



Performance and Anode Consumption of Electrocoagulation and Hybrid Electrocoagulation–Ultrasonic Processes for Optimized Landfill Leachate Treatment

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ABSTRACT

This study investigates the effectiveness of electrocoagulation (EC) and hybrid electrocoagulation-ultrasound (EC-US) processes for treating landfill leachate from Alor Pongsu Landfill and Pulau Burung Sanitary Landfill in Malaysia. The treatment performance of the EC and EC-US processes was evaluated using aluminum (Al) and iron (Fe) electrodes under various operating conditions, including applied voltage (2 V - 10 V), inter-electrode distance (1 cm - 3 cm), and electrolysis time (5 - 30 minutes) in influencing COD removal efficiency from both landfill leachates. The results showed that the hybrid EC-US process significantly outperformed the EC process. Using Al electrodes, the maximum COD removal efficiency reached 95.05% and 96.31% for Alor Pongsu and Pulau Burung leachates, respectively, in the EC-US process, compared to 80.80% and 81.90% in the EC process. Both methods shared optimal operational parameters: 10 V, 2 cm inter-electrode distance, and 25 minutes of electrolysis time. The highest percentage of anode weight loss was 19.4% and 21.4%, as recorded in the EC-US process using Al electrodes for Alor Pongsu and Pulau Burung leachates, respectively, indicating enhanced coagulant generation. The findings demonstrate that the EC-US process is a promising and efficient approach for improving COD removal in landfill leachate treatment.

Keywords: Electrocoagulation, Ultrasonic, Hybrid process, Anode dissolution.

INTRODUCTION

Landfilling is the most widely used method for solid waste management, making it essential to effectively treat the resulting leachate (Rookesh et al., 2022). As water percolates through waste, it generates leachate containing a complex mixture of organic and inorganic substances, heavy metals, and xenobiotic organic compounds (Lindamulla et al., 2022; Ma et al., 2022).

Untreated leachate can pose serious environmental risks, especially since it can continue to be generated for 30 to 50 years after a landfill has been closed (Apaydin & Özkan, 2020). Leachate properties and quantity are influenced by factors, including precipitation, run-off, infiltration, evaporation, waste compaction, landfill age, transpiration, temperature, waste composition, density, depth, moisture content, flow rate, degradation cycle, and disposal method (Abdel-Shafy et al., 2023;

Lindamulla et al., 2022; Yaashikaa et al., 2022). Chemical oxygen demand (COD) measures the pollutant strength of biodegradable and non-biodegradable organic matters in a sample. The COD ranges for young, intermediate, and stabilized leachates are 10,000 mg/L and above, 4,000–10,000 mg/L, and less than 4,000 mg/L, respectively (Zakaria & Aziz, 2017).

Diverse methods, encompassing chemical, physical, and biological processes, have been employed for the treatment of landfill leachate (Al-Nawaiseh et al., 2021). Treatment of landfill leachate primarily utilizes physical and chemical processes. These processes demonstrate drawbacks, such as the generation of significant sludge and a requirement for extensive chemical constituents (Guo et al., 2022). In addition, biological treatment has certain limitations. The persistent nature of organic carbon in landfill leachate often results in inadequate removal efficiency when treating aged leachate with low biodegradability (Nohara et al., 2023; Yang et al., 2022). Moreover, the high concentrations of ammonia nitrogen and heavy metals commonly found in aged leachate can significantly inhibit microbial activity in leachate (Guo et al., 2022). Membrane separation, ion exchange, and reverse osmosis are effective methods for reducing metal ions in landfill leachate; however, they face operational challenges and incur high installation costs (Hassani et al., 2022). Traditional treatment methods present significant challenges for effective leachate treatment and can lead to long-term environmental issues. Consequently, more effective treatment methods for landfill leachate have been identified.

Electrocoagulation (EC) is a promising wastewater treatment technique that integrates electrochemical, coagulation, and flotation processes to remove a wide range of pollutants (Tejera et al., 2021). EC stands out for its environmental friendliness, high removal efficiency, low sludge production, and minimal chemical requirements, while also being simple to operate, energy-efficient, and economically viable (Deng et al., 2021; Kundu et al., 2024). In the mechanism of the EC process that uses sacrificial electrodes, such as aluminum (Al) or iron (Fe), metal ions are released under the application of direct current and subsequently form hydroxide species that act as coagulants (Kundu et al., 2024; Muvel et al., 2024). These species effectively destabilize and aggregate organic and inorganic contaminants, including

refractory compounds commonly found in landfill leachate (Chen et al., 2022). In recent years, EC has been successfully applied in the treatment of various wastewater types, including landfill leachate, textile effluents, tannery wastewater, and domestic sewage (Al-Qodah & Al-Shannag, 2019). Due to its operational flexibility and ability to handle complex waste streams, EC has become an attractive alternative to conventional treatment methods.

The EC process is a popular method for treating landfill leachate. However, this process has some limitations, such as the formation of a passive film on the electrode surface over time (Khoramipour et al., 2021). Some studies have combined the EC process with other treatment technologies, such as sonolysis, photolysis, adsorption, and ozonation, to address these drawbacks (Asaithambi et al., 2020). Ultrasonic treatment is an advanced oxidation process (AOP) that demonstrates significant efficacy in treating landfill leachate by generating highly reactive hydroxyl radicals ($\bullet\text{OH}$) and initiating pyrolysis (Soomro et al., 2020). The primary mechanism involves acoustic cavitation, where the creation and rapid implosion of micro-bubbles under ultrasonic irradiation generate extreme localized conditions of high temperature and pressure (Ritesh & Srivastava, 2020). These conditions facilitate the decomposition of organic and inorganic pollutants through oxidative degradation and physical disruption (Lei et al., 2023). Ultrasonic treatment has been successfully applied in various fields, including wastewater treatment, due to its ability to oxidize a wide range of contaminants, such as refractory organics and ammonia, without the need for chemical additives (Chen et al., 2022). Recently, researchers have increasingly concentrated on the combined US-EC process for the treatment of landfill leachate, as shown by (Afsharnia et al., 2018; Al-Rubaiey et al., 2018; Asaithambi & Govindarajan, 2021; Khoramipour et al., 2021). Therefore, the combined process of EC with US treatment presents a promising method for enhancing landfill leachate quality by increasing the removal efficiency of various contaminants. Considering that existing leachate processes are often site-specific, complex, and expensive, there is an urgent need for additional studies into low-cost, efficient, and scalable alternatives.

Therefore, a significant research gap exists regarding a comprehensive evaluation of the effectiveness of EC

and EC-US in treating landfill leachate. This study aims to address this gap by comparing the performance of EC and EC-US in terms of COD removal efficiency at two distinct landfill sites in Malaysia. Additionally, the study examines electrode consumption by assessing the weight loss of various anode materials, specifically Al and Fe, under various controlled operating conditions, including voltage, inter-electrode distance, and electrolysis time.

MATERIALS AND METHODS

Landfill Leachate Characteristics

This study employed landfill leachate samples collected from two distinct landfill sites in Malaysia:

Alor Pongsu Landfill situated in Perak, and Pulau Burung Sanitary Landfill located in Penang. Alor Pongsu Landfill is positioned at 5°04' N and 100°35' E, while Pulau Burung Sanitary Landfill is located at 5.1942° N latitude and 100.42° E longitude. The sample was collected by a grab sample.

Both landfill sites receive municipal solid waste and produce old leachate, with Alor Pongsu operating as an anaerobic landfill and Pulau Burung Sanitary Landfill being a semi-aerobic landfill. These variations in landfill operations contribute to differences in the leachate composition. The physico-chemical characteristics of the leachate samples from both sites were analyzed and are summarized in Table 1.

