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# Removal of Ammonia and Colour from Landfill Leachate using Integrated Electrocoagulation-Zeolite Adsorption Processes

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**Abstract**

The amount of waste generated in Malaysia had increased annually due to both economic and population accelerations. This resulted in an increase in leachate production that contains a high amount of ammonia nitrogen ($NH_3$-N) and colour, which is a severe problem for both environments as well as for human health. Therefore, this study aimed to determine the potential of Electrocoagulation (EC) – zeolite adsorption for removing $NH_3$-N and colour from landfill leachate. In the experimental works, two batch studies consist of batch EC using aluminium (Al) cylindrical electrode and batch adsorption study using clinoptilolite zeolites adsorbent were conducted using leachate sample collected from Pulau Burung Sanitary Landfill (PBSL). The batch EC experiment was carried out to determine the optimum removal of $NH_3$-N and colour by the effect of current density (7 - 35 mA/mm$^2$), initial pH (5 - 9), electrolysis time (0 - 90 minutes) and inter-electrode distance (5 - 25 mm). An integrated EC - zeolite adsorption was carried out under the optimum condition of 2.0 g/150 mL adsorbent dosage, contact time of 100 minutes and an initial pH value of 8. Based on the experiment, almost 50% of $NH_3$-N and 95% of colour were removed using a single EC under the optimum current density of 14.0 mA/mm$^2$, pH 5, electrolysis time of 75 minutes and 15 mm of inter-electrode distance. A comparison of colour removal between single EC and EC-Zeolite showed the same degradation performance. The interaction of hydrogen ion ($H^+$) with aluminium hydroxide ion (Al $(OH)_3$) produces during EC stabilises small particles forming larger sludge that reduces colour concentration. The removal of $NH_3$-N increased from 50.0% to 93% when EC was integrated with clinoptilolite zeolite adsorption. In conclusion, the results show that both processes can potentially remove colour while the integrated process effectively removes $NH_3$-N from landfill leachate. The proposed treatment helps optimise process performance while minimising the operational cost of the treatment. Thus, the proposed integrated EC – zeolite adsorption processes can be an alternative to landfill leachate treatment which aligns with the 12$^{th}$ Malaysia Plan to reduce pollution and carbon emissions to become a carbon neutral nation as early as 2050.

## 1. Introduction

Leachate is formed when the water carries organic and inorganic matter, heavy metals, impurities, and other harmful substances flowing from the waste into the solid ground [1]. The organic matter in leachate, such as ammonia nitrogen ($NH_3$-N) along with the colour can change the properties of groundwater and surrounding soils. In Malaysia, environmental regulations governing leachate treatment are primarily enforced by the Department of Environment (DOE) under the Ministry of Natural Resources and Environment. The regulations and guidelines included Environmental Quality Act 1974, Solid Waste and Public Cleansing Management Act 2007, Guidelines for the Design and Operation of Sanitary Landfills (2007) and National Environmental Policy (NEP). The regulatory system in Malaysia for leachate treatment is specifically designed to protect the environment and public health. It ensures that leachate is collected, treated, and disposed of in a proper manner, in accordance with the country's environmental regulations and guidelines. Leachate effluent acceptable conditions were governed by the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation 2009 (PU(A) 433) Second Schedule (Regulation 13), also known as the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation.

Leachate is treated by four different methods: physical treatment, chemical treatment, physicochemical treatment, and biochemical treatment and it needs to be treated before being discharged into the receiving rivers [1], [ 2]. There are many factors that affect the quality of leachate such as age, waste composition and the weather conditions of the landfill, but the most important factor influencing the quality and effectiveness of leachate treatment is age [3]. As landfill leachate ages from young to old, the organic concentration of COD decreases, and the $NH^3$-N concentration starts to increase. Additionally, the hydrolysis rate and fermentation of nitrogen-containing fractions of biodegradable refuse substrates lead to high $NH_3$-N concentrations in old leachate which require extensive treatment compared to young leachates [1], [4].

Malaysia's solid waste production is expected to rise from 292 kilograms per capita in 2000 to 511 kilograms per capita in 2025 [5]. This situation had increased the production of landfill leachate with a high amount of pollutants such as $NH_3$-N and colour. Several leachate characteristic studies indicate that $NH_3$-N constitutes as high as 1000 mg/L while for colour as high as 5000 Pt-Co [6]-[8]. This shows that the treated leachates regularly reach the permissible limit of leachate effluent based on the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation. Exposure to this contamination can lead to a danger to health as it is exposed to communities and the ecosystem such as acceleration of eutrophication processes caused by prolonged exposure to $NH_3$-N [9], [10]. Leachate frequently contains organic and inorganic compounds that are not easily treated using conventional methods, requiring the use of additional or advanced treatment systems. Elevated concentrations of ammonia can inhibit the activity of microorganisms in biological treatment processes, hence reducing treatment effectiveness. Due to the high concentration of organic and inorganic matters and impurities, natural treatments such as wetland are insufficient to treat this polluted leachate.

