristics of the wastewater sample are shown in Table 1.

TABLE 1. Characteristics of wastewater sample collected from the catchment lake adjacent to Faculty of Engineering and Built Environment, University Kebangsaan Malaysia, Malaysia

Parameter	Sample 1	Sample 2	Average
pH	7.01	6.77	6.89 ± 0.12
COD (mg/L)	72.0	43.0	57.5 ± 14.5
Colour (PtCo)	314	142	228 ± 86
Turbidity (NTU)	17.61	19.60	18.61 ± 1.00

FIGURE 1. Satellite map for catchment lake adjacent to Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia

CHEMICAL COAGULATION

Chemical coagulation process was carried out in a conventional jar test apparatus (Model ZR4-6, Zhongrun Water, China). Approximately 500 mL of wastewater was treated with chemical coagulation with the addition of $Al_2(SO_4)_3$ as the coagulant. The dosage of $Al_2(SO_4)_3$ was manipulated at the concentration of 2 mg/L - 8 mg/L to investigate the optimal dosage with greatest percentage of removal for chemical oxygen demand (COD), colour, and turbidity from the collected wastewater. The coagulation procedures consist of three steps: vigorous stirring after the addition of coagulant (120 rpm for 2 minute), mild stirring (20 rpm for 20 minutes), and settling (30 minutes).

ELECTROCOAGULATION

The electrocoagulation set-up for the experimental work in this study is shown in Figure 2. Electrocoagulation experiments were carried out in lab-scale electrocoagulation reactor with the total volume of 500 mL. Two parallel monopolar electrodes: one anode and one cathode with the total effective electrode area of 7.2 cm^3 were connected vertically to the lab-scale electrocoagulation reactor. The spacing between the electrodes was fixed at 1 cm. A digital DC power supply (Smart Power, 1 kW SMART Programmable DC Power Supplies, TSP5020, 0 ~ 50 v, 0 ~ 20 A) was employed to supply regulated electricity current to the electrocoagulation reactor. Prior each testing, the aluminium electrodes were rinsed with acetone to eliminate the surface impurities; it was then dipped into a mixture of 100 cm^3 HCl solution (5% v/v) and 200 cm^3 hexamethylenetetramine aqueous solution (2.80%) for 5 min and dried (Kobya et al. 2003). All testing were conducted at constant temperature (25ºC) and mixing speed (130 rpm). The voltage was regulated to the wanted value and the test was started. The variation of electrocoagulation operation duration and voltage in studying

the performance of electrocoagulation process was presented in Table 2. At the end of the test, the treated wastewater was allowed to stand for 30 min and the supernatant was collected for the analysis of pH, COD, colour, and turbidity.

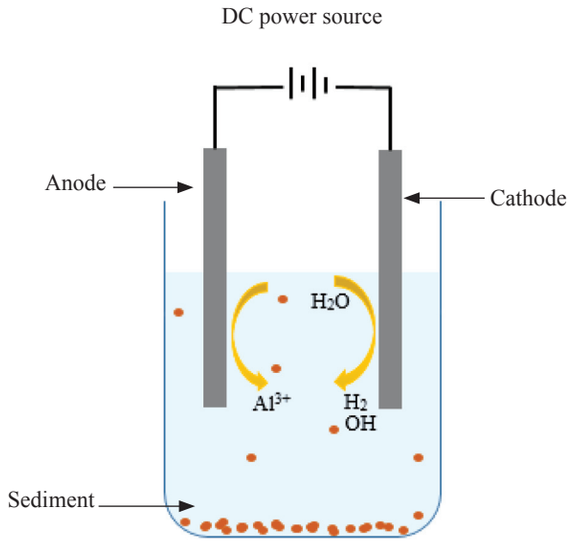

FIGURE 2. Experiment set-up for electrocoagulation process

TABLE 2. Electrocoagulation operation duration and voltage in studying the performance of electrocoagulation process

Duration (min)	Voltage (V)	Immersed depth of aluminium electrode (cm)
30	5	8
30	10	8
45	5	8
45	10	8

WATER SAMPLE ANALYSIS

Chemical and physical parameters including pH, COD, colour, and turbidity were measured to compare the efficiency of chemical coagulation and electrocoagulation in wastewater treatment. pH of the water sample was determined using a pH meter (Hanna Instruments Inc. USA). Whereas, COD and colour of the water sample were measured using a HACH DR3900 spectrophotometer med RFID-technology (HACH, USA) with standard program preloaded in the machine. Turbidity in water sample was measured using a Hach DR/2000 direct reading spectrophotometer (HACH USA).