Table 1. Landfill leachate characteristics (mean $\pm$ standard deviation)

Parameter	Min.	Max.	Alor Pongsu	Min.	Max.	Pulau Burung landfill
Temperature (°C)	25.9	31.6	29.8 $\pm$ 1.51	29	33.5	30.12 $\pm$ 0.9
pH	7.85	8.64	8.09 $\pm$ 0.2	7.92	8.32	8.12 $\pm$ 0.1
Color (Pt-Co)	788	2204	1496 $\pm$ 354	880.2	2427.8	1654 $\pm$ 386.9
Salinity (ppt)	9.10	11.3	10.2 $\pm$ 0.55	8.11	10.51	9.31 $\pm$ 0.6
Conductivity (μ S/cm)	8357.11	12285.11	10321.11 $\pm$ 987	15785.97	20451.97	18118.97 $\pm$ 1166.5
Turbidity (NTU)	24.2	43.45	25.45 $\pm$ 9	28.85	84.85	56.85 $\pm$ 14.0
BOD ₅ (mg/L)	92	232.23	106.23 $\pm$ 63	98	243.63	115.63 $\pm$ 64.0
COD (mg/L)	1540	3309	2145 $\pm$ 582	2221.2	3494.8	2858 $\pm$ 318.4
TDS (mg/L)	6007.22	7167.23	6587.23 $\pm$ 580	9244.05	11802.45	10573.25 $\pm$ 664.6
DO (mg/L)	0.57	1.37	0.97 $\pm$ 0.4	0.63	1.83	1.23 $\pm$ 0.3
BOD ₅ /COD	0.03	0.08	0.05 $\pm$ 0.02	0.01	0.07	0.04 $\pm$ 0.015

EXPERIMENTS' SET-UP

Electrocoagulation Set-up

The EC experiments were conducted in a 250-mL cylindrical glass beaker, which functioned as the EC reactor, as illustrated in Figure 1. The reactor was filled with 150 mL of landfill leachate wastewater as the working volume. Two rectangular plates (2 cm W $\times$ 15 cm H) were used as electrodes, with one serving as the anode and the other serving as the cathode. A direct current (DC) power supply (OJES PS6005, 60V/5A) provided the required current intensity for the EC process. To ensure uniform mixing and maintain the homogeneity of the wastewater throughout the experiment, a magnetic stirrer was employed for continuous stirring. The EC system utilized rectangular electrodes, which served as both anode and cathode.

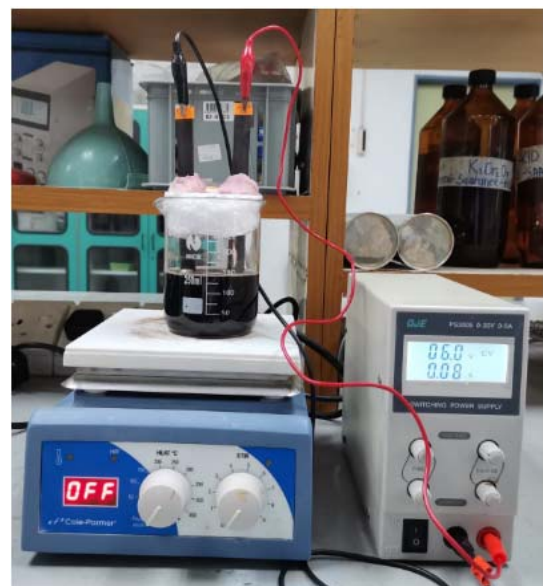


Figure 1. EC set-up

Electrocoagulation-Ultrasonic Set-up

The EC-US experiments were conducted using an ultrasonic bath filled with distilled water to facilitate ultrasonic wave transmission. The EC reactor, containing 150 mL of wastewater, was placed inside the ultrasonic bath, ensuring effective ultrasonic energy transfer during the treatment process, as illustrated in Figure 2. A continuous ultrasonic wave was applied throughout the experiment at a frequency of 40 kHz with an output power of 110 W to enhance pollutant removal. The combination of EC and US treatments aimed to improve the removal efficiency of pollutants in landfill leachate.



Figure 2. EC-US set-up

Procedure and Analytical Method

The EC and EC-US processes were conducted in a batch reactor to evaluate the removal of COD from landfill leachate. Al and Fe electrodes were used as anodes and positioned vertically and parallel to each other with varying inter-electrode distances of 1 cm, 2 cm, and 3 cm. A DC power supply was used to regulate the applied voltage, which was varied at 2V, 4V, 6V, 8V, and 10V. Each experiment was conducted for different electrolysis times of 5, 10, 15, 20, 25, and 30 minutes to assess the impact of electrolysis time. Before each experiment, the electrodes were dried and weighed using an analytical balance to determine their initial mass. The treatment was performed in batch mode, with a fixed volume of landfill leachate. For EC-US experiments, ultrasonic waves were applied simultaneously with EC to evaluate their effect on COD removal and anode dissolution. After each experiment, the treated samples were left for 30 minutes to allow floc settling before the analysis.

RESULTS AND DISCUSSION

To achieve the objectives of this study, both the EC process and the combined EC-US process were applied to assess the effectiveness of COD removal from landfill leachate collected from Alor Pongsu landfill and Pulau Burung landfill. The treatment performance was evaluated using Al and Fe electrodes. For both EC and EC-US processes, the influence of key operational parameters, such as inter-electrode distance, electrolysis time, and applied voltage, was systematically investigated to determine their impact on COD removal efficiency.

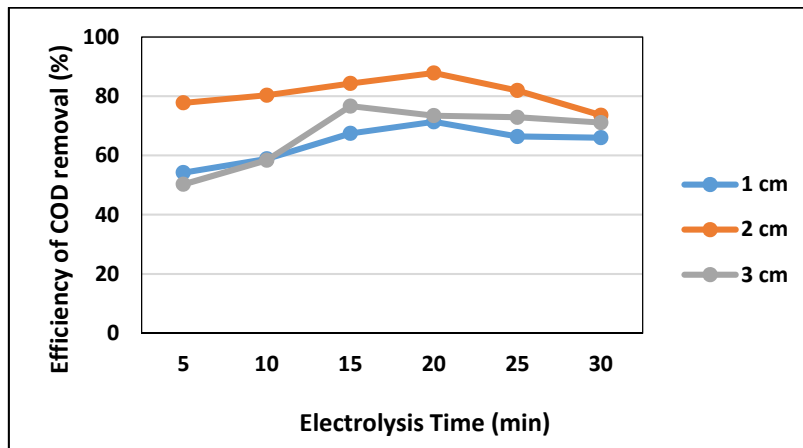
Effect of the Operational Condition on the EC and EC-US Processes

Effect of Inter-Electrode Distance on the Processes

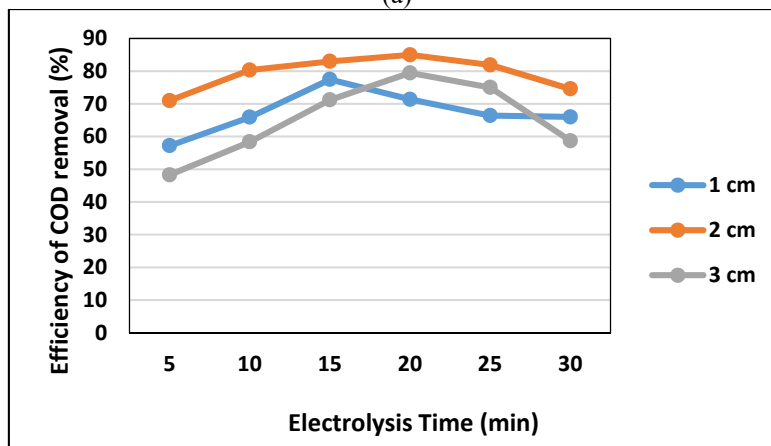
The inter-electrode distance is a vital factor in the EC process. This relates to the electro-static field, where the optimal inter-electrode distance results in the most efficient pollutant removal. Figure 3. (a) and (b) present the trends in COD removal efficiency over a 30-minute treatment period for landfill leachate samples from Alor Pongsu and Pulau Burung, respectively, using Al electrodes during the EC process. In both cases, the experiments were conducted under a constant applied voltage of 10 V, with Al electrodes serving as both the anode and cathode. The inter-electrode distances were varied at 1 cm, 2 cm, and 3 cm to evaluate their influence on COD removal performance. The figure illustrates the temporal evolution of COD removal efficiency, highlighting the effect of inter-electrode spacing on treatment effectiveness at each landfill site. The results indicate that COD removal efficiency increases with electrolysis time, reaches a maximum at an optimal point, and then gradually declines. This pattern suggests the existence of an optimal electrolysis time beyond which the process becomes less effective, likely due to the saturation of coagulating agents or re-dissolution of flocs. These findings are consistent with those reported by Lu et al. (2023), who observed similar trends in their studies on COD removal from landfill leachate using the EC process. In this study, the highest COD removal efficiency for Alor Pongsu landfill leachate was 71.38%, achieved at an inter-electrode distance of 1 cm, an applied voltage of 10 V, and an electrolysis time of 20 minutes. Similarly, for Pulau Burung landfill leachate, the maximum COD removal reached 77.47% under

identical voltage and inter-electrode spacing conditions, but with a shorter treatment time of 15 minutes. The removal of COD in the EC process primarily occurs through the coagulation of organic matter facilitated by *in situ*-generated coagulants (Rajaei et al., 2021). Furthermore, reducing the inter-electrode spacing below 1 cm can hinder the free flow of the solution between