One of the major issues related to leachate treatment is developing an efficient system that can provide treatment for a large volume of leachate with lower operational costs. Different combinations of biological, physical, and chemical processes have been approached; however, most of the combinations require various methods that are both expensive and complicated [1]. As a result, cheaper technologies combining various methods have been introduced.

Currently, electrocoagulation (EC) process has widely been used as an alternative to conventional leachate treatment. This process consists of the arrangement of two electrodes (anode and cathode) where the cathode is oxidized while the water is reduced, resulting toward a better by-product treatment [11]. When direct current is applied, the sacrificial anode is dissolved to produce in situ coagulant while the cathode will undergo passivation [12]. In general, the EC process undergoes three stages which are the oxidation of sacrificial electrodes to form coagulant, destabilization, suspension and breaking of contaminants and the formation of flocs which are crucial for the EC process to be successfully performed [13]. In most previous studies, aluminium electrode performed best with percentage removal of $NH_3$-N and colour more than 60.0% [14]-[16]. In this study, aluminum was used as an electrode for batch EC study. However, EC also faces a major problem such as high consumption of electricity while treating the landfill as it will directly affect the operational cost [17]. Most of the EC were integrated to maximise the process performance while minimising the operational cost. However, existing EC processes were integrated with more expensive processes such as chemical precipitation, ozonation or ammonium stripping which resulted in higher operational costs [18].

As a solution to overcome these problems, an integrated treatment of EC with zeolite adsorption was proposed. The potential of integrating EC with clinoptilolite zeolite adsorption was observed in this study. Previous literature studies have used clinoptilolite zeolite as an adsorbent to remove $NH_3$-N [17], [18] and colour [19], [20] from wastewater at a rate as high as 90%. Removal occurs when these pollutants separate and bind on the surface of the absorbent from the effect of Van der Waals force.

In this study, the performance removal for a single EC and integrated EC – Zeolite adsorption was investigated to provide an alternative treatment for landfill leachate. The aims of this paper are to determine the optimum removal of $NH_3$-N and colour by considering the effects of current density, initial pH value, electrolysis time and inter-electrode distances; and to compare the performance removal between a single EC and integrated EC – Zeolite adsorption potential to remove $NH_3$-N and colour from a leachate sample.

## 2. Materials and Methods

This section consists of the leachate sample collection and characterization, analytical method, EC setup and integrated EC – Zeolite adsorption setup. The batch EC and integrated EC-Zeolite adsorption processes are discussed in Section 2.2 and Section 2.3, respectively.

### 2.1 Leachate Collection and Characterization

In this study, leachate samples were collected from Pulau Burung Sanitary Landfill (PBSL), Nibong Tebal, Penang twice a month for six (6) months. Then, the sample was collected in a 10 Litre HDPE bottle and stored immediately below 4°C in an ice box to avoid natural chemical and biological reactions occurring prior to laboratory analysis. The collection and preservation of the sample were followed in accordance with Standard Methods for the Examination of Water and Wastewater (1992). The initial concentration of raw pH, temperature, colour, salinity and conductivity were analysed on site by using portable analysis instruments (YSI ProODO multi-meter, YSI Trulab pH meter and Hash SensIon 5 conductivity meter) while $NH_3$-N and color were analysed in lab using a Spectrophotometer (DR6000). However, the collected samples were brought to the Environmental Laboratory, UiTM Cawangan Pulau Pinang to analysed for $NH_3$-N and colour. The characteristics of PBSL leachate were analysed in Table 1. The characterization study of leachate quality is important for protecting the environment, preserving public health, ensuring regulatory compliance, and improving waste management practices. The results showed that the leachate samples from PBSL exceeded the standard limit, which is the permissible limit for leachate effluent according to the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation. Section 3.1 provides a detailed discussion of the leachate sample characteristics from PBSL.

### 2.2 Batch Electrocoagulation

A batch EC study was conducted using a cylindrical pair of aluminum mesh electrodes with an anode electrode (30 mm diameter, 100 mm height) and a cathode electrode (50 mm diameter, 100 mm height). It was placed inside a 1000 mL beaker and connected to a 30V/10A DC power supply as shown in Fig. 1. The leachate sample was filled into a beaker for approximately 600 mL so that 50% of the electrodes were dipped into the sample. The batch study was placed on a magnetic stirrer and stirred for an optimum stirring speed of 150 rpm [21].

The study was conducted by evaluating current density from 7.0 mA/mm$^2$ to 35.0 mA/mm$^2$, initial pH from 5.0 to 9.0, electrolysis time from 30 minutes to 90 minutes, and inter-electrode distance from 5 to 25 mm. After 20 minutes of settling time, 100 mL of supernatant leachate was extracted from the beaker. After the EC process was completed, the sample was settled for 20 minutes for the flocs to form. The result from this experiment was used to analyse the optimum removal of $NH_3$-N and colour, as shown in Eqn. 1.

$$Removal\ (\%) = \left(\frac{C_i - C_o}{C_i}\right) \times 100\% \tag{1}$$

where $C_i$ and $C_o$ is the initial and final concentration of pollutant.

