

## The Combination of Coagulant and Electrocoagulation in Stabilized Leachate Treatment

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### Abstract

The contaminated leachate production is a result of solid waste disposal that has led to a negative impact on water and the environment. Therefore, leachate treatment is necessary to solve the problem. This study was conducted to determine the removal of color, suspended solids, chemical oxygen demand (COD), turbidity and ammonia nitrogen (NH<sub>3</sub>-N) for chemical coagulation alone under the influence of pH and dosage of alum, and also for combined coagulant+electrocoagulation (alum and electrocoagulation) under the effect of current density and the duration of current flow. The combination of coagulant and electrocoagulation performance in this experiment was compared with chemical coagulation. The results of this study revealed that the optimum dosage and pH for alum were 3 g/L and pH 6, respectively for chemical coagulation. The percentage removal of turbidity, color, suspended solids, COD and NH<sub>3</sub>-N by 3 g/L alum at pH 6 was 89.16%, 87.5%, 97.3%, 49.86% and 17.32%, respectively. Coagulant+electrocoagulation was conducted by using a combination of iron electrode plates (7 cm × 5 cm × 0.1 cm) and chemical coagulant (alum) in an electrolytic cell to determine the optimum current density and the duration of current flow. The result showed that the optimum coagulant dose, current density and duration of current flow in the coagulant+electrocoagulation were 3 g/L alum at pH 6 and 42.86 A/m<sup>2</sup> of current density at 30 minutes duration of current flow, respectively. The percentage removal of the turbidity, color, suspended solids, COD and NH<sub>3</sub>-N by using the combination method were 93.39%, 89.47%, 96.58%, 78.05% and 44.25%, respectively. Coagulant+electrocoagulation enhanced the removal efficiency of coagulation especially for COD and NH<sub>3</sub>-N compared to one stage coagulation. Thus, the combination of coagulant and electrocoagulation method has the potential to be used in the leachate treatment.

## 1. Introduction

Leachate is generated from precipitation, surface runoff, and the infiltration of groundwater into landfills [1]. Moreover, the factors that affect the production of leachate are the physical and chemical disposal reactions of wastes, the humidity rate of the waste on-site, and the increase in the underground water level [2]. Furthermore, leachate comprises a variety of complex substances and organic molecules such as fatty acids, humic substances, heavy metals and harmful chemicals [3]. Therefore, the discharge of leachate can lead to major environmental issues [1]. Generally, contaminants in leachate depend on solid waste composition that undergoes physical, chemical and biological activities within the landfill [4]. The characteristics of leachate are important in determining the suitable leachate treatment method [2]. Moreover, the characteristics of landfill leachate are affected by many factors, such as the age of the landfill, precipitation, weather variation, and the type of waste [5]. However, the age of the landfill is the main element that determines the overall content and characteristics of leachate [6].

Coagulation–flocculation is a physical and chemical method that is usually implemented in treating stabilized and older leachate [7]. Moreover, coagulation-flocculation is a good treatment for stabilized leachate [8]. The coagulation process involves the addition of a coagulant into the contaminated water under the intensity of rapid mixing [9]. The injection of chemical coagulant in the coagulation process destabilizes the colloidal particles, followed by the flocculation of unstable particles into bulky flocs, allowing the sludge to be settled more easily [4]. Moreover, several factors influence the efficiency of the coagulation-flocculation process, such as pH, coagulant dosage, mixing time and speed and retention time [10]. The advantages of physical and chemical methods in leachate treatments are mainly simplicity, low cost, good efficiency in removing pollutants and easy on-site implementation [11].

