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Enhanced removal of mercury from leachate using electrocoagulation: Reaction kinetics

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ABSTRACT

Leachate treatment represents a key problem related to landfill operations, a liquid waste generated from a mixture of rainwater infiltration, waste decomposition, and surface runoff. Mercury is among the hazardous contaminants commonly found in landfill leachate. The use of electrocoagulation has shown potential in eliminating mercury from this leachate. This study aimed to investigate the impact of varying electric voltages (4 V, 8 V, and 12 V), contact times (15, 30, 45, and 60 minutes), and electrode spacing (1 cm and 2 cm) on the efficiency of mercury removal during a 60-minute electrocoagulation process. It was found that the most effective mercury removal, with an efficiency of 99.32%, occurred at 12 V with a 60-minute treatment duration and a 1 cm gap between electrodes. An ANOVA analysis revealed that 86.1% of the influence of the investigated variables on mercury reduction was statistically significant. Furthermore, data from the electrocoagulation process were utilized to examine the effects of operational parameters, evaluated to determine the most appropriate adsorption kinetics and isotherm models. The pseudo-second-order kinetic model and the Freundlich isotherm exhibited the highest correlation with the experimental findings. Among the models evaluated, the kinetic model presented the strongest correlation with the experimental results, as evidenced by a high correlation coefficient ($R^2 = 0.9895$).

Keywords: electrocoagulation, mercury, adsorption kinetics, adsorption isotherm, aluminum.

INTRODUCTION

Leachate typically contains a high concentration of suspended solids, which necessitates the use of coagulants to bind these particles together. Originating from the infiltration of rainwater or surface runoff into waste deposits, leachate dissolves numerous organic and inorganic substances, resulting in a liquid with extremely high pollutant levels. These pollutants often include significant concentrations of organic matter, which can be quantified through parameters such as Biochemical Oxygen Demand (BOD), ammonia, and various heavy metals (Hasyyati et al., 2020; Nurfahasdi et al., 2024a; Nurfahasdi et al., 2024b; Nurfahasdi et al., 2024c). According to Mahmad et al. (2016), leachate is generated through the decomposition of waste when infiltrated by external water sources, leading to a mixture rich in organic contaminants and heavy metals. The discharge of leachate containing elevated mercury concentrations poses serious risks to aquatic ecosystems and food chains. Certain forms of mercury, once deposited in sediment, may undergo

methylation and become concentrated in aquatic organisms such as fish and benthic species. The procedure is regulated by parameters specific to the site, which encompass mercury speciation and the existing geochemical conditions and the availability of labile organic matter (Korejwo et al., 2022; Eckley et al., 2020; Hsu-Kim et al., 2018; Driscoll et al., 2013). Researchers and engineers continue to face challenges in designing mercury removal technologies that are efficient, cost-effective, and capable of handling different mercury species – dissolved, elemental, and nanoparticulate. Mercury is a non-essential and toxic heavy metal often found in landfill leachate, and human exposure can result in central nervous system (CNS) disorders (Said, 2010). In Indonesia, leachate treatment commonly relies on pond-based systems such as retention ponds, anaerobic and aerobic ponds, and stabilization ponds. However, these methods are limited by long hydraulic retention times – typically 30 to 50 days – and the need for extensive land area (Herlina et al., 2021).