RESULTS AND DISCUSSION

CHEMICAL COAGULATION

With the purpose of verify the effectiveness of chemical coagulation process and to gain the optimal $Al_2(SO_4)_3$ dosage for a favorable result, the pH, COD, colour, and turbidity of the treated wastewater underwent chemical coagulation at different $Al_2(SO_4)_3$ dosage were measured and the results were shown in Figure 3. Once the $Al_2(SO_4)_3$ had been injected into the wastewater sample in conventional jar test apparatus, small and fluffy flocs started to form. During the slow stirring process, these flocs collided with each other and entangled into larger flocs, as shown in Figure 4. After a series of experiments, the optimal $Al_2(SO_4)_3$ dosage was determined from the set of run that gave the best performance in greatly reduced the concentration of the studied parameters. As depicted by the results in Figure 3, the optimal $Al_2(SO_4)_3$ dosage was at 6 mg/L where the percentage removal of COD, colour, and turbidity was 93.04%, 93.86%, and 93.98%, respectively. This proved that chemical coagulation using $Al_2(SO_4)_3$ as coagulant can be a good wastewater treatment technique. However, when the $Al_2(SO_4)_3$ dosage was added beyond the optimal dosage, the concentration of the studied parameters were started to increase, suggesting an overdose of $Al_2(SO_4)_3$ in chemical coagulation process which provides antithetical effect in coagulation performance (Ang et al. 2015).

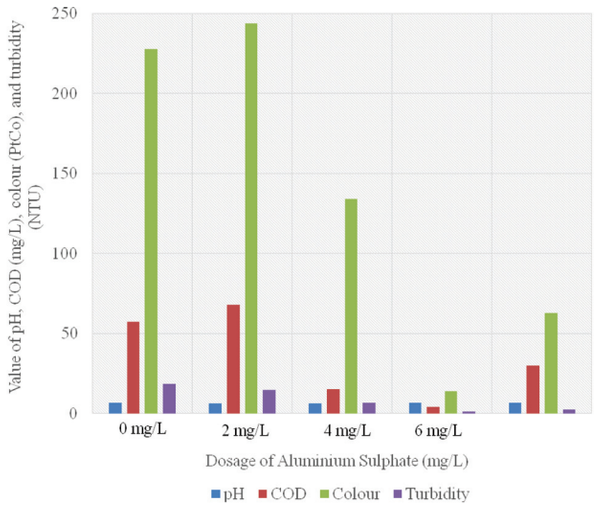

FIGURE 3. Characteristics of wastewater treated with chemical coagulation at different $Al_2(SO_4)_3$ dosage

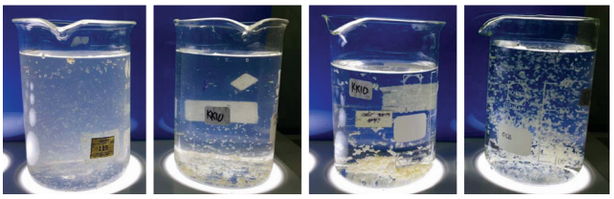

FIGURE 4. Wastewater treated with chemical coagulation in (a) 2 mg/L $Al_2(SO_4)_3$ (b) 4 mg/L $Al_2(SO_4)_3$ (c) 6 mg/L $Al_2(SO_4)_3$ and (d) 8 mg/L $Al_2(SO_4)_3$

Figure 5 shows the zeta potential for the wastewater samples treated with different $Al_2(SO_4)_3$ dosage. It was postulated that sweep coagulation was expected to take place in chemical coagulation process using $Al_2(SO_4)_3$ as coagulant. Chemical coagulation mechanism is generally divided into charge neutralization/electrostatic interaction

and sweep coagulation/co-precipitation (Ng et al. 2013). The difference of coagulation mechanism is highly depended on the types of coagulant used in that respective chemical coagulation process. For a charge neutralization coagulation mechanism, cationic coagulants will neutralized the negative charge pollutant particles due to the electrostatic shielding among the negatively charge pollutant particles through a binding action. This charge neutralization mechanism will consequently resulting in the reduction of particle's surface charge near to zero and precipitate (Wang et al. 2014; Zhong et al. 2011). Whereas, for sweep coagulation mechanism, the zeta potential value of the treated water sample will still retain at negative value even at optimum dosage. Since the zeta potential of the wastewater samples after treated with chemical coagulation process in this study was in the range of -11.3 mV to -32.5 mV, it designates that charge neutralization is not the predominant mechanism and it is attributed by the sweep coagulation. Similar conclusion has been drawn by Ng et al. (2013), Zhong et al. (2011), and Ang et al. (2015) where charge neutralization mechanism was eliminated due to the negative zeta potential value obtained. As an alternative, sweep coagulation which due to the adsorption and entrapment of pollutant particles onto aluminium hydroxides was the dominant mechanism for flocs formation in this coagulation process.