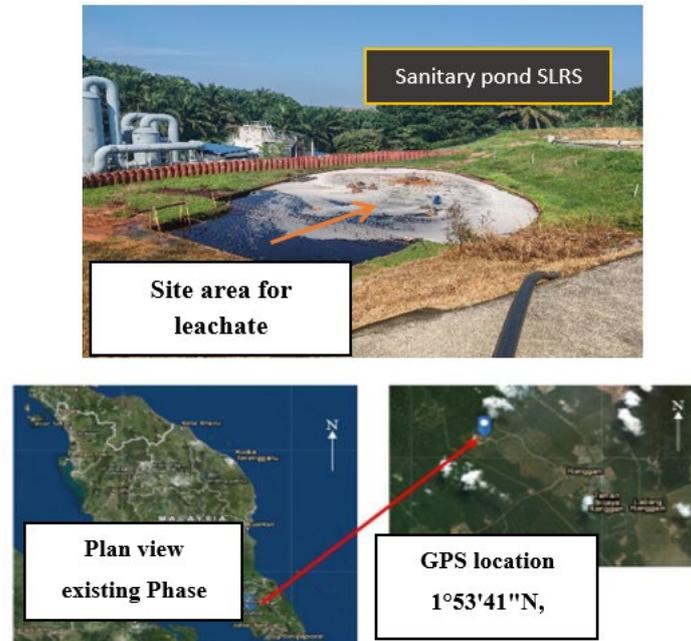
Electrocoagulation is an electrochemical method that is applied in both wastewater and surface water treatment [12]. Electrocoagulation is used in treating effluents that contain suspended solids, oil and grease, and organic and inorganic pollutants [13]. According to Ricordel and Djelal [14], electrocoagulation is efficient in removing a wide range of pollutants from water and wastewater. Moreover, the electrocoagulation process consists of flocs formation of metallic hydroxides in the effluent to be treated by using electro-dissolution of soluble anodes, which are aluminium (Al) or iron (Fe). Several factors that influence the performance of pollutant removals in electrocoagulation include the type of electrode used, the distance between electrodes, current density and reaction time [12]. Theoretically, electrocoagulation has an advantage in removing the smallest colloidal particles [15]. This is because the smallest charge particles are more likely to be coagulated as a result of an applied electric field that causes them to move faster [15]. The electrocoagulation process also gives advantages such as easy operation, simple equipment, and decreased amount of sludge [14]. Electrocoagulation is a rapid process compared to traditional chemical coagulation [16]. Moreover, the electrocoagulation process provides higher efficiency than chemical coagulation [2]. According to Ilhan *et al.* [17], the efficiency of COD removals obtained in electrocoagulation is 33% in 30 minutes of contact time with Fe electrodes, while chemical coagulation obtained around 22% in the same situation.

The conventional method of coagulation-flocculation that is often employed in the leachate treatment only helps to moderately remove COD [18]. Moreover, the usage of electricity in electrocoagulation may demand significant costs, and an impermeable oxide film that may form on the cathode can cause the electrocoagulation to be less efficient [19]. According to Can *et al.* [16], combined operations of electrocoagulation and chemical coagulant in textile wastewater reach the ultimate COD removal value of 65% in nearly 10 minutes by adding alum, whereas electrocoagulation takes at least 20 minutes to achieve the final COD removal value of 63%. Chemical coagulation gives an advantage in the formation of flocs in a leachate treatment. However, this method may require a high level of chemical coagulant consumption to treat leachate. In addition, the electrolysis process used in electrocoagulation has the ability to remove the smallest colloidal particles in leachate. Therefore, coagulant+electrocoagulation (chemical coagulation (alum) and electrocoagulation) were proposed to be used in this leachate treatment to investigate whether this method can increase the removal of suspended solids, turbidity, color, NH<sub>3</sub>-N and COD in the leachate treatment. This study was conducted to determine the removal of the turbidity, color, suspended solids, ammonia nitrogen (NH<sub>3</sub>-N) and chemical oxygen demand (COD) for chemical coagulation (alum) (under the influence of pH and dosage of alum) and for coagulant+electrocoagulation (under the effect of current density and duration of current flow). The performance of the coagulant+electrocoagulation in this experiment was compared with chemical coagulation.

## 2. Materials and Methodology

Leachate samples were taken from the Simpang Renggam landfill. This location is in the Kluang district of Johor, at latitude 1° 53'41.64" north and longitude 103° 22'34.68" (Fig. 1). The samples were collected, stored, and preserved following the APHA standard technique for the examination of water and wastewater [20]. The leachate samples were collected and preserved at the temperature of 4°C to prevent any changes to their characteristics

[20]. The leachate sample was taken out from the chiller and left at room temperature for around two hours before the experimental work was conducted.



**Fig. 1** Simpang Renggam landfill site location

Through this study, the characteristics of the raw leachate from Simpang Renggam landfill were determined, as shown in Table 1. Based on the value of pH, which is higher than 7.5 and COD, which is lower than 4000 mg/L, leachate at Simpang Renggam landfill was classified as an old landfill leachate. Therefore, the physical and chemical methods are suitable for leachate treatment at Simpang Renggam landfill.