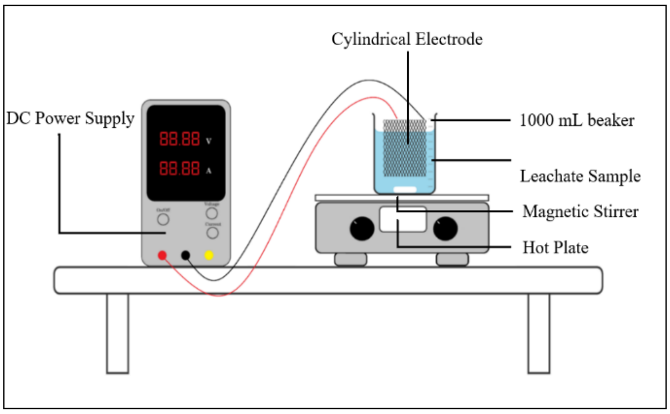



**Fig. 1** *Schematic diagram of batch electrocoagulation*

## 2.3 Batch Electrocoagulation – Zeolite Adsorption

The integrated EC – zeolites adsorption study was conducted to investigate the optimum percentage removal of targeted parameters by integrating both processes and was finally used to compare the performance removal of parameters between the integrated processes and the single EC process. Fig. 2 shows the configuration of the batch EC – zeolite adsorption study. For the EC process, the same optimisation process from Section 2.2 was used to treat the raw leachate sample and then the treated sample (from the EC process) was retreated again using the adsorption process. Clinoptilolite zeolites were used as the adsorbent media in this study. The adsorption study was conducted using an orbital shaker (Protech Model 719). The experiment was conducted based on an optimum dosage of 2.0 g in 150 mL of leachate sample in a conical flask with a contact time of 100 minutes and an initial pH value of 7 to 8 [22]. The selected range is based on previous studies [19], [22], [23]. In the experiment, pH was selected between pH 7 and pH 8 due to the characteristic of stabilised leachate being pH 7.5 and leachate discharge was also between pH 6 to pH 9. Then, after 20 minutes of settling process, the sample was drawn and analysed to determine the treated concentration of $NH_3$-N and colour using integrated treatment.

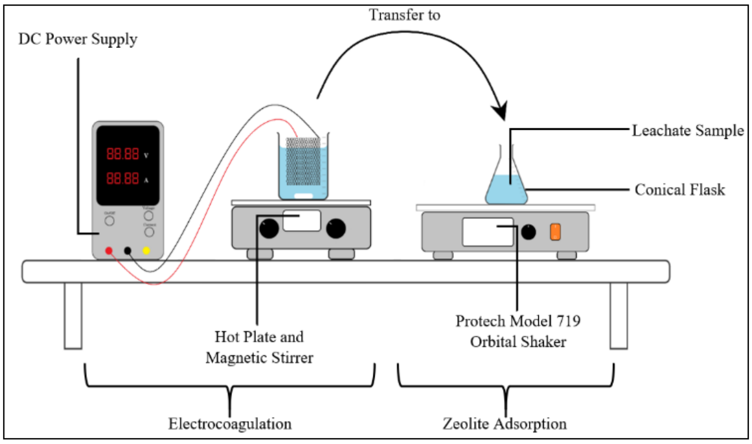



**Fig. 2** *Schematic diagram of integrated electrocoagulation – zeolite adsorption configuration*

## 3. Results and Discussion

In this study, the results and analysis are divided into three (3) sections. Section 3.1 discussed the leachate characteristics of PBSL, Section 3.2 discussed the performance of the electrocoagulation process and Section 3.3 investigated the performance of integrated electrocoagulation with zeolite adsorption in removing $NH_3$-N and colour from landfill leachate.

The performance removal of the EC process was influenced by several factors such as the effect of current density, electrolysis time and inter-electrode distance. In this study, the optimum removal of $NH_3$-N and colour using a single electrocoagulation process is explained in Section 3.2. Then, the performance removal of both pollutants using integrated treatment of electrocoagulation – zeolite adsorption process is discussed in Section 3.3.

### 3.1 Leachate Characteristics

The characteristics of PBSL leachate were analysed in Table 1. From Table 1, the leachate sample from PBSL was above the standard limit based on the permissible limit of leachate effluent based on the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation. In this study, the concentrations of colour increased, indicating that the leachate became more blackish with a mean value of 6363 Pt-Co whose permissible limit is 100 Pt-Co. The higher concentration of leachate color was primarily due to resistant dissolved organic molecules, such as humic substances [24].