Electrocoagulation (EC) integrates both electrochemical and conventional coagulation–flocculation mechanisms. The process operates on principles similar to those of an electrolytic cell, which consists of two electrodes—an anode and a cathode. During electrocoagulation, the anode serves as a source of coagulant through oxidation, while reduction reactions at the cathode generate hydrogen gas. These gas bubbles aid in lifting suspended particles or flocs that would otherwise remain settled within the system (Hanum et al., 2015). As noted by Attour et al. (2014), the success of electrocoagulation relies on a range of operational parameters, including electrode characteristics, electrode distance, applied voltage, and pH of the solution. Commonly, aluminum or iron is used as the anode, where oxidation reactions release coagulant species, while reduction takes place at the cathode (Suprihatin & Aselfa, 2020; Tibebe et al., 2019; Ascon, 2020). Previous investigations have shown that electrocoagulation is an effective method for removing both dissolved ionic substances and heavy metals from wastewater. A key advantage of this method lies in the continuous generation of coagulants directly within the contaminated medium. Coagulants generated in situ play a significant role in enhancing the effectiveness of pollutant removal for different types of contaminants. Furthermore, electrocoagulation presents a promising alternative to conventional wastewater treatment systems by offering a more sustainable and economically feasible solution with fewer limitations.

Adsorption kinetics refers to the time-dependent process through which substances adhere to the surface of an adsorbent. The rate at which adsorption occurs is typically characterized by the adsorption rate constant (k) and the reaction order, as described by various kinetic models. To evaluate the equilibrium of the adsorption process, models such as the Langmuir and the Freundlich isotherms are frequently employed. These models help in identifying the most appropriate representation of the adsorption behavior (Hamayani & Faisal Anwal, 2016).

While electrocoagulation has demonstrated efficacy in removing various heavy metals, the specific kinetics and equilibrium mechanisms governing mercury (Hg) removal from complex matrices like landfill leachate using aluminum electrodes remain insufficiently explored. Many studies focus on removal efficiency, but a comprehensive analysis linking operational parameters to the underlying adsorption kinetics and isotherms is needed for process optimization and scaling. Therefore, this study aims to explicitly investigate the influence of applied voltage and electrode spacing on mercury removal efficiency from real landfill leachate. Furthermore, the research seeks to identify the most appropriate kinetic and isotherm models to describe the adsorption behavior of mercury during the electrocoagulation process, thereby providing deeper mechanistic insights and validating the process's applicability.

RESEARCH METHODS

Experimental set-up

This research employed a batch electrocoagulation process using two aluminum (Al) electrodes (purity >99.5%) in a monopolar configuration. The electrodes, with dimensions of 15 cm × 5 cm × 0.06 cm (length × height × thickness), were immersed to an effective area of 10 cm × 5 cm per side, providing a total effective surface area of 100 cm² per electrode (200 cm² total). The leachate sample was added to the reactor and exposed to electrocoagulation under the determined conditions of voltage and contact time. The sampling procedure adhered to SNI 6989.59:2008, which outlines the Standard Method for Wastewater

Sampling. The independent variables in this study included electric voltage (4 V, 8 V, and 12 V) and electrode distance (1 cm and 2 cm). The dependent variable was the volume of wastewater treated, set at 2000 mL, with sampling intervals at 15, 30, 45, and 60 minutes. The electrodes used had dimensions of 10×4 cm with a thickness of 0.6 mm. Primary data were obtained by measuring the mercury concentration in the leachate before and after the electrocoagulation process. A DC power supply was employed as the power source. In addition, nitric acid (HNO_3) was added to adjust the pH to below 2 in order to prevent changes in mercury concentration. Mercury content was determined using Inductively Coupled Plasma (ICP) at the Environmental Health Engineering Center (BTKL) in Medan. The purpose of the data analysis was to investigate the role of operational parameters such as electric voltage, contact time, and electrode distance on the performance of mercury (Hg) removal. Experimental data analysis was performed using multiple linear regression, while adsorption kinetics and isotherm models were applied to clarify the mercury removal mechanism in electrocoagulation.

Kinetics study

Batch experiments were performed to assess the adsorption kinetics of Hg(II). A series of 50 mL Pyrex glass tubes was prepared, each containing a specific volume of reagent solution, and kept in a shaking water bath maintained at a controlled temperature. Once the desired temperature was achieved, 0.01 g of activated carbon was added to each tube. The mixtures were then agitated using a mechanical shaker to ensure proper contact between the adsorbate and adsorbent. At predetermined time intervals, samples were taken, and the solutions were separated from the adsorbent to analyze the remaining Hg(II) concentration in the solution. These measurements were used to calculate the adsorption capacity at each time point.