**Table 1** Characteristic of raw leachate at Simpang Renggam landfill

Parameter	Range	*Average
Temperature (°C)	29-34	31
pH	8.66-8.69	8.68
Suspended solid (mg/L)	628-642	634
Turbidity (NTU)	210-370	347
NH <sub>3</sub> -N (mg/L)	1625-1628	1626
COD (mg/L)	2185-2190	2187
Color (Pt. Co)	9322-9344	9333

\*Three leachate samples were taken from February to April 2024

## 2.1 Chemical Coagulation (Alum)

Chemical coagulation was carried out by using a jar test. From this experimental work, the parameters of suspended solids, NH<sub>3</sub>-N, turbidity, COD and color were determined. Alum 3% (aluminium sulphate) was used in this experiment as a chemical coagulant. The pH of the leachate sample was altered by using 1N of sulphuric acid or sodium hydroxide before conducting a jar test. For this experiment, 0.5 liter of leachate samples were prepared in six different beakers. A chemical coagulant of alum with different dosages was added to each beaker (0, 2, 2.5, 3, 3.5 and 4 g/L). One beaker acted as a control sample.

During the process, the leachate sample was stirred rapidly at 200 rpm for 4 minutes [21]. For the next stage, the speed was reduced to 30 rpm for 15 minutes [21]. Then, the flocs were allowed to settle for around 30 minutes [21]. After the end of settling time, the leachate sample was collected around 3 cm from the surface of the

supernatant to measure the final suspended solids, turbidity, color, COD and  $\text{NH}_3\text{-N}$  of the sample. The experiment was repeated with varied pH value (pH 4, 5, 6, 7, 8 and 9).

## 2.2 Coagulant+electrocoagulation

Coagulant+electrocoagulation was conducted in an electrolytic cell that contained leachate sample (Fig. 2). In this experiment, chemical coagulation of alum was used along with electrocoagulation. Electrode plates of iron with the dimension of  $7\text{ cm} \times 5\text{ cm} \times 0.1\text{ cm}$  were used and connected as anode and cathode. The electrodes were dipped into the solution up to an active surface area of  $35\text{ cm}^2$ . The electrode plates were placed at a spacing of  $9.5\text{ cm}$  at the electrolytic cell and were connected to DC power supply. The pH of leachate sample was adjusted by using  $1\text{ N}$  of sulphuric acid or sodium hydroxide before conducting electrocoagulation.

During the process, optimum dosage of alum that was determined from chemical coagulation process was added into the electrolytic cell during rapid mixing. During slow mixing,  $40\text{ ml}$  of sodium chloride with the concentration of  $0.58\text{ g/L}$  was added to the leachate sample to enhance electrocoagulation process. In this experiment, the electrodes were dipped into the beaker that containing  $1\text{ litre}$  of leachate sample. The time and speed of rapid mixing and slow mixing in this experiment were set with an automatic controller. Then, the flocs were allowed to settle for around  $30\text{ minutes}$  [21]. After the end of settling time, leachate sample was collected around  $3\text{ cm}$  from the surface of leachate sample to measure the suspended solid, turbidity,  $\text{NH}_3\text{-N}$ , COD and color of the leachate sample. The experiment was repeated with varied duration of current flow ( $0, 5, 10, 15, 20, 25, 30, 35$  and  $40\text{ minutes}$ ) and current density ( $0, 5.71, 14.29, 42.86, 71.43$  and  $100\text{ A/m}^2$ ).