The presence of humic acids was also revealed by the dark brown colour of landfill leachate [25]. In addition, leachate is regarded as one of the most toxic compounds for living organisms since it has such a high concentration of $NH_3$-N among its constituents. Problems such as depletion of dissolved oxygen, rapid eutrophication, and algae production may arise from the presence of a high concentration of $NH_3$-N [26]. During this study, it was discovered that the typical concentration of $NH_3$-N was 2044 mg/L; however, this concentration required to be reduced to less than 5 mg/L in order to be in compliance with the authorised limit of discharge requirement. In leachate, there is no limit on the discharge requirements of some parameters, such as salinity, conductivity, and turbidity. The key component that encourages algae development, increases eutrophication, interrupts biological treatment operations, and diminishes dissolved oxygen [27]. Coloured leachate can reduce the amount of light passing through the water bodies, affecting photosynthesis and the growth of aquatic plants. It also indicates the presence of hazardous substances that affect water supply quality and safety. As a result, both the high strength of $NH_3$-N and the color of the PBSL leachate required treatment before discharging to the river.

**Table 1** *Characteristic of PBSL leachate*

| Parameter | Minimum | Maximum | Average | Standard B |
|---|---|---|---|---|
| **Physical Characteristics:** | | | | |
| Turbidity, NTU | 126 | 499 | 256.22 | - |
| Colour (True), Pt-Co | 5020 | 8375 | 6363.00 | 100 |
| **Chemical Characteristics:** | | | | |
| Ammoniacal Nitrogen, mg/L | 1700 | 2700 | 2044.00 | 5 |
| pH | 10 | 12 | 11.00 | 6.0-9.0 |
| Total Dissolved Solid, g/L | 10 | 11.66 | 10.74 | - |
| Salinity, g/L | 10.52 | 17.61 | 13.66 | - |
| Conductivity, mS/cm | 10.5 | 18.8 | 14.37 | - |

### 3.2 Batch Electrocoagulation Study

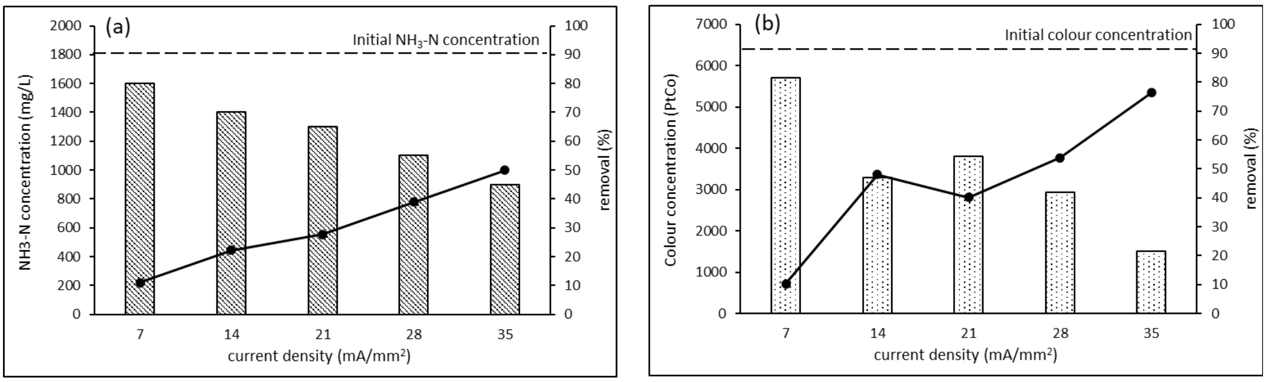
The batch electrocoagulation experiment was conducted to determine the optimum operation conditions such as the effect of current density, initial pH sample, electrolysis time and electrode inter-distance involved in this process. Sections 3.2.1 to 3.2.4 explain the findings obtained from the experiment of the single electrocoagulation study.

#### 3.2.1 The Effect of Current Density

Current density is one of the important factors that affects the removal efficiency in the EC process. This is because current density supplies electrical charges for ions to stabilise pollutants [28]. Theoretically, a higher current density will result in more pollutant removal [21]. Fig. 3(a) and (b) show the percentage removal of $NH_3$-N and colour resulting from the effect of current density on the EC process. The highest percentage removal of colour occurred at 48%, which was observed when the current was increased from 7 mA/mm$^2$ to 14 mA/mm$^2$. The percentage removal of $NH_3$-N increased at a constant rate as the current density increased, with the highest percentage of 50 % when the current density was configured at 35 mA/mm$^2$. At 14 mA/mm$^2$, $NH_3$-N was reduced from 1800 mg/L to 1400 mg/L followed by colour which was reduced from 6350 Pt-Co to 3300 Pt-Co, as shown in Fig. 3(a) and 3(b).

The results showed that a configuration of 14 mA/mm$^2$ significantly improved the efficiency in removing $NH_3$-N and colour. The result obtained has similarities to previous studies [29], [30], where their findings found that current density was optimised at 16.3 mA/mm$^2$ and 15.0 mA/mm$^2$ respectively. Furthermore, a lower current density reduced the electrode's oxidation rate, leading to a longer electrolysis time for the particles to stabilise, resulting in the formation of more flocs and sludge. Although the performance of $NH_3$-N and colour increased proportionally to the current density, an optimum current density of 14 mA/mm$^2$ was selected due to the ability of the electrodes to supply current to the reactor. Beyond 14 mA/mm$^2$, the Al electrode degraded and leached out into the leachate sample.