The equilibrium metal adsorption capacity (qe) and the removal efficiency percentage were determined using Eqs. (1) and (2), respectively.

$$q_e = V\,(C_i - C_e)/M \quad (1)$$

$$Removal\ (\%) = (C_i - C_e)/Ci \times 100 \quad (2)$$

where: C_i and C_e represent the initial and equilibrium concentrations of metal ions in ppm, respectively; V denotes the volume of the solution in liters, and M stands for the mass of the adsorbent in grams.

The data from the experiment were analyzed by applying to linear forms of commonly used adsorption isotherms and kinetic models, as outlined in Table 1. In these models, qt (μg/g) represents the level of Hg(II) uptake at a particular time, with qe (μg/g) showing the amount adsorbed at equilibrium. The constant k_1 (min^{-1}) represents the pseudo-first-order rate constant, while k_2 (g/μg·min) denotes the pseudo-second-order rate constant. Ce (μg/L) represents the concentration of Hg(II) at equilibrium in the solution. For the isotherm models, KF (L/μg) indicates the adsorbent's adsorption capacity, and bF stands for the heterogeneity factor linked to surface energy variations (Freundlich model). Meanwhile, KL (L/ μg) indicates the solute's affinity for the adsorbent, and aL (L/μg) reflects the energy of adsorption (Langmuir model). These parameters were used to evaluate which model best described the adsorption behavior of mercury onto the adsorbent material.

Equilibrium adsorption capacity q_e(μg/g), adsorption capacity at time t, q_t(μg/g) and removal efficiency R (%) are calculated as follows:

$$q_e = (C_0 - C_e) / m \times V \quad (3)$$

$$q_t = (C_0 - C_t) / m \times V \quad (4)$$

$$R = (C_0 - C_e) / C_0\,\% \quad (5)$$

where: C_0 (μg /L) and C_e (μg /L) represent the initial and equilibrium concentrations of Hg (II) solution, respectively; C_t represents the concentration of time t; m(g) is the quality of adsorbent while V (L) is the solution volume.

The pseudo-first-order and pseudo-second-order kinetic models were selected for this analysis as they are the most prevalent models for elucidating adsorption mechanisms. The pseudo-first-order model is often applicable in the initial stages of adsorption, while the pseudo-second-order model is typically indicative of a chemisorption process, which is rate-controlling. Fitting experimental data to these models helps determine the nature of the adsorption process and the potential rate-limiting steps involved in the electrocoagulation of mercury.

Table 1. Adsorption models for kinetics analysis

Kinetics models	Type	Plot (y vs x)	Constants
Pseudo first order			
$ln(q_e - q_t) = lnq_e - k_1 t$	Linear	$ln(q_e - q_t)$ *vs t*	k_1 (min^{-1})
Pseudo second order			
$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$	Linear	$\frac{t}{q_t}$ *vs t*	k_2 (g/µg.min)
Isotherm models	Type	Plot (y vs x)	Constants
Langmuir			
$\frac{1}{q_e} = \frac{1}{K_L C_e} + \frac{a_L}{K_L}$	Linear	$\frac{1}{q_e}$ *vs* $\frac{1}{C_e}$	K_L (L/µg) a_L (L/ µg)
Freundlich			
$logq_e = b_F logC_e + logK_F$	Linear	$logq_e$ *vs* $logC_e$	K_F (L/g) b_F (dimensionless)

RESULTS AND DISCUSSION

The water utilized for this research was gathered from the inlet of the landfill storage pond. Table 2 illustrates the properties of the collected leachate.

The experimental results were acquired by adjusting key parameters in the electrocoagulation process, including voltage (volts), contact time (minutes), and distance (cm). These results are illustrated in Figure 1.