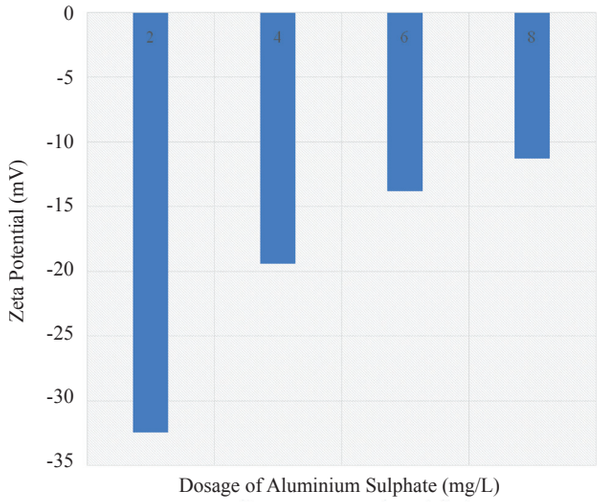

FIGURE 5. Zeta potential of chemical coagulation treated wastewater effluent at different $Al_2(SO_4)_3$ dosage

ELECTROCOAGULATION

Figure 6 depicted the photograph of wastewater treated with electrocoagulation process. The pollutant particles were attached to aluminium hydroxides's surface to form larger aggregates and moving to the surface of water as flocs by the produced hydrogen bubbles at cathode. Whereas, Figure 7 shows the characteristics of wastewater treated with electrocoagulation at different voltage and different operation duration. It was observed that the concentration of COD, colour, and turbidity was reduced after the experiment runs for electrocoagulation process. The concentration of these parameters was further decreased as the the DC power supply was increased from 5 V to 10 V for both operation duration (30 min and 45 min). This agreed well with the statement of Elnenay et al. (2017) saying that one of the important factors that affect the electrochemical process is the current density, not only for the coagulation dosage rate but also for the bubble production rate and the size and the growth of flocs which can influence the treatment efficiency of the electrocoagulation as well as strongly influencing both solution mixing and mass transfer at the electrodes. The great removal of these parameters at higher was due to the formation of more compounds like $Al_2(OH)_2^{4+}$, $Al(OH)^{2+}$, $Al(OH)_2^{+}$, $Al_{13}(OH)_{32}^{7+}$, $Al(OH)_3$ due to faster dissolution of anode at larger charge, which was then contributed to good coagulation effect (John et al. 2016). Additionally, higher number of hydrogen bubbles were observed at cathode as the DC power supply is increasing. These hydrogen bubbles had improved the mixing rate between aluminium hydroxides and pollutant particles which is then enhanced the flotation ability of the cell, consequently caused an increased in removal efficiency of the studied parameters (Guo et al. 2006). Besides, it was also observed that the number of hydrogen bubbles was increased and their size was decreased with the increasing of DC power supply. This mechanism was further support for fast removal of pollutant particles and sludge flotation (Golder et al. 2007). Apart from adsorption of pollutant particles on aluminium hydroxides and its compounds, Al^{3+} ions released from anode could also directly interact with the pollutant particles, which were then precipitates as insoluble salt (Phutdhawong et al. 2000).

For the factor of operation duration, the concentration of COD, colour, and turbidity concentration was found to be significantly reduced at longer operation duration (45 min). By prolonged the operation duration from 30 min to 45 min, the percentage removal of COD, colour, and turbidity was increased 50.00%, 40.58%, and 9.82%, respectively for the electrocoagulation reactor operated at 5 V. Whereas, the percentage removal of colour and turbidity of the wastewater sample underwent electrocoagulation at 10 V had increased 75.38% and 11.79%, respectively for longer operation duration of electrocoagulation process, except COD which experience a slight increase. This might possibly due to the small variation during the water sample analysis. The existence of higher amount of Al^{3+} ions (coagulants) in the wastewater sample at longer operation duration time was enhancing the efficiency of coagulation process. This agrees with previous reported results (El-Ashtoukhy et al. 2013; Elnenay et al. 2017; Errami et al. 2013).

Unlike other studied parameters, pH of the wastewater sample was experienced an increment after the electrocoagulation process. The final pH value of the wastewater sample was shown to be more alkaline with the increment of DC power supply from 5 V to 10 V and by prolonging the operation duration from 30 min to 45 min. This is possibly owing to the generation of Al^{3+} ions which was then affects the pH of the solution. During an electrocoagulation process which operated at higher DC power supply and longer

operation duration, higher amount of Al^{3+} ions will generated in-situ in electrocoagulation reactor due to electrochemical dissolution of aluminium electrode at anode, thus the pH value of wastewater sample was changed and shown to be more alkaline.

In overall, it was found that highest removal efficiency of COD, colour, and turbidity was obtained a high voltage and longer operation duration. The removal is maximum for a electrocoagulation reactor operated at 10 V for 45 min in this study, where the percentage of removal for COD, colour, and turbidity was 73.91%, 92.98%, and 71.44%, respectively.