In the experiment, the removal of both $NH_3$-N and colour still increased beyond the current density of 35 A/mm$^2$. However, the experiment could not be continued due to the limitation of the DC power supply, which was able to generate a maximum current density of 35 mA/mm$^2$ and will be sparked when it exceeds the maximum current density.

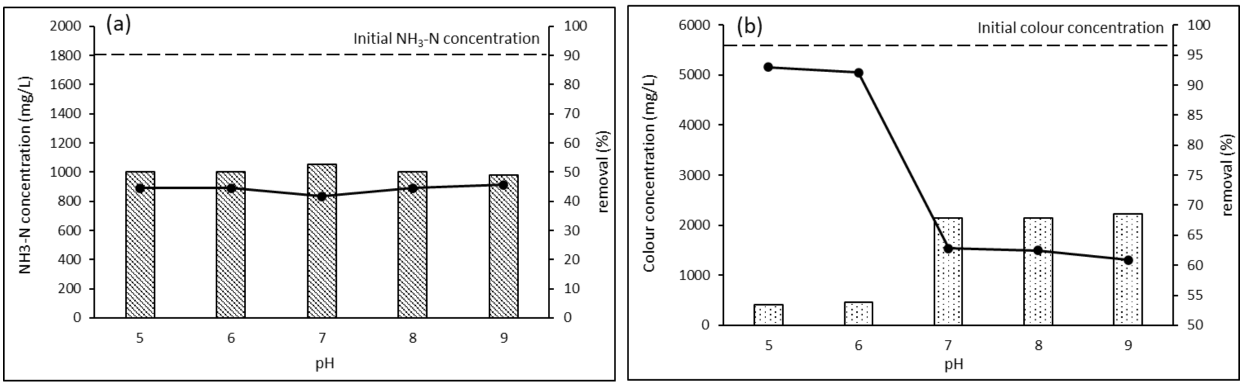



**(a)** **(b)**

**Fig. 3** *The effect of current density on (a) NH3-N; (b) colour*

### 3.2.2 The Effect of Initial pH

The effect of pH was investigated to determine the optimum pH required for the leachate to be effectively removed. $NH_3$-N and colour as the initial pH 9.0 act as controlling factors in the EC process. In this study, the initial pH value of the leachate sample was adjusted accordingly to 5.0, 6.0, 7.0, 8.0, and 9.0 of pH value. The range was selected based on the Environmental Quality (Control of Pollution from Solid Waste Transfer Stations and Landfill) Regulation. The batch EC study was performed using an optimum current density of 14.0 mA/mm$^2$ obtained from Section 3.2.1. The percentage removal of $NH_3$-N from pH 5.0 to pH 9.0 constantly at equilibrium, around 40% to 50% as shown in Fig. 4(a). In contrast, the initial pH of leachate is important for colour removal using the EC process, as shown in Fig. 4(b). Adjusting the pH value to an acidic condition led to an increase in the percentage of color removal from 63% at pH 8.0 (raw leachate) to 92% at pH 6.0. The leachate sample's pH decreased to pH 5.0, resulting in a slight increase in $NH_3$-N removal to 93%. However, [12] in their study indicates that the percentage removal of $NH_3$-N decreases when the pH values are too acidic (<pH 3.9) or too basic (>pH 10.0). Their finding also indicates the same percentage removal between 40% when pH 5.0 was applied in the EC process. Since the removal of $NH_3$-N between pH 5.0 and pH 9.0 was not significantly different, the optimum initial pH value was selected at pH 5.0 with the highest removal efficiency for both $NH_3$-N and colour.




**(a)** **(b)**

**Fig. 4** *The effect of pH on (a) NH3-N; (b) colour*

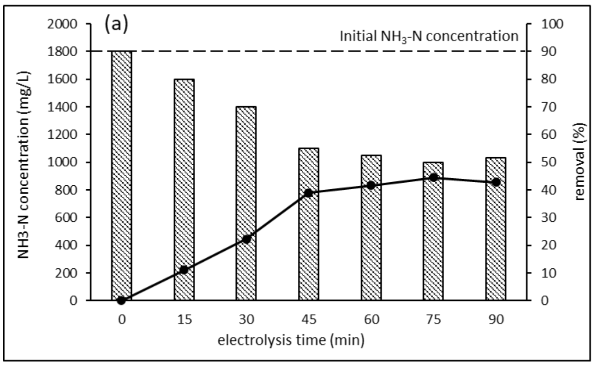
Since lowering the pH value increased the number of $H^+$ ions, the formation of flocs increased significantly with the lighter particles floating with the help of $H^+$ ions. A higher oxidation rate in an acidic state led to an increase in the production of hydroxide ions ($OH^-$) during electrode oxidation [29]. Higher $OH^-$ ion reacts with Al electrodes to form aluminium hydroxide (Al $(OH)_3$) during the EC process and is used as a supporting electrolyte to treat pollutants from landfill leachate [12]. Eqn. 2 shows the formation of Al $(OH)_3$ during this study.