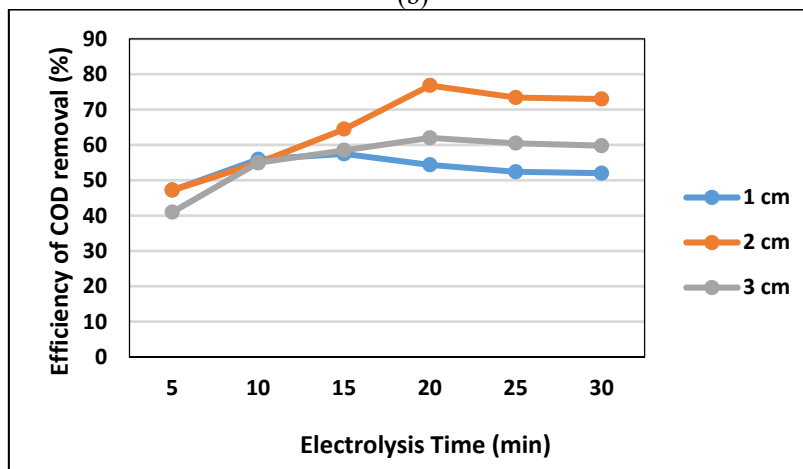
electrodes, thereby limiting mass transfer and adversely affecting removal efficiency (Bharath et al., 2018). These observations underscore the importance of carefully optimizing operational parameters, such as inter-electrode distance, electrolysis time, and voltage, to maximize the performance of the EC process in landfill leachate treatment.



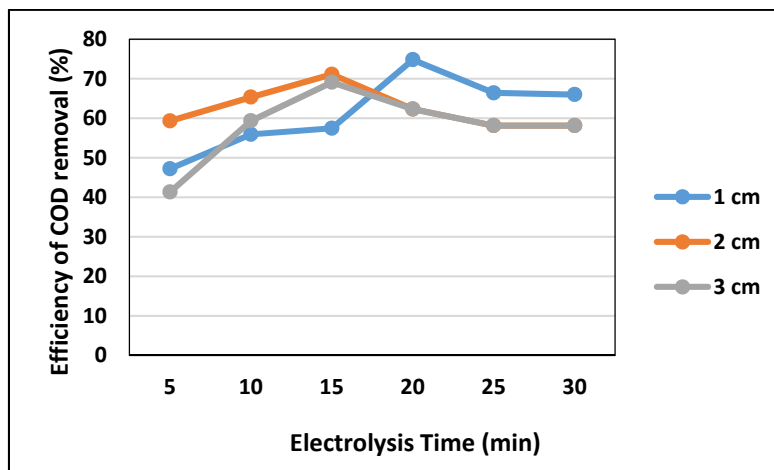
(a)



(b)



(c)



(d)

Figure 3. Effect of inter-electrode distance on COD removal efficiency during the EC process with the voltage of 10 V using (a) Al electrode for Alor Pongsu landfill; (b) Al electrode for Pulau Burung landfill (c) Fe electrode for Alor Pongsu landfill; (d) Fe electrode for Pulau Burung landfill

At an inter-electrode distance of 2 cm, the Alor Pongsu landfill leachate exhibited the highest COD removal efficiency of 87.87% after 20 minutes of treatment using the EC process. This represents a 16.5% improvement compared to the efficiency achieved at a 1-cm spacing, indicating that increasing the inter-electrode distance had a significant positive impact on pollutant removal performance. Similarly, in the case of the Pulau Burung landfill leachate, the maximum COD removal efficiency of 85% was attained at an applied voltage of 10 V and a treatment time of 20 minutes. An increase in the inter-electrode distance from 1 cm to 2 cm also led to an improvement in COD removal, albeit more modest, with an increase of approximately 6%. This indicates that the removal efficiency of COD in landfill leachate was increased by increasing the electrode distance during the EC process. On the other hand, at 3-cm inter-electrode distance, the optimum operating conditions to achieve the highest COD removal efficiency varied slightly between the two landfill leachates. For Alor Pongsu landfill leachate, the highest COD removal efficiency of 76.67% was obtained at an applied voltage of 10 V and a treatment time of 15 minutes. In contrast, the Pulau Burung landfill leachate demonstrated a slightly higher COD removal efficiency of 79.47% under the same voltage (10 V), but with a longer treatment time of 20 minutes. These findings suggest that the optimal treatment time may differ depending on the leachate composition. This highlights the importance of tailoring the operational

parameters based on site-specific leachate characteristics to maximize treatment efficiency. An increase in COD removal efficiency was observed when the inter-electrode distance using Al electrodes increased from 1 cm to 2 cm; however, a further increase to 3 cm resulted in a decline in removal efficiency. An increased inter-electrode distance results in a diminished speed of the generated ions. Due to their slower movement, ions require additional time to generate the floc necessary for pollutant coagulation (Nassar et al., 2023). This trend is consistent with the findings of Manikandan and Saraswathi (2023), who reported that increasing the distance between electrodes reduces the electro-static attraction between the electrodes and the dissolved ions, thereby slowing the flocculation process.

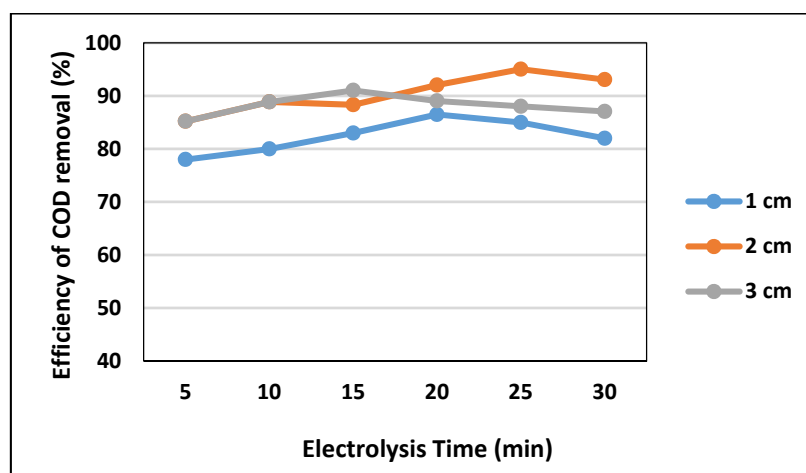
In conclusion, the maximum COD removal efficiency of 87.87% for Alor Pongsu landfill leachate was achieved using aluminum electrodes as both anode and cathode under optimal operating conditions: a voltage of 10 V, an inter-electrode distance of 2 cm, and a treatment time of 20 minutes. For Pulau Burung landfill leachate, the highest COD removal efficiency recorded was 85%, also using Al electrodes under the same operational conditions. These findings demonstrate that an inter-electrode distance of 2 cm is optimal for enhancing COD removal efficiency in the EC process.