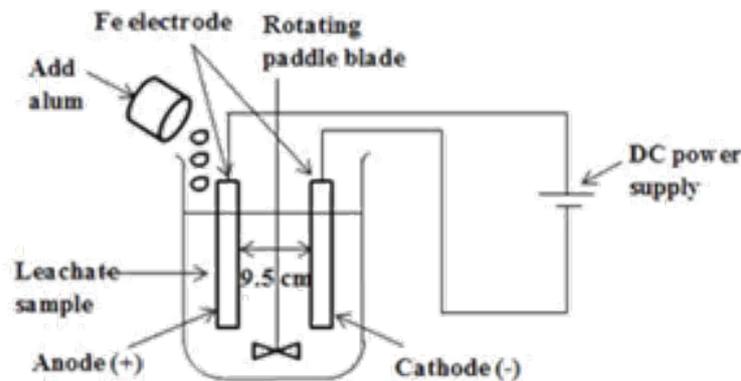


Fig. 2 Schematic diagram for coagulant+electrocoagulation

## 3. Result and Analysis

### 3.1 Optimum Dose by Chemical Coagulation

Fig. 3 below shows the percentage removal by alum under different dosages. Based on Figure 3, the dosage of alum at  $3\text{ g/L}$ ,  $3.5\text{ g/L}$  and  $4\text{ g/L}$  shows better removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  in contrast to other dosages of alum. The percentage removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  at  $3\text{ g/L}$  dosages of alum was  $85.71\%$ ,  $88.89\%$ ,  $96.34\%$ ,  $40.38\%$  and  $20.14\%$ , respectively. On the other hand, the percentage removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  at  $3.5\text{ g/L}$  dosage of alum was  $78.02\%$ ,  $83.33\%$ ,  $93.9\%$ ,  $34.04\%$  and  $15.33\%$ , respectively. Meanwhile, the removal percentage of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  at  $4\text{ g/L}$  does not show much difference compared to the dosage of alum at  $3\text{ g/L}$  and  $3.5\text{ g/L}$ , which was  $82.42\%$ ,  $72.22\%$ ,  $95.12\%$ ,  $25\%$  and  $16.48\%$ , respectively. However, the consumption of alum dosage at  $3.5\text{ g/L}$  and  $4\text{ g/L}$  was higher. Therefore,  $3\text{ g/L}$  was chosen as the optimum dosage of alum in this chemical coagulation since it was more economical. A sufficient coagulant dose is vital to ensure optimum performance of coagulation. Over dosage as shown in Fig. 3 beyond  $3\text{ mg/L}$  of alum resulted in charge reversal that contributed to disaggregation of floc and decreased pollutant removal [22]. A similar optimum dosage of alum was recorded by Omar *et al.* [23]. However, the optimum dosage of alum obtained by Marañón *et al.* [24] was  $0.8\text{ g/L}$  at pH 6, this is probably because every leachate has different characteristics, and the reaction between alum will be different.

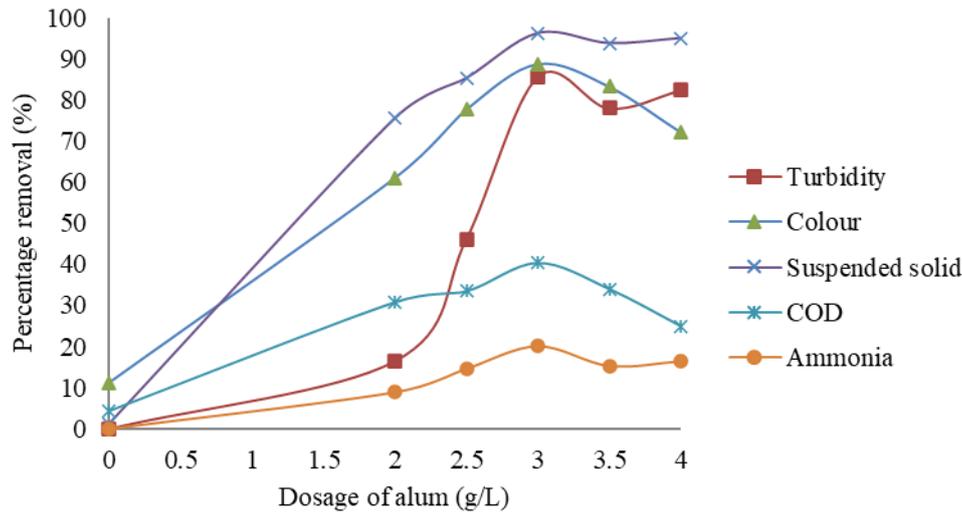


Fig. 3 Percentage removal by alum at pH 6

### 3.2 Optimum pH by Chemical Coagulation

Fig. 4 shows the percentage removal by alum of 3 g/L at different pH in the range of 4 to 9. In overall, better performance was recorded at acidic range compared to alkaline range. At right pH, the process can neutralize the pollutant charge and initiated coagulation. Based on Figure 4, the alum dosage of 3 g/L at pH 5 and pH 6 gives high removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  compared to other pH values. The percentage removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  at pH 5 was 79.52%, 81.25%, 95.95%, 44.85% and 17.04%, respectively. Meanwhile, the removal percentage of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  by alum at pH 6 was 89.16%, 87.5%, 97.3%, 49.86% and 17.32%, respectively. However, the consumption of reagents used in changing the pH value of the leachate sample to pH 5 was more than pH 6. Therefore, pH 6 was selected as the optimum pH since fewer reagents were used to alter the pH of the sample, and the removals were at par between both pHs. A similar optimum pH was recorded by Omar *et al.* [23]. Another study by Marañón *et al.* [24] showed that the optimum pH by using alum was also obtained at pH 6.