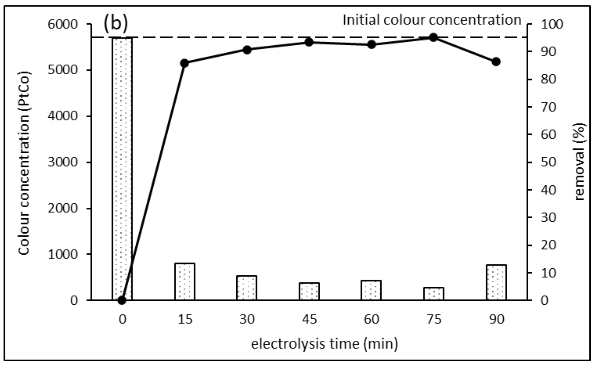
$$Al^{3+} + 3H_2O \quad \rightarrow \quad Al(OH)_3 + 3H^+ \qquad (2)$$

Al ion ($Al^{3+}$) produced by Al electrodes react with the leachate sample to produce Al $(OH)_3$ and additional $H^+$ ions [13]. The interaction of Al $(OH)_3$ with these particles stabilised them to form heavier sludge which settled at the bottom of the tanks [31].

### 3.2.3 The Effect of Electrolysis Time

Electrolysis time is a measurement of the overall duration of the ion exchanger process that occurs when direct current is applied to the leachate sample. Fig. 5(a) and (b) illustrate the percentage removal of $NH_3$-N and color, respectively, as a function of electrolysis time. Fig. 5(a) shows a significant increase in the percentage removal of $NH_3$-N from 11.11% to 38.89% from 15 to 45 minutes. This percentage then remains constant for 60 minutes, reaching an optimum removal of 44.44% at 75 minutes. The concentration of $NH_3$-N was reduced from 1800 mg/L to 1000 mg/L at optimum electrolysis of 75 minutes, as shown in Fig. 5(a). According to Fig. 5(b), the percentage removal of color increased for the first 30 minutes to 90.79% and gradually increased until the optimum percentage removal of 95.18% at 75 minutes. The colour concentration at 75 minutes was reduced from 5700 Pt-Co to 275 Pt-Co as shown in Fig. 5(b). The decrease in the percentage removal of $NH_3$-N and colour to 42.86% and 88.17% could potentially be attributed to the exhaustion of all Al ions due to the extended electrolysis time. The results showed that we achieved the highest percentage removal for both $NH_3$-N and colour at 75 minutes. Overall, the optimum electrolysis time occurred at 75 minutes with the highest percentage removal of $NH_3$-N and colour at 44.44% and 95.18%, respectively. The result can be compared to previous studies, which indicate a similar electrolysis time for the EC process between 60 and 80 minutes [32], [33].