Figure 3 (c) and (d) show the percentage of COD removal efficiency in Alor Pongsu and Pulau Burung landfill leachate by placing Fe electrodes, respectively,

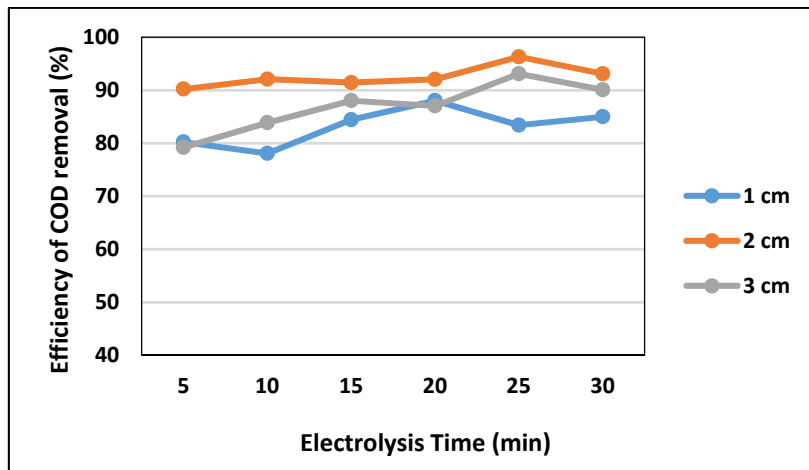
at varying inter-electrode distances. A comparison between Al and Fe electrodes revealed that Al consistently achieved higher COD removal efficiencies in both Alor Pongsu and Pulau Burung landfill leachates. For Alor Pongsu leachate, Al electrodes achieved 87.87% COD removal compared to 76.85 % by Fe electrodes at an inter-electrode distance of 2 cm, indicating an improvement of approximately 11.02 % at 10 V and 20 minutes of treatment. Similarly, in Pulau Burung leachate, Al electrodes recorded 85% COD removal, outperforming Fe electrodes, which reached 62 % under the same conditions at 2 cm. This superior performance of Al electrodes can be attributed to the differences in the electro-chemical dissolution behavior of the electrode materials. Fe electrodes undergo dissolution at the anode to produce Fe^{2+} ions, which are subsequently oxidized to Fe^{3+} in the presence of dissolved oxygen. These Fe^{3+} ions then hydrolyze to form $\text{Fe}(\text{OH})_3$, which acts as the primary coagulant. In contrast, Al electrodes dissolve to directly release Al^{3+} ions, which rapidly react with OH^- ions to form monomeric and polymeric hydrolyzed species. These Al-based coagulants are more reactive and efficient in destabilizing and aggregating organic pollutants, contributing to the improved COD removal performance observed (Taib et al., 2021). Moreover, since the cost of Al and Fe electrodes is relatively similar, the selection of Al electrodes represents a more cost-effective option for achieving higher COD removal in landfill leachate

treatment processes. The results align with a previous study conducted by Somroo et al. (2020), who observed that Al electrodes were more efficient than Fe electrodes for COD removal in landfill leachate.

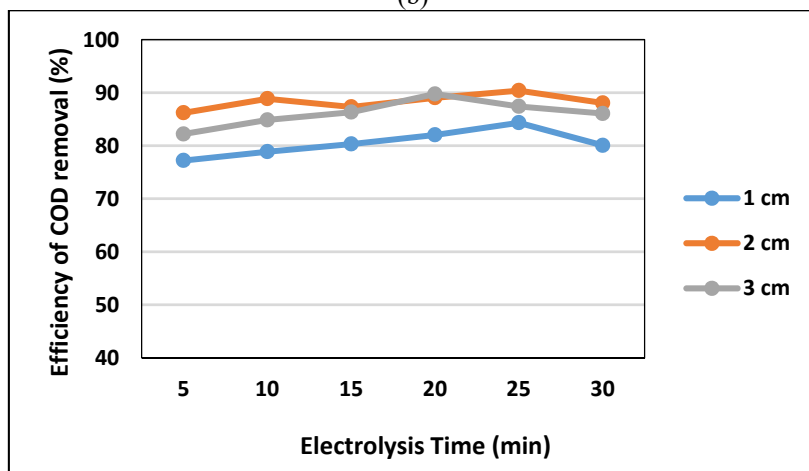
The effect of increasing the inter-electrode distance from 1 cm to 2 cm between Al electrodes was evaluated for both Alor Pongsu and Pulau Burung landfill leachates during the hybrid EC-US treatment. At a 2 cm spacing, the highest COD removal efficiencies of 95.05% and 96.31% were achieved for Alor Pongsu and Pulau Burung leachates, respectively, under a voltage of 10 V and a treatment time of 25 minutes. Based on Figure 4 (a) and (d), these results indicate a notable improvement in the EC process when using Al and Fe electrodes. The production of hydroxyl ions, crucial for the destabilization and precipitation of pollutants, is reduced at wider electrode distances due to decreased electro-chemical activity (Nassar et al., 2023). Conversely, when the inter-electrode distance is insufficient (i.e., below 2 cm), the electro-static field intensity becomes excessive, leading to intense collisions among the developing flocs. The robust electro-static attraction may result in floc dis-integration or re-dispersion, thereby impairing treatment efficacy (Tahreen et al., 2020). Consequently, sustaining an optimal inter-electrode distance is essential for enhancing coagulant production while reducing floc destabilization, thereby ensuring efficient pollutant removal.



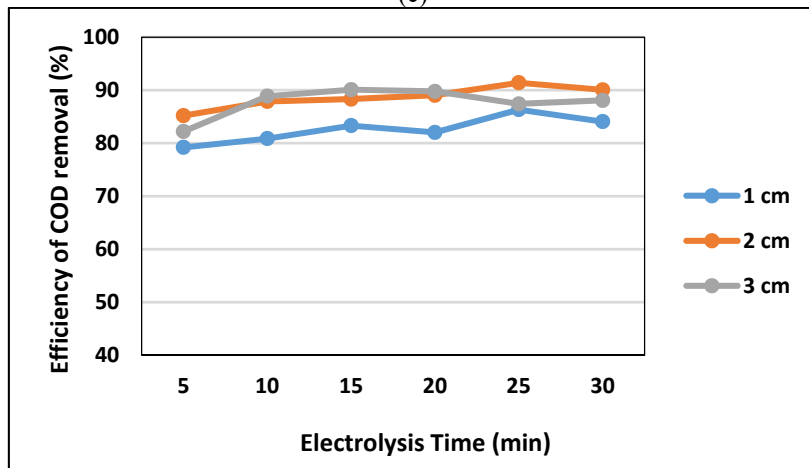
(a)



(b)



(c)



(d)

Figure 4. Effect of inter-electrode distance on COD removal efficiency during the EC-US process with the voltage of 10 V using (a) Al electrode for Alor Pongsu landfill; (b) Al electrode for Pulau Burung landfill (c) Fe electrode for Alor Pongsu landfill; (d) Fe electrode for Pulau Burung landfill

This observation is consistent with findings from Moradi et al. (2021), who reported improved COD and

turbidity removal at greater electrode distances due to reduced electrode passivation and more efficient

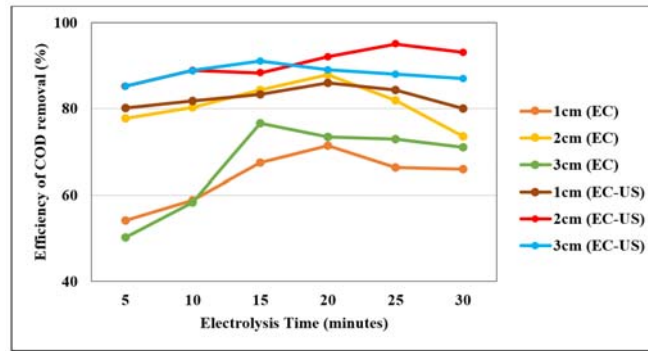
coagulant generation. In contrast, the highest COD removal efficiencies of 90.4% and 91.4% were achieved for Alor Pongsu and Pulau Burung landfill leachates, respectively, using Fe electrodes as both anode and cathode during the hybrid EC-US process. The optimal results were obtained using Al under operating conditions of a 2-cm inter-electrode distance, an applied voltage of 10 V, and a treatment duration of 25 minutes. The findings confirm that the integration of ultrasonic irradiation with EC significantly enhances the removal of organic pollutants from landfill leachate, particularly under optimized electrode spacing and operational parameters. The improved performance of the EC-US system can be attributed to the synergistic effect of ultrasonic cavitation, which promotes strong micro-mixing and facilitates the dispersion and collision of coagulant flocs, thereby enhancing mass transfer and pollutant destabilization. Additionally, the presence of hydroxyl radicals ($\cdot\text{OH}$), which are strong oxidants capable of eliminating various organic and inorganic materials, contributes to this improved performance (Dizge et al., 2018; Ritesh & Srivastava, 2020). These findings highlight the potential of integrating ultrasonic irradiation with EC to enhance the degradation of refractory organic pollutants in leachate treatment.