Figure 1 shows the impact of the interaction between electrodes, duration, and voltage on the effectiveness of mercury removal during electrocoagulation. The maximum efficiency of mercury removal from the electrocoagulation process is 99.32% at 12 volts, 60 minutes, with an electrode spacing of 1 cm, and the lowest mercury reduction effectiveness is 39.91% at 4 volts treatment, 15 minutes, and 2 cm distance.

Effects of electrode distance, contact time, and voltage on mercury level reduction

A higher applied voltage increases the current density, which in turn accelerates the anodic

Table 2. Leachate characteristics

No	Parameter	Unit	Concentration	Standard*	Description
1	Mercury (Hg)	µg/l	0.0401	0.005	Exceed

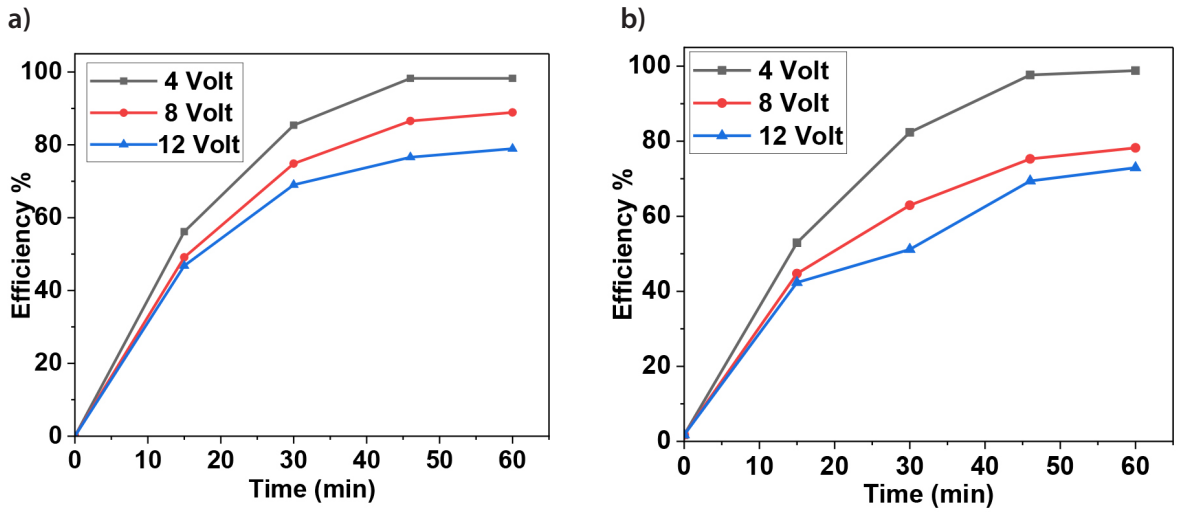

Figure 1. Experimental results (a) distance 1 cm and (b) distance 1.5 cm

dissolution of aluminum according to Faraday's law ($Al \rightarrow Al^{3+} + 3e^-$). The increased release of Al^{3+} ions leads to the rapid in-situ formation of polymeric aluminum hydroxides (e.g., $Al(OH)_3$), which are effective coagulants for the sweep flocculation and enmeshment of mercury species. Concurrently, the enhanced current promotes the generation of smaller and more numerous hydrogen bubbles at the cathode ($2H_2O + 2e^- \rightarrow H_2 + 2OH^-$), improving the flotation of the formed flocs. Thus, the efficiency obtained is greater (Puji et al., 2017). Besides regulating the dosage of the coagulant, the electric voltage also influences the production rate, floc formation, and bubble size, all of which contribute to the removal efficiency of the electrocoagulation process (Bazrafshan et al., 2012).