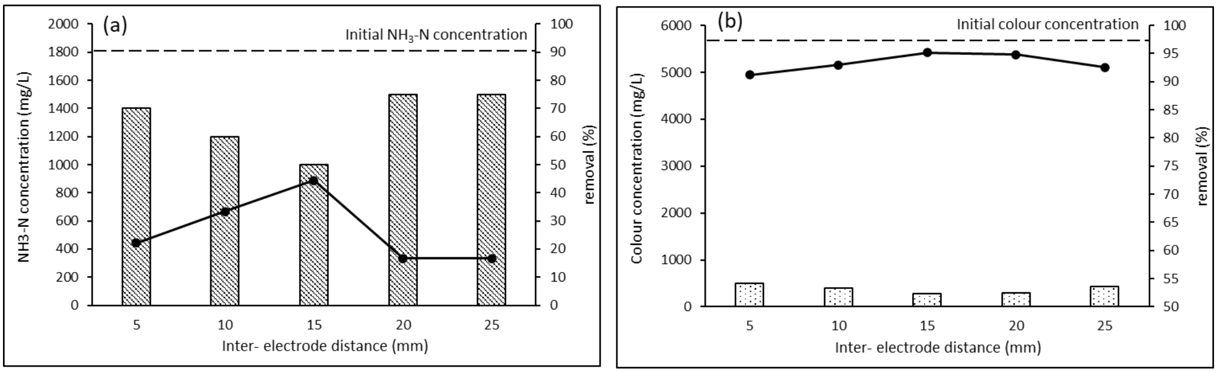

**(a)** **(b)**

**Fig. 5** *The effect of electrolysis time on (a) $NH_3$-N; (b) Colour*

### 3.2.4 The Effect of Inter-Electrode Distance

The effect of inter electrode distances against the percentage removal (a) $NH_3$-N (b) colour are presented in Fig. 6. The maximum percentage removal of $NH_3$-N was observed when the distance was increased from 5 mm to 15 mm. At this distance, the percentage removal of $NH_3$-N was increased from 22% to the maximum percentage of 44% with the concentration reduced from 1800 mg/L to 1000 mg/L as shown in Fig. 6(a). The efficiency of removal decreased when the distances were further apart, at 20 mm and 25 mm, with a constant removal rate of 28%. The removal trend of colour is also similar to the $NH_3$-N removal, which increased the colour removal from 91% to 95% when the distance between electrodes increased from 5 to 15 mm and suddenly dropped after an inter-distance of 15 mm (Fig. 6(b)). The results indicate that the efficiency decreases at lower electrode distances. When the electrode distance is extremely high, the solution resistance will increase [34]. If the solution resistance increases, the dissolution rate of aluminium will decrease, thus decreasing the efficiency of both pollutants.

From the observation, the efficiency of the EC process drops when the electrodes are too close or too far apart from each other. The configuration of two consecutive electrodes too far apart led to a decrease in efficiency, as the resistance within the leachate reduced the potential of current transfer [28]. Meanwhile, as the space between two electrodes becomes smaller, nearer distances reduce the volume of solution under treatment [35]. In conclusion, the optimum inter-electrode distances were at 15 mm, which indicates good agreement with previous studies [36].




**(a)** **(b)**

**Fig. 6** *The effect of inter-distance on (a) $NH_3$-N; (b) Colour*

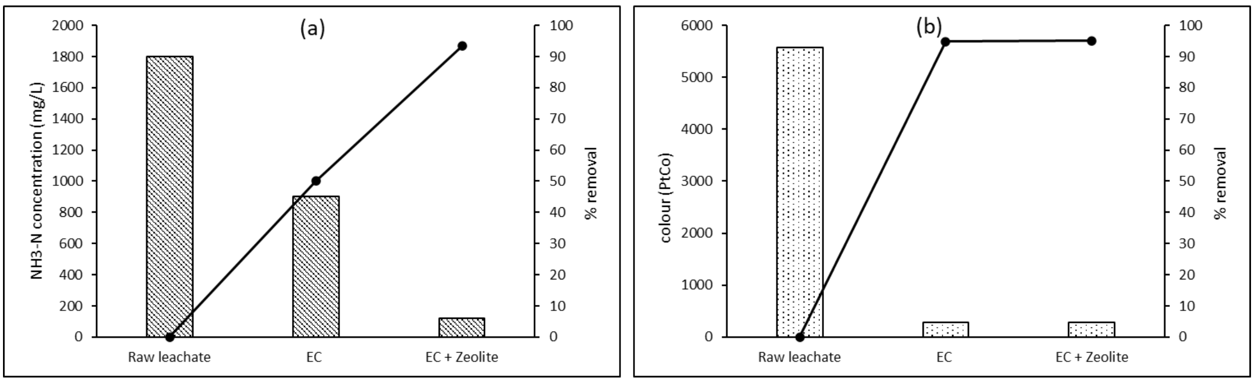
## 3.3 Integrated Electrocoagulation- Zeolite Adsorption Process

Integrated EC – zeolite adsorption processes were investigated to enhance the performance of the removal of $NH_3$-N and colour from the leachate sample. In this study, a raw leachate sample from PBSL was treated under an optimised EC process. Then, the treated effluent was retreated again using clinoptilolite zeolite in the adsorption process. Fig. 7 shows the raw and treated concentrations of $NH_3$-N and colour using both the single EC process and integrated EC – zeolite adsorption processes. Fig. 7 shows that an integrated process efficiently removed both $NH_3$-N and colour almost entirely from the leachate sample. As presented in Fig. 7(a), the treated concentration of $NH_3$-N was reduced from 1800 mg/L to 120 mg/L equivalent to 93.33% removal. A single EC process removed half of the $NH_3$-N concentration, while the clinoptilolite-zeolite adsorption process helped remove the remaining $NH_3$-N concentration.

In this study, a small amount of clinoptilolite zeolites dosage (2.0 g/ 150 mL) was able to perform similarly to the EC process. The performance of clinoptilolite zeolites to remove $NH_3$-N was due to the high capability of ion-exchange properties to dissolve $NH_3$-N into ammonia gas ($NH_3$) and ammonium ($NH_4^+$) as shown in Eqn. 3 and Eqn. 4 [37]. Additionally, clinoptilolite zeolites are also highly selective to adsorb $NH_4^+$ into their skeletal pores [19]. Based on previous studies, the capability of a single EC process to remove NH3-N from leachate was limited to a maximum of 50% capability [15], [33]. However, by integrated the process, the removal percentage would increase significantly, as proven by previous studies [19], [38].

$$NH_3 + H_2O \leftrightarrow NH_4{}^+ + 3H^+ \tag{3}$$

$$Clinoptiolite - Na^+ + NH_4{}^+ \leftrightarrow Clinoptiolite - NH_4{}^+ + Na^+ \tag{4}$$