Effect of the Electrolysis Time on the Processes

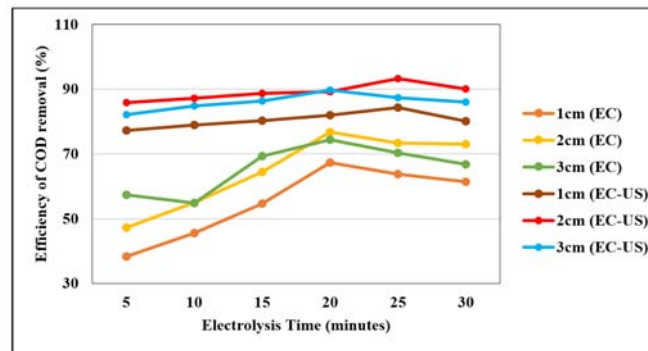
Electrolysis time is a critical operational parameter influencing the efficiency of COD removal from landfill leachate in both the EC process and the combined EC-US process. In this study, a fixed treatment duration of 30 minutes was applied for each experimental run. Figure 5 (a) and (b) present the COD removal efficiency in Alor Pongsu landfill leachate using Al and Fe electrodes, respectively, comparing the EC and EC-US processes at inter-electrode distances of 1 cm, 2 cm, and 3 cm. As anticipated, the hybrid EC-US process consistently outperformed the EC process, demonstrating significantly higher COD removal across all conditions. This is attributed to the chemical and physical effects of the cavitation (Lei et al., 2023). The chemical effects demonstrate that the ultrasound irradiation produces cavitation micro-bubbles that collapse, leading to the *in situ* production of free radicals, such as hydroxyl radicals ($\cdot\text{OH}$) (Emerick et al., 2020), which interact with and degrade organic matter (Ritesh & Srivastava, 2020; Thokchom et al.,

2015). Additionally, the physical effects include shock waves, micro-jets, turbulence, and acoustic streaming, which accelerate the chemical reaction rate, augment the mass transfer of pollutants between the electrodes, and reduce the thickness of the diffusion layer (Asaithambi et al., 2020; Patidar & Srivastava, 2021; Ritesh & Srivastava, 2020). These effects maintain the electrode's active sites, thereby improving coagulant generation and overall treatment efficiency. During the EC process, anodic dissolution leads to the continuous release of metal ions, which subsequently form metal hydroxide flocs acting as coagulants. According to Tahreen et al. (2020), under constant current density, extending the electrolysis time allows for a greater accumulation of these hydroxides, thereby increasing floc formation and promoting more effective coagulation and sedimentation. However, this improvement continues only up to an optimal treatment time. Al electrodes exhibited enhanced performance at 25 minutes, attaining COD removal efficiencies of 81.9% (EC) and 95.05% (EC-US), in contrast to 73.4% (EC) and 90.4% (EC-US) for Fe electrodes. Beyond that point, the availability of dissolved ions may decrease due to the saturation or passivation of electrode surfaces, leading to a decline in floc production and, consequently, a reduction in COD removal efficiency. Therefore, identifying an optimal treatment duration is essential to balance effective contaminant removal with energy efficiency and process stability.

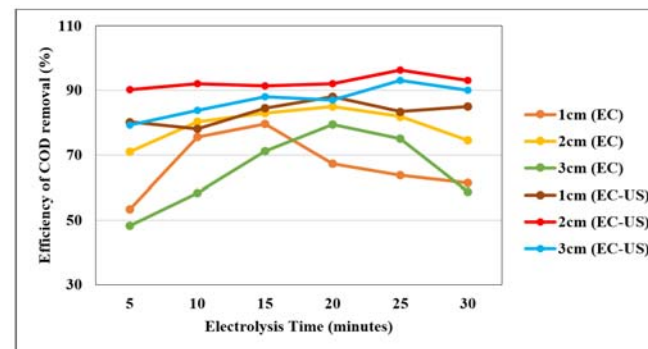
Figure 5 (c) and (d) illustrate the effect of electrolysis time on COD removal efficiency in Pulau Burung landfill leachate using Al and Fe electrodes, comparing the performance of the EC process and the hybrid EC-US process at inter-electrode distances of 1 cm, 2 cm, and 3 cm. As shown in both figures, COD removal efficiency for both types of electrodes increases with electrolysis time. However, beyond an optimum treatment duration, the removal efficiency plateaus, indicating that further extension of electrolysis time does not yield additional improvement. This phenomenon can be attributed to certain organic matter not interacting with the coagulants and not decomposing via EC (Galvão et al., 2020; Lu et al., 2023). In addition, extended treatment may lead to anodic passivation and cathodic polarization, which impair the performance of the EC system by reducing the availability of reactive electrode surfaces (Manikandan & Saraswathi, 2023).



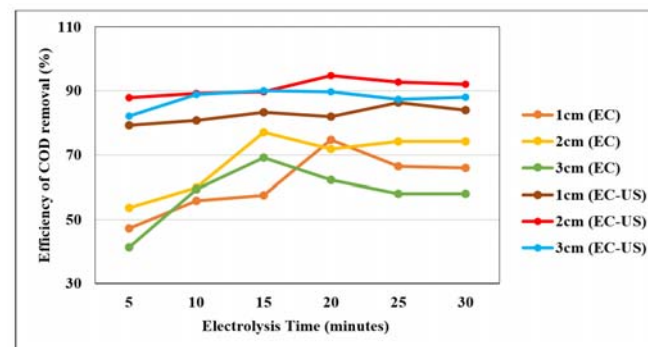
(a)



(b)



(c)



(d)

Figure 5. Effect of electrolysis time on COD removal efficiency using (a) Al in Alor Pongsu landfill; (b) Fe in Alor Pongsu landfill; (c) Al in Pulau Burung landfill; (d) Fe in Pulau Burung landfill; during EC process and EC-US process

A similar trend has been observed when examining the influence of applied voltage on COD removal efficiency. The effectiveness of COD removal is closely linked to the concentration of metal ions generated at the electrodes. According to Joshi and Gogate (2019), the concentration of metal ions and their associated hydroxide flocs increase with electrolysis time, enhancing coagulation efficiency. This correlation is evident in Figure 5 (c) and (d), where longer electrolysis times correspond to higher COD removal for both Al and Fe electrodes. At all tested electrode spacings (1 cm, 2 cm, and 3 cm), the highest COD removal rates were observed at electrolysis times exceeding 15 minutes for both the EC and EC-US processes. In the case of Al electrodes, extended electrolysis time leads to increased release of Al^{3+} ions, following Faraday's law, thereby enhancing the formation of aluminum hydroxide flocs that improve pollutant removal (Tahreen et al., 2020). For Fe electrodes, Fe^{2+} ions released through anodic dissolution help neutralize the surface charge of colloids, aiding in their destabilization and aggregation. As highlighted by Moradi et al. (2021), insufficient electrolysis time results in limited charge neutralization, thereby reducing the effectiveness of coagulation. Additionally, the generation of hydrogen gas bubbles during electrolysis contributes to the flotation of precipitates and flocs, which further assists in the separation of organic matter (Marmanis et al., 2021). The trends observed in Pulau Burung are consistent with those reported in Alor Pongsu, where COD removal efficiency increased with electrolysis time up to an optimal point, after which the efficiency either stabilized or declined.

These findings are in agreement with those of prior studies. For instance, Moradi et al. (2021) reported a maximum COD removal efficiency of 68.77% within the first 20 minutes, followed by a significant decline to 12.34% after 30 minutes of treatment. Similarly, Afsharnia et al. (2018) demonstrated that the integration of ultrasonic irradiation significantly enhances COD removal, achieving efficiencies of 40% in the US system alone and up to 98% when combined with EC. This improvement was attributed to a combination of factors, including electrolyte reactions at the electrode surface, adsorption of pollutants by coagulants, and hydroxyl radical production induced by ultrasonic cavitation.