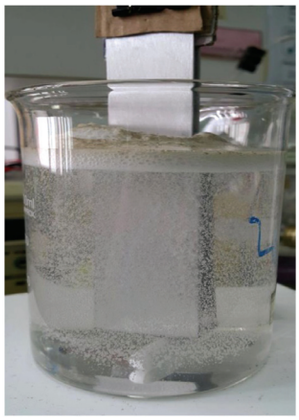

FIGURE 6. Wastewater treated with electrocoagulation process

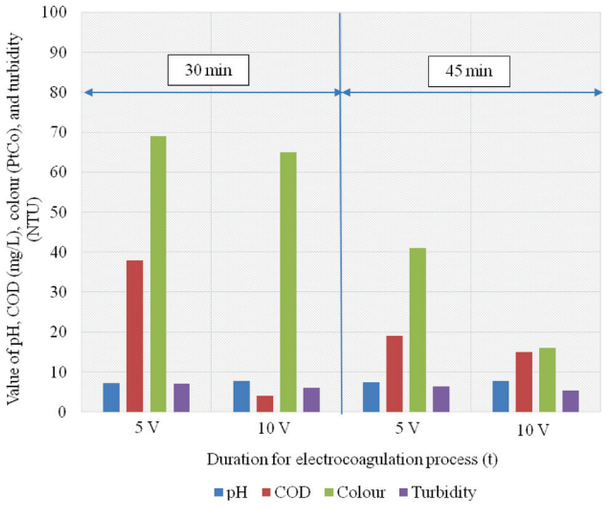

FIGURE 7. Characteristics of wastewater treated with electrocoagulation at different voltage and different operation duration

Figure 8 shows the variation of zeta potential value for electrocoagulation treated wastewater effluent at different voltage and different operation duration. During electrocoagulation, pollutant particles get physically attached to the aluminium hydroxide coagulants and form sludge. Since the formation of aluminium hydroxide compounds are greater at higher DC power supply and longer operation duration of electrocoagulation process due to faster dissolution of anode at larger charge and more dissolution of anode at longer period, it will then contributed to good coagulation effect where most of the pollutant particles were attached to the aluminium hydroxide and precipitated at the bottom of electrocoagulation reactor. This will then reduce the zeta potential value of the solution which corresponded to the presence of negatively charge pollutant particles.

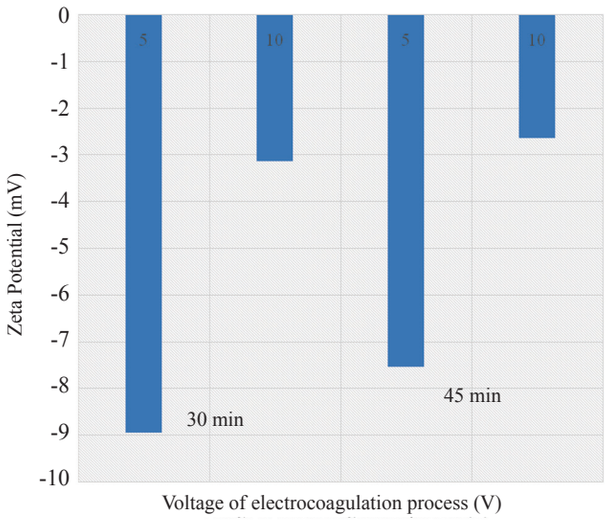

FIGURE 8. Zeta potential of electrocoagulation treated wastewater effluent at different voltage and different operation duration

CONCLUSION

This study was comparing the performance of chemical coagulation and electrocoagulation for lake water remediation. pH, COD, colour, turbidity, and zeta potential are the major analysis conducted for this study. From this work, it was found that chemical coagulation is an effective treatment process for lake water remediation compared to electrocoagulation where it manage to remove the COD, colour, and turbidity of the wastewater sample up to 93.04%, 93.86%, and 93.98%, respectively. Besides, the results obtained from this study presented that the percentage of removal of the parameters in electrocoagulation process is directly related to the applied voltage and operation time. The percentage removal of COD, colour, and turbidity were increased with the increment of DC power supply voltage and pro-longed the operation duration. Results shown the percentage of remoal is optimum for electrocoagulation reactor operated at 10 V for 45 min in this study, where the percentage of removal for COD, colour, and turbidity was 73.91%, 92.98%, and 71.44%, respectively.

ACKNOWLEDGEMENT

The authors wish to acknowledge Dana Penyelidikan Strategik (KRA-2017-016) and Geran Universiti Penyelidikan (GUP-2017-098) for the financial support of this research work.

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Received date: 20th March 2018
Accepted date: 31st August 2018
Online first date: 30th November 2018
Published date: 31st December 2018