**(a)** **(b)**

**Fig. 7** *Performance removal of (a) $NH_3$-N; and (b) colour between single electrocoagulation and integrated electrocoagulation-zeolite adsorption process*

On the other hand, Fig. 7(b) shows that a single EC process effectively removed the colour. The colour concentration of the leachate reduces from 5570 Pt-Co to 290 Pt-Co and slightly to 275 Pt-Co using the integrated EC – zeolite adsorption process. The overall percentage removal of colour using integrated EC – zeolite processes was 95%. The effectiveness of a single EC process to treat high concentrations of colour is due to the interaction of ions such as $H^+$ and Al $(OH)_3$ with smaller particles, which were stabilised to form flocs and sludge. However, clinoptilolite zeolites were unable to reduce colour concentration, which may be due to the low dosage of the adsorbent media. This was strongly proved by a previous study [38], which indicates optimum removal of colour as high as 73% was achieved when a higher dosage of clinoptilolite zeolites (110 g/sample) was configured. However, this simulated study might result in higher operational costs. Besides, previous studies found that integrated EC-activated carbon adsorption removed $NH_3$-N up to 98% as compared to single electrocoagulation, which can remove only 44% [39]. When integrated EC-adsorption is used, less electrolysis time is needed to lower the leachate's ideal concentration of $NH_3$-N. Eulmi et al. [40] found that the removal of $NH_3$-N and COD is more than 97% in 15 minutes of electrolysis time; however, using a single electrocoagulation process, it takes more than 60 minutes to remove around 50% of $NH_3$-N [12], [15], [40]. Thus, integrated EC – zeolite was highly recommended as an alternative leachate treatment to effectively remove high strength $NH_3$-N and colour by minimising the use of both aluminium and clinoptilolite zeolites.

## 4. Conclusions

The use of aluminium (Al) electrodes for the electrocoagulation (EC) process was capable of removing the highest amount of ammonia, nitrogen ($NH_3$-N), and colour under optimum conditions. Al electrodes formed Aluminium Hydroxide (Al $(OH)_3$) as supporting electrolytes that helped in ion exchange during the EC process. A batch EC study was carried out to determine the optimum removal of targeted pollutants by evaluating the effects of current density, initial pH value, electrolysis time, and inter-electrode distance. From the batch electrocoagulation experiment, the highest percentage removal of $NH_3$-N and colour were recorded at 47% and 90% respectively, when the EC process was optimised with a current density of 14.0 mA/mm$^2$, an electrolysis time of 75 minutes, and an inter-electrode distance of 15 mm. However, due to the lower percentage removal of $NH_3$-N at 44%, integrated EC – zeolite adsorption was required to enhance the removal of $NH_3$-N from landfill leachate.

The integration of EC with clinoptilolite zeolite adsorption resulted in a significant increase in the percentage removal of $NH_3$-N, from 47% to 87%, while the performance in removing colour stayed steady at 90%, despite the integration of the process. The result indicates that the single EC process was very effective in removing colour, while the integrated EC – zeolite adsorption process performed best to remove high strength of both $NH_3$-N and colour from leachate. The effectiveness of clinoptilolite zeolites to remove $NH_3$-N from leachate was due to a highly ammonium-ion selective present in clinoptilolite zeolites. Clinoptilolite zeolite adsorption removes ammonia through ion exchange and adsorption processes. Furthermore, clinoptilolite zeolites also have a higher adsorption rate due to their porous surfaces, which attract more pollutants, primarily $NH_3$-N. As a result, the effectiveness of removing $NH_3$-N and colour using EC increased when the process was integrated with clinoptilolite zeolite adsorption. The integrated EC – zeolite adsorption process was recommended as an alternative to replace the current physiochemical leachate treatment process and ensure that leachate management practices comply with environmental regulations and standards.

Besides, there are several limitations demonstrated from both single Electrocoagulation (EC) and integrated EC – zeolite adsorption processes during treatment of pollutants. Therefore, a few recommendations for improvement were required to address these limitations in future research such as to analyse the physical and chemical properties of the clinoptilolite zeolites adsorbent and to study the operating cost benefits analysis of single EC process and integrated EC – zeolite adsorption in term of economic point of view. Implementing these suggestions can enhance the efficacy of the proposed approach for managing landfill leachate.

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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:*** *Nor Azliza Akbar, Mohd Azhar Abd Hamid;* ***data collection:*** *Muhamad Hazwan Abdul Rahim, Muhammad Nasiruddin Hakimi;* ***analysis and interpretation of results:*** *Nur Shaylinda Mohd Zin, Badrul Nizam Ismail, Mohamad Sazali Said;* ***draft manuscript preparation:*** *Nor Azliza Akbar , Muhamad Hazwan Abdul Rahim . All authors reviewed the results and approved the final version of the manuscript.*

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