Effect of Voltage on the Processes

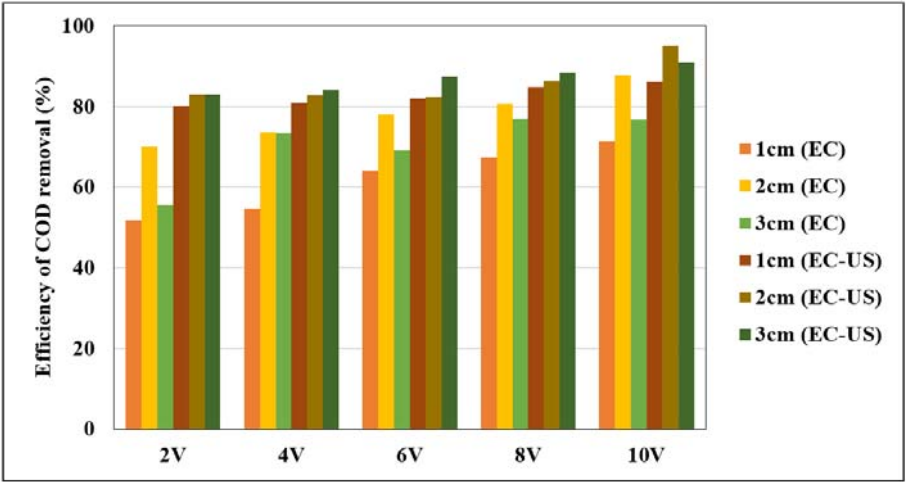
Voltage is a critical operational parameter

influencing the COD removal efficiency in landfill leachate during both the EC process and the hybrid EC-US process. In this study, voltages of 2V, 4V, 6V, 8V, and 10V were investigated to evaluate COD removal from Alor Pongsu and Pulau Burung landfill leachates using Al and Fe electrodes. Figure 6 (a) and (b) illustrate the percentage of COD removal in Alor Pongsu leachate using Al and Fe electrodes, respectively, at varying voltages and fixed inter-electrode distances of 1 cm, 2 cm, and 3 cm, comparing both the EC and EC-US processes. The combined EC-US process consistently achieved higher COD removal efficiencies compared to the EC process for both electrode types. The highest removal rates were observed at 10V for both Al and Fe electrodes. Notably, Al electrodes demonstrated superior performance at this voltage, achieving COD removal efficiencies of 81.9% (EC) and 95.05% (EC-US), compared to 73.4% (EC) and 90.42% (EC-US) for Fe electrodes. These findings align with the results of Moradi et al. (2021), who reported that Al electrodes are more effective in adsorbing dissolved organic compounds and colloidal particles, primarily due to the formation of $\text{Al}(\text{OH})_3$ flocs with a higher specific surface area, enhancing their coagulation capacity.

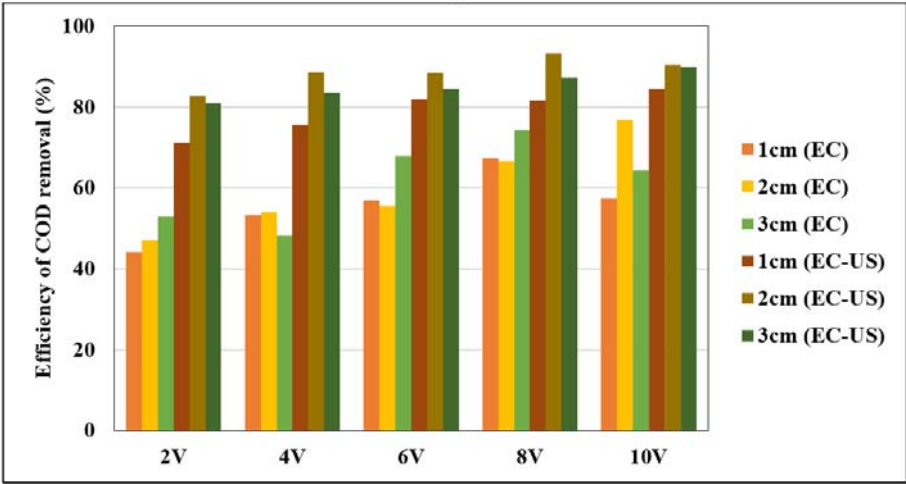
Figure 6 (c) and (d) present the results for Pulau Burung leachate under similar conditions. The highest COD removal efficiency was achieved at the maximum applied voltage of 10 V for both Al and Fe electrodes. However, the Al electrode exhibited superior performance, with removal efficiencies of 81.9% (EC) and 96.31% (EC-US), compared to 58.1% (EC) and 91.41% (EC-US) for the Fe electrode. This enhanced performance of Al electrodes can be attributed to the higher release of Al^{3+} ions under increased voltage, which in turn promotes the formation of aluminum hydroxide flocs with greater coagulation capacity (Nassar et al., 2023). According to Faraday's law, at constant current intensity, the amount of metal ions released into the solution is directly proportional to the applied voltage and electrolysis time, leading to improved pollutant removal and the generation of bubbles of hydrogen (H_2) at the cathode (Shahedi et al., 2020). Moreover, the application of ultrasonic waves in the EC-US hybrid process enhances the mass transfer of reactants and facilitates the detachment of gas bubbles from the electrode surfaces through cavitation, thereby maintaining electrode activity throughout the process (Khoramipour et al., 2021). This synergistic effect

significantly boosts the overall treatment efficiency compared to that in the EC process. However, it is important to note that beyond a certain current density threshold, cathode passivation may occur, resulting in a

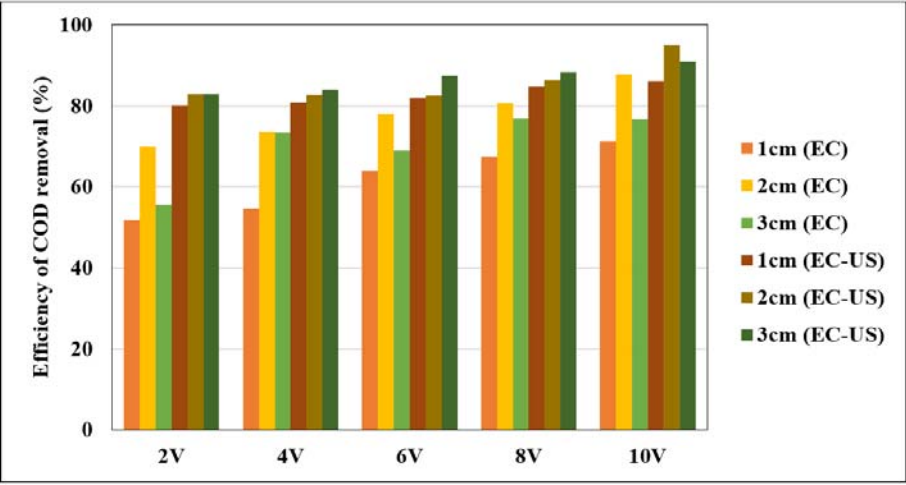
reduction or cessation of anode dissolution. This phenomenon ultimately impairs floc formation and decreases removal efficiency (Huda et al., 2017).



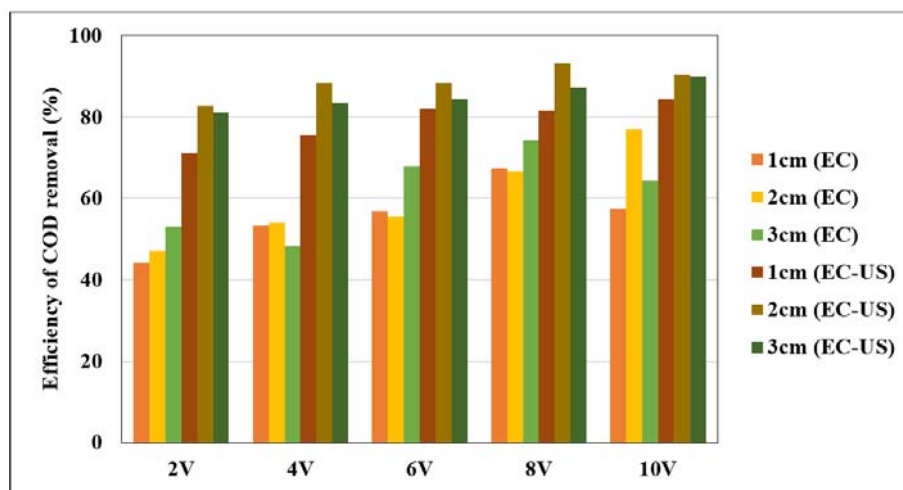
(a)



(b)



(c)



(d)

Figure 6. Effect of voltage on COD removal efficiency using (a) Al in Alor Pongsu landfill; (b) Fe in Alor Pongsu landfill; (c) Al in Pulau Burung landfill; (d) Fe in Pulau Burung landfill; during EC process and EC-US process

Generally, increasing the applied voltage and, consequently the current density, enhances COD removal for both electrode types. Consequently, in this study, the optimal voltage was 10 V. Our findings correspond with those of a previous study conducted by Taib et al. (2021), which indicated that at 10 V and an electrode gap of 0.5 cm, COD removal efficiencies of 43% and 45% were attained using Fe and Al electrodes, respectively, in the EC process. These findings reinforce the direct relationship between applied voltage and COD removal efficiency, as predicted by Faraday's law. Higher current leads to increased dissolution of electrode material, enhancing both charge neutralization of colloids and the formation of metal hydroxide coagulants, thus improving overall treatment effectiveness.