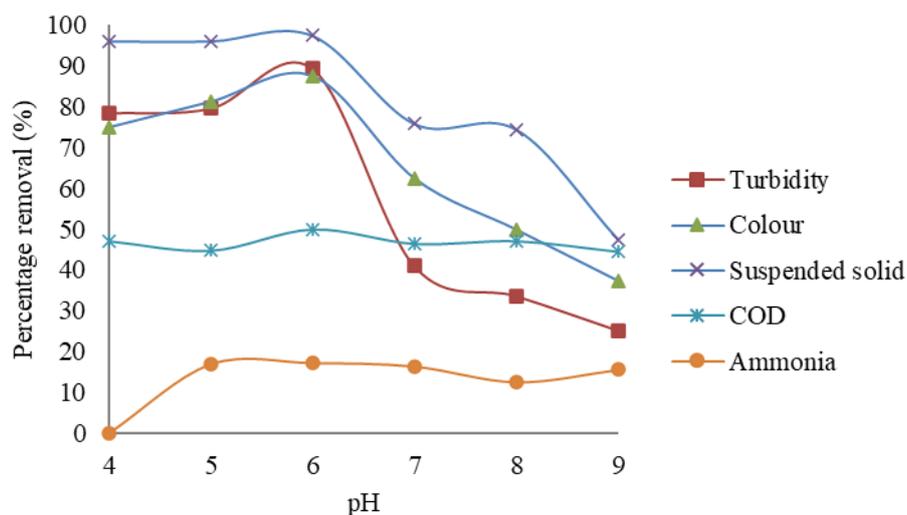


Fig. 4 Percentage removal by alum (3 g/L) at different pH

### 3.3 Optimum Duration of Current Flow by Coagulant+electrocoagulation

Fig. 5 showed the percentage removal of turbidity, color, suspended solid, COD and  $\text{NH}_3\text{-N}$  by using coagulant+electrocoagulation under the influence of duration of current flow. Based on Fig. 5, the duration of current flow at 30 minutes and 35 minutes showed good percentage removal of turbidity, color, suspended solid,

COD and  $\text{NH}_3\text{-N}$ . The percentage removal of turbidity, color, suspended solid, COD and  $\text{NH}_3\text{-N}$  at 30 minutes duration of current flow was 85.08%, 94.74%, 97.76%, 71.88% and 28.5%, respectively. Meanwhile, the percentage removal of turbidity, color, suspended solid, COD and  $\text{NH}_3\text{-N}$  at 35 minutes duration of current flow was 88.95%, 78.95%, 97.76%, 75% and 30.19%, respectively. The increment of duration of current flow leading to a better coagulation and flocculation of contaminants. However, beyond optimum value the removal decreased possibly resulted from the effects of electrode scaling, mass transfer limitations, and the dynamics of electrochemical reactions over time [25][26]. Furthermore, longer duration may increase the energy costs and electrode degradation. Therefore, 30 minutes was selected as the optimum duration of current flow since lesser time was used to give at par removal of 35 minutes. The similar optimum duration of current flow was also recorded by Ilhan *et al.* [17], and MoayeriKashani *et al.* [27].