The time needed to produce metal hydroxides and finalize pollutant coagulation is known as the electrocoagulation duration. This duration is a key factor in the electrocoagulation process. The influence of this treatment time on the overall mercury removal is shown in Figure 3. As indicated in Figure 3, the efficiency of mercury removal increased with longer treatment times. According to Said (2010), the leachate's metal content decreases with the length of the electrocoagulation process. The greater the electrocoagulation time, the more time is required to release the coagulant. Thus, more flocs were produced. This is according to Faraday's law, that states that, the longer the electrocoagulation process, the more coagulants are formed. However, at 45 to 60 min, there was a decrease in the leachate's ability to eliminate mercury effectively. The observed decrease in efficiency after 45 minutes is attributed to electrode passivation, where a layer of oxide or adsorbed flocs forms on the anode surface, inhibiting further dissolution of aluminum ions. Additionally, the re-stabilization of flocs due to excessive mixing over extended periods could also contribute to the slight decline in removal efficiency. This is according to the research results by Masrulitta et al. (2021) and Nurfahasdi et al. (2024a) which states that, saturation occurs on the electrode plate used. This saturation is due to the increasing number of flocs that block the formation of coagulants, so that the efficiency also decreases.

Mercury concentration reduction depends on time, voltage, and electrode spacing. The distance between electrodes controls the electrostatic charge distribution between the anode and cathode, significantly impacting the electrocoagulation process (Setyawati et al., 2021; Farouk et al., 2022; Karichappan et al., 2014; Ahangarnokolaei et al., 2018). Figure 4 shows that a distance of 1 cm was more efficient than a distance of 2 cm. According to Trisnaawati and Purnama (2021), Time, voltage, and the separation between electrodes collectively affect mercury removal efficiency. The spacing modulates the electrostatic interactions between the anode and cathode, which is critical in the electrocoagulation process (Bouguerra et al., 2015). This occurs because the greater the distance between the electrodes, the greater the resistance, so that the production of coagulant in this case $Al(OH)_3$ will also decrease, which causes the efficiency of reducing contaminants to decrease. The contaminant