Electrode Weight Loss

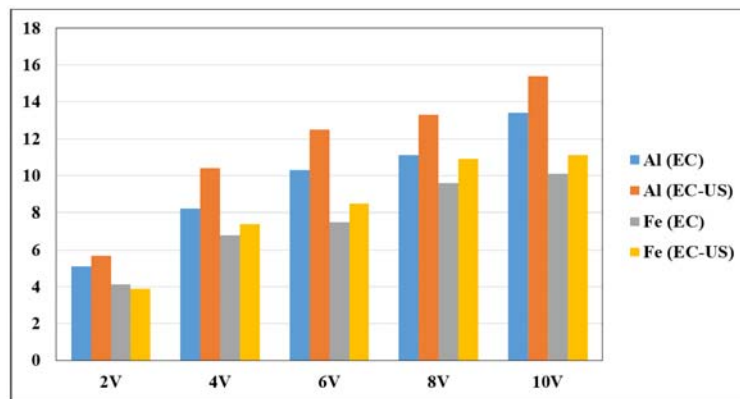
The electrode weight loss observed during both the EC process and the combined EC-US process was evaluated to assess electrode consumption and process efficiency. Experimental results for both Al and Fe electrodes are presented and discussed. This analysis provides insight into the rate of electrode dissolution under different operating conditions, which is critical for understanding the cost-effectiveness and sustainability of the treatment process. The key operating parameters include inter-electrode distance, electrolysis time, and applied voltage. As reported by Shahedi et al. (2020), electrode dissolution is strongly affected by the applied

voltage, with higher voltages leading to increased anodic degradation. In the present study, the focus was placed on the anode, given that electro-chemical reactions between hydroxyl ions and metal atoms at the anode surface result in the dissolution of the electrode and the subsequent release of metal ions that function as coagulants. Therefore, both the EC and the hybrid EC-US processes were investigated to evaluate the influence of these control parameters on electrode consumption.

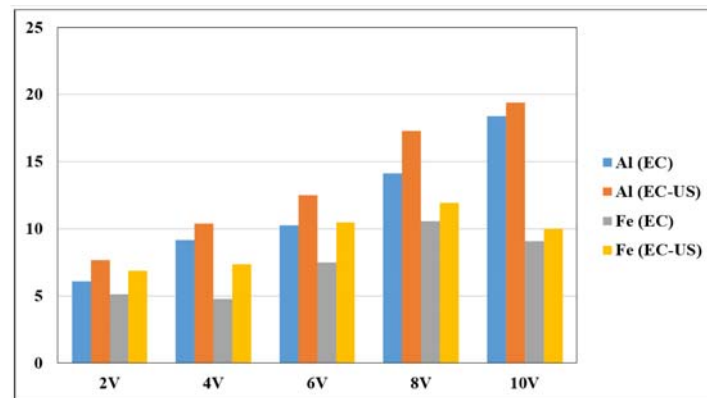
Figure 7 (a)-(c) present the percentage of anode weight loss for Al and Fe electrodes used in the treatment of Alor Pongsu landfill leachate under both treatment processes at inter-electrode distances of 1 cm, 2 cm, and 3 cm. The results revealed a consistent reduction in electrode mass following each experimental run, indicating progressive anode dissolution. An increase in voltage and inter-electrode distance corresponded to higher percentages of weight loss for both Al and Fe electrodes. Notably, the highest anode consumption was observed under the EC-US process at 10 V, with weight losses of 19.4% for Al and 10% for Fe. This behavior aligns with Faraday's law, which states that the mass of electrode material consumed during electrolysis is directly proportional to the total electric charge passed through the system. The dissolution of sacrificial Al and Fe anodes leads to the generation of Al^{3+} and Fe^{2+} ions, which subsequently hydrolyze to form metal hydroxides, the key agents in pollutant coagulation. These metal hydroxides possess the capacity to neutralize negatively charged particles in

solutions, facilitating their aggregation into microflocs. Through van der Waals forces, these micro-flocs

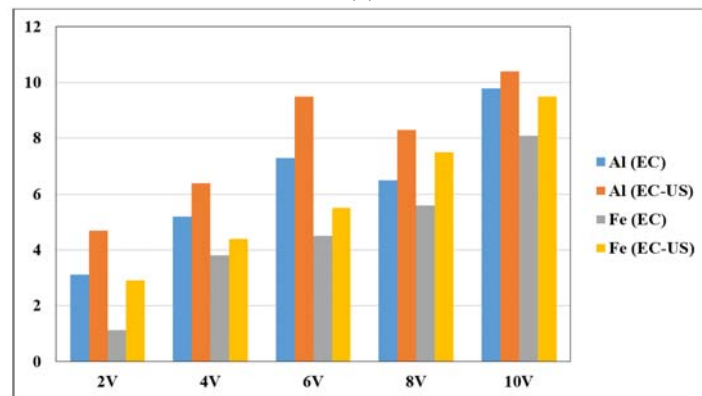
coalesce into larger flocs, enhancing pollutant removal efficiency (Taib et al., 2021).



(a)



(b)



(c)

Figure 7. Percentage of weight loss of Al and Fe electrode as an anode in EC process and combined EC-US process at an inter-electrode distance of (a) 1 cm, (b) 2 cm, (c) 3 cm in Alor Pongsu landfill leachate

Figure 8 (a)-(c) illustrate the percentage of weight loss of the anode for both Al and Fe electrodes used for COD removal in Pulau Burung landfill leachate during the EC process and the hybrid EC-US process. Based on the results, it was observed that the percentage of weight loss of Al and Fe anode was rising from 1 cm to 2 cm,

but subsequently decreased at 3 cm for both EC process and hybrid EC-US process at highest voltage of 10 V. Specifically, in the EC process at inter-electrode distances of 1 cm and 2 cm and voltage of 10 V, weight loss of Al anode increased from 16.4% to 18.4%, while for the Fe anode, weight loss increased from 12.11% to

14.11% over the same distance. However, at 3 cm, the weight loss of the Al and Fe anodes dropped to 16.8% and 13.1%, respectively. A similar trend was observed in the hybrid EC-US process, indicating a consistent relationship between inter-electrode distance and anode dissolution. These findings suggest that an inter-electrode distance of 2 cm represents an optimal

configuration, as it corresponded to both the highest COD removal efficiency and the greatest electrode consumption. The observed decrease in weight loss at 3 cm may be attributed to the reduced efficiency of ion migration across the larger gap, leading to weaker electro-static interactions and slower ion movement (Moradi et al., 2021).