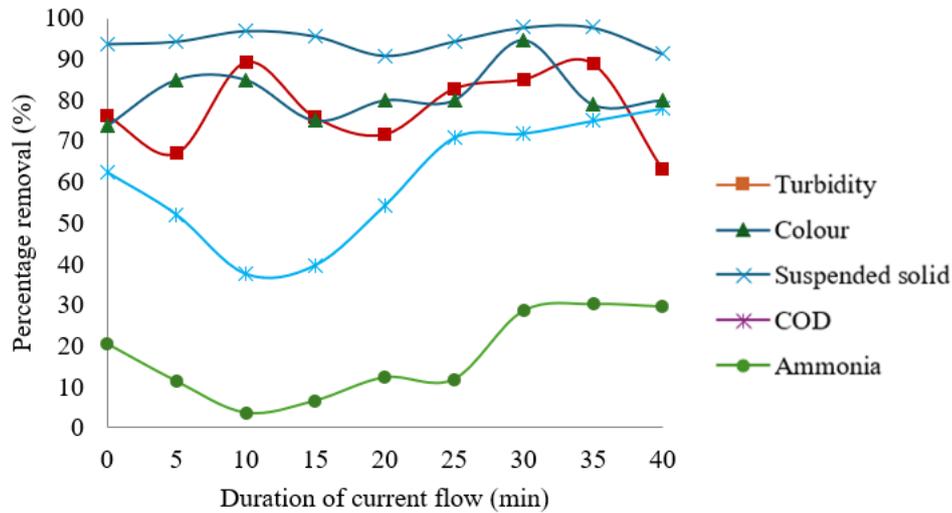
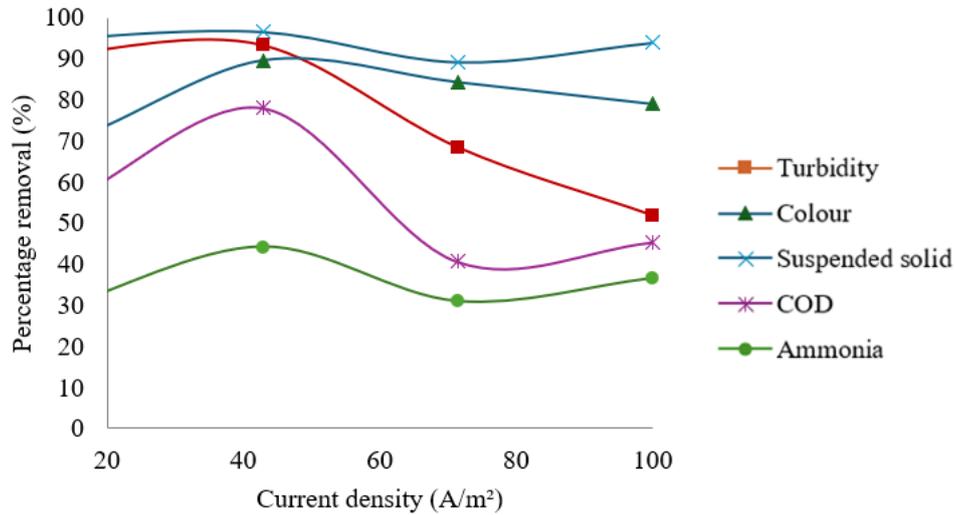


Fig.5 Percentage removal by two stages coagulation under the influence of duration of current flow

### 3.4 Optimum Current Density by Coagulant+electrocoagulation

Fig. 6 shows the percentage removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  by using coagulant+electrocoagulation under the influence of current density. Based on Figure 6, the current at  $42.86 \text{ A/m}^2$  showed good percentage removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  compared to other current densities. The percentage removal of turbidity, color, suspended solids, COD and  $\text{NH}_3\text{-N}$  at  $42.86 \text{ A/m}^2$  of current density was 93.39%, 89.47%, 96.58%, 78.05% and 44.25%, respectively. Therefore,  $42.86 \text{ A/m}^2$  was selected as the optimum current density since it gives good percentage removal. According to the previous study by Ilhan *et al.* [17], the obtained optimum current densities were at  $631 \text{ A/m}^2$ , at 30 minutes duration of current flow for single electrocoagulation. This is probably because single electrocoagulation requires more current density in removing pollutant compared to coagulant+electrocoagulation. From this study, proven that the addition of coagulant (alum) probably resulted in enhancement of floc structure and increasing the removal efficiency of electrocoagulation at less current density value [28].



**Fig. 6** Percentage removal by two stages coagulation under the influence of current density

#### 4. Conclusions

The results obtained from the optimization study confirm the removal ability enhancement of combined coagulant and electrocoagulation method. Coagulant+electrocoagulation has achieved 93.39%, 89.47%, 96.58%, 78.05% and 44.25%, respectively for turbidity, color, suspended solids, COD and NH<sub>3</sub>-N. As compared to chemical coagulation alone using alum the removal percentage of turbidity, color, suspended solids, COD and NH<sub>3</sub>-N were 89.16%, 87.5%, 97.3%, 49.86% and 17.32%, respectively. The data analysis showed that the application of coagulant+electrocoagulation significantly improved the removal of COD and NH<sub>3</sub>-N. However, the removals increment of turbidity, color and suspended solid values are not significant since the increased percentage is less than 5%. This suggest that combine coagulant and electrocoagulation were more successful in removing COD and NH<sub>3</sub>-N for leachate treatment than chemical coagulation alone.

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#### Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

#### Author Contribution

The authors confirm contribution to the paper as follows: **study conception and design:** Nur shaylinda and Nor Azliza; **data collection:** roseliz azman, **analysis and interpretation of results:** Laila Wahidah, Ain Natasya; **draft manuscript preparation:** Sitor Aishah. All authors reviewed the results and approved the final version of the manuscript.

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