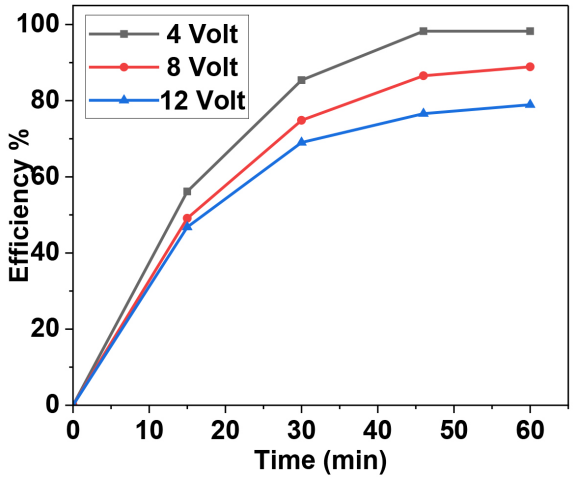

Figure 2. Effect of voltage on lowering mercury levels

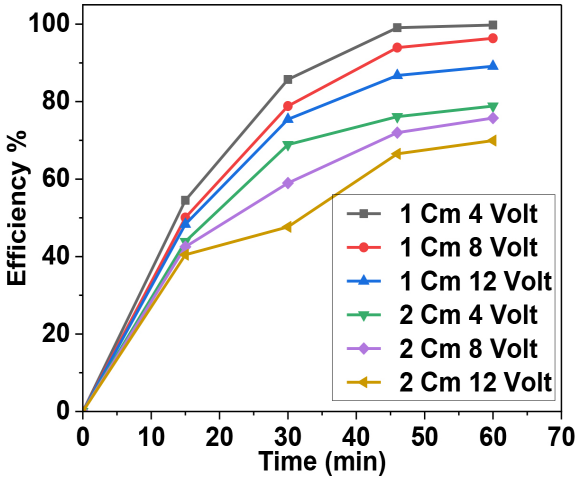

Figure 3. Effect of contact time on reducing mercury levels

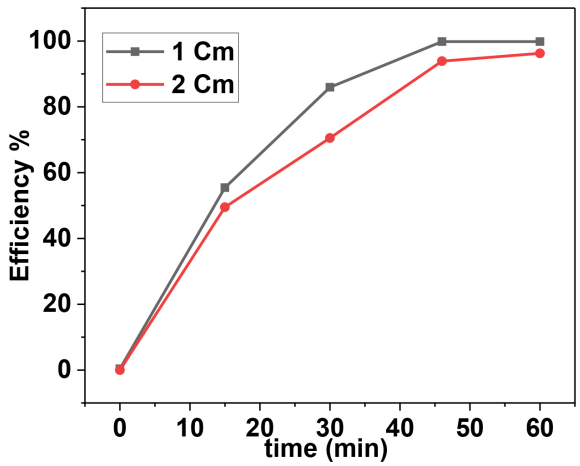

Figure 4. Effect of electrode distance on the decrease in mercury levels

reduction efficiency increases as the electrode distance shortens (Bouguerra et al., 2015; Chen et al., 2018). The reduced electrode spacing aids ion migration by minimizing resistance.

Table 3 demonstrates that the adjusted R-squared value stands at 0.861, indicating that 86.1% of the variation in mercury removal efficiency from leachate is concurrently affected by voltage, time, and electrode spacing. The remaining 13.9% are other variables that affect the electrocoagulation process, such as variations in electrodes. Reaction kinetics is the quantitative study of chemical reaction rates and the variables influencing these rates. The rate of a chemical reaction is the number of moles of reactant per unit volume that reacts in a given unit of time. In every electrochemical reaction such as electrocoagulation, electroplating, electrorefining the factors that affect the reaction are the current strength and density which will control the reaction rate (Largitte & Pasquier, 2016; Kim et al., 2021). If a curve is constructed to decrease the concentration of reactants as a function of time, a curve will be obtained in which the slope of the curve at each point is always negative because the concentration of reactants always decreases. Therefore, the reaction rate at any point along the curve is = − dC/dt. However, if the reaction rate is written as the product formation rate, the reaction rate will be positive. The total reaction order is calculated by summing the exponents of the reactant concentrations in the differential rate equation. Theoretically, the order of the reaction is a small integer, but in some cases, it is a fraction or zero. Generally, the reaction order for a particular substance is not the same as that in the stoichiometric reaction equation (Simonin, 2016; Ilhan et al., 2019).

Adsorption kinetics and isotherm

Adsorption is the process by which a solid or liquid adsorbent absorbs a gas or liquid adsorbate, which can then accumulate as a layer on the adsorbent's surface (Largitte & Pasquier, 2016). The adsorption kinetics provided insight into the temporal rate of adsorbate removal by the adsorbent. Based on the Table 3, It is discovered that the pseudo-second order R^2 value surpasses that associated with the pseudo-first-order kinetic model. The result shows that the data closely resembles the second pseudo-adsorption kinetics model, specifically the R2 > 0.9 value. Figure 5 shows the R2 value of each equation, which is used to determine the compatibility of data with a linear function and shows the level of linearity of a graph (Simonin 2016). A value's fit to the available data is greater the closer it is to one.

The adsorption isotherm explains how the adsorbent's (an aluminum plate) performance and the adsorbate interact during the adsorption process (Ayawei et al., 2017; Loez-Luna et al., 2019; Di et al., 2022; Alotaibi et al., 2023). In this study, the adsorption behavior was examined using both the Freundlich and Langmuir isotherm models. Analysis of the linear plot revealed that the Freundlich model yielded a regression coefficient (R^2) of 0.607. Given its closer proximity to 1 compared to the Langmuir model, this suggests that the Freundlich isotherm offers a more accurate representation of the adsorption process under the studied conditions Freundlich's method presumes that adsorption creates many layers and that the adsorbent surface is heterogeneous. This permits unrestricted movement of the adsorbate until the adsorption process takes place, which

Table 3. Model summary

Model	R	R Square	Adjusted R Square	The Error in the Estimate
1	0.938[a]	0.879	0.861	6.94

Note: a – predictors: (constant), electrode distance (cm), contact time (min), voltage (V) (SPSS, 2022).