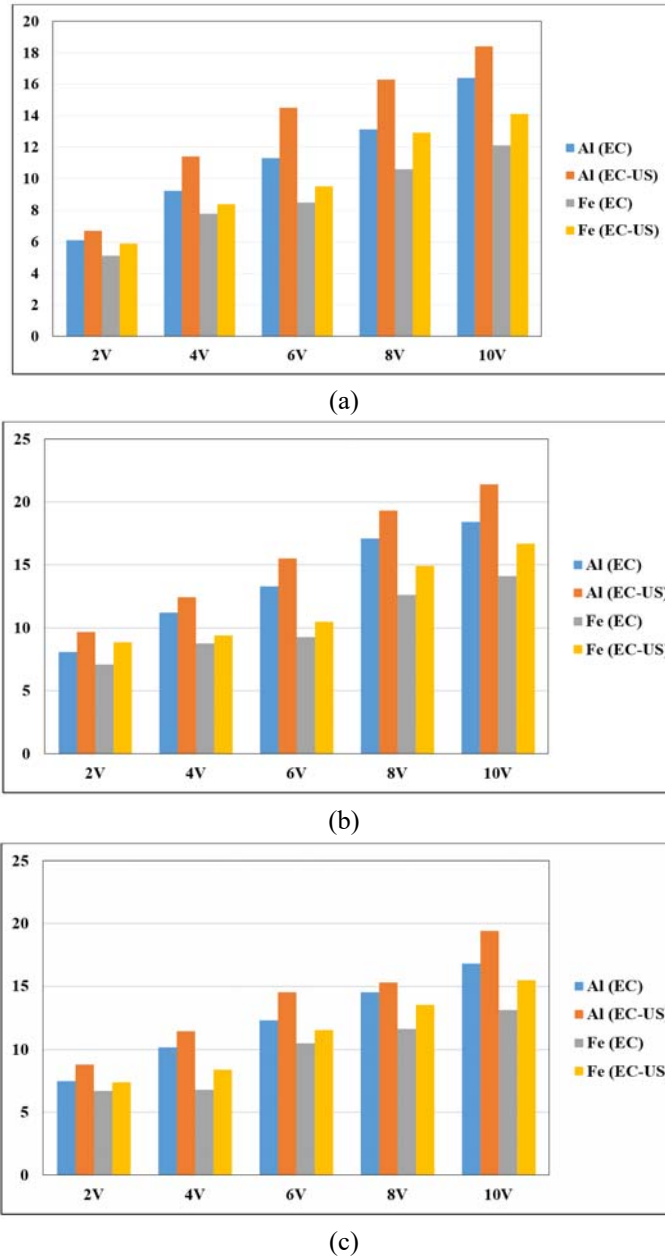


Figure 8. Percentage of weight loss of Al and Fe electrode as an anode in EC process and combined EC-US process at an inter-electrode distance of (a) 1 cm, (b) 2 cm, (c) 3 cm in Pulau Burung landfill leachate

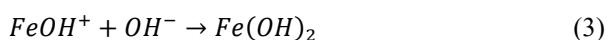
Both the Alor Pongsu and Pulau Burung landfill leachates exhibited the highest anode weight loss for Al and Fe electrodes at an inter-electrode distance of 2 cm

and an applied voltage of 10 V. Notably, across all inter-electrode distances (1 cm, 2 cm, and 3 cm), the percentage of anode weight loss in the EC process was

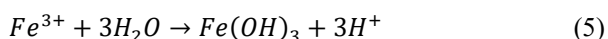
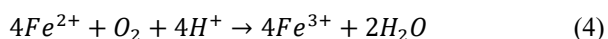
consistently lower than that observed in the hybrid EC-US system. This difference can be attributed to the enhanced energy input from ultrasonic irradiation, which promotes sonolysis of water molecules, resulting in the formation of reactive free radicals and improved ion mobility toward the electrodes. The cavitation effect generated during ultrasonication further increases the dissolution of electrode material, thereby contributing to greater electrode consumption compared to the single EC process (Al-Rubaiey et al., 2018).

The Al electrodes consistently exhibited a higher percentage of weight loss than the Fe electrodes in both treatment configurations. This observation aligns with the findings revealed by Taib et al. (2021), who reported a similar trend during the treatment of sanitary landfill leachate in southern Malaysia. The higher weight loss of Al anodes is likely due to their higher electro-chemical dissolution rate relative to Fe under comparable operating conditions. The previous study also explained the mechanism of Fe dissolution, where Fe^{2+} ions are initially generated *via* anodic oxidation, as described in Equations (1)-(3). These Fe^{2+} species are subsequently oxidized to Fe^{3+} in the presence of dissolved oxygen Equation (4), and the resulting Fe^{3+} ions hydrolyze to form $Fe(OH)_3$, as shown in Equation (5). These sequential reactions highlight the complex transformation of Fe species during the EC process and their role in coagulant formation (Rajaniemi et al., 2021).

At alkaline conditions:

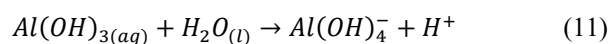
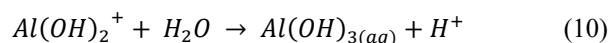
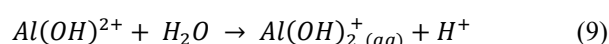
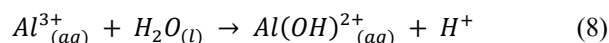
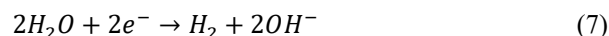


At acidic conditions with the presence of dissolved oxygen:



In contrast to the Fe sacrificial anode, the Al sacrificial anode undergoes direct dissolution to release Al^{3+} ions, as described in Equation (6). Simultaneously, hydrogen gas and hydroxide ions (OH^-) are generated at the cathode, as represented by Equation (7). The overall

mechanism governing the release of Al ions from the dissolution of Al anodes is illustrated in Equations (8)-(11). (El-Ashtoukhy et al., 2020). Upon release, the Al^{3+} ions readily react with OH^- ions present in the aqueous medium to form various monomeric and polymeric hydrolyzed aluminum species.



The superior pollutant removal efficiency observed with Al as the sacrificial anode can be largely attributed to the direct dissolution of Al^{3+} ions during the EC process. This efficiency is closely related to the favorable flocculation and settling properties of aluminum hydroxide $Al(OH)_3$, which forms denser and more readily separable flocs than iron hydroxide $Fe(OH)_3$ (Taib et al., 2021). Previous studies supporting this observation also found that Al electrodes achieved higher COD removal efficiencies compared to Fe electrodes (De La Luz-Pedro et al., 2019; Soomro et al., 2020). In the current study, Al demonstrated superior performance over Fe in both COD reduction and operational effectiveness, indicating its greater suitability for treating landfill leachate from the Alor Pongsu and Pulau Burung sites. Experimental data confirmed that both Al and Fe anodes experienced measurable weight loss throughout the EC process, with variations depending on the inter-electrode spacing (1 cm, 2 cm, and 3 cm). A positive correlation was observed between applied voltage and anode dissolution, with the greatest weight losses recorded at 10 V for both EC and hybrid EC-US systems. This trend underscores the influence of energy input on the rate of electrode consumption. Additionally, the hybrid EC-US process consistently resulted in greater anode weight loss compared to the conventional EC system. This enhancement is attributed to the synergistic action of ultrasonic cavitation, which generates localized

turbulence and acoustic streaming. These phenomena intensify mass transfer and accelerate the electro-chemical reactions between metal ions and hydroxide ions at the electrode surface (Hassani et al., 2022; Zanki et al., 2020), thereby promoting more rapid anode dissolution. Moreover, under identical operating conditions, Al anodes exhibited a higher percentage of weight loss than Fe anodes in both treatment approaches. This finding not only confirms the higher electro-chemical reactivity of Al, but also suggests its greater capacity for generating coagulant species during treatment. Thus, Al electrodes offer distinct advantages in terms of both treatment performance and material reactivity, making them a more effective choice for landfill leachate remediation.

CONCLUSIONS

This study successfully characterized landfill leachate from Alor Pongsu Landfill and Pulau Burung Sanitary Landfill, revealing that the COD levels in both samples exceeded the discharge limits set by the Malaysia Environmental Quality Act (MEQA) 1974, highlighting the need for effective leachate treatment. The performance of EC and hybrid EC-US processes was evaluated using Al and Fe electrodes under various operating conditions. A comparison between the EC and the hybrid EC-US processes revealed that the integration of ultrasound significantly enhanced COD removal efficiency. The EC-US process attained a high removal efficiency of 95.05% and 96.31% for the Alor Pongsu Landfill and Pulau Burung Sanitary Landfill, respectively, using Al at 25 minutes. This improvement can be attributed to the synergistic effect of ultrasonic irradiation, which enhances mass transfer, improves the dispersion of coagulants, and disrupts the structure of organic pollutants, making them more accessible for coagulation. Despite the difference in removal

performance, both processes shared the same optimum operating conditions: 10 V applied voltage, 2 cm inter-electrode distance, and 25 minutes of treatment time, demonstrating that the additional hydroxyl radicals ($\bullet\text{OH}$) are the key factor contributing to the enhanced treatment efficiency in the hybrid system. Al electrodes proved to be more efficient than Fe electrodes in both processes. Additionally, the highest anode weight loss occurred under optimum conditions in the hybrid EC-US process, indicating enhanced coagulant generation. Overall, the integrated EC-US system demonstrated superior effectiveness and potential as an advanced treatment method for the removal of COD from landfill leachate. However, certain limitations should be considered. The work was carried out under controlled laboratory-scale conditions. Therefore, further research at pilot scale or full scale is recommended to assess long-term performance, and energy consumption. In addition, more research work is needed to prevent or reduce the weight loss of anode in single EC process and hybrid EC-US process.

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Data Availability

The data presented in this study is available in the article.

Conflict of Interests

The authors have no conflict of interests to declare.

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