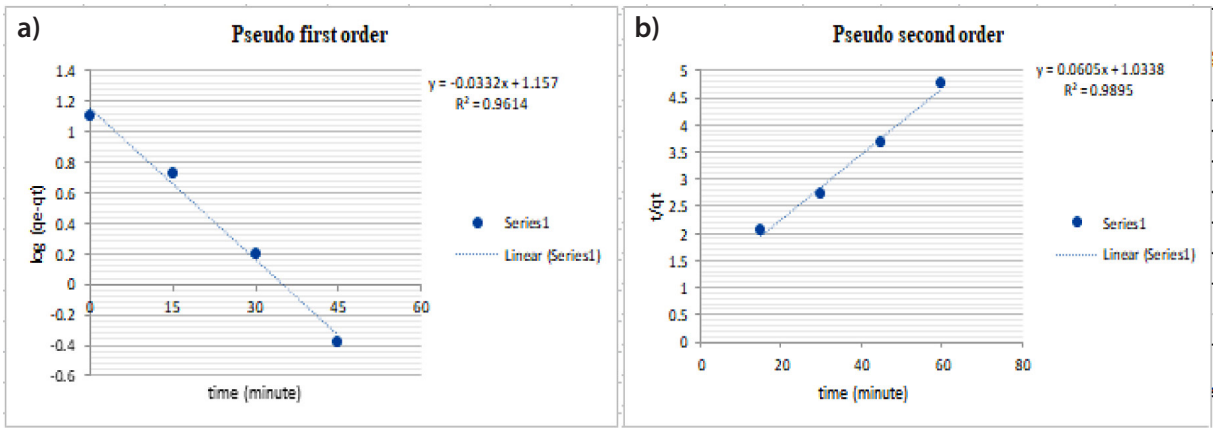

Figure 5. Linear chart of adsorption kinetics on (a) pseudo first order and (b) pseudo second order

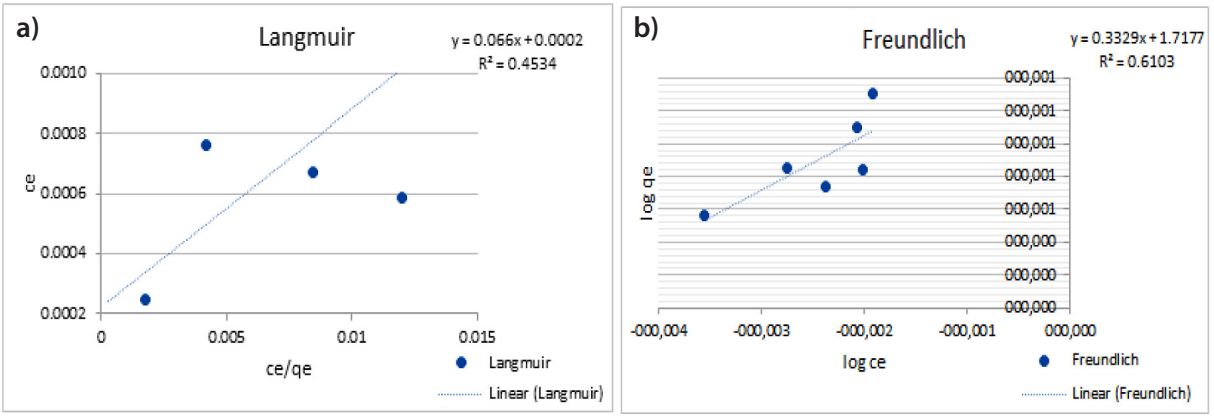

Figure 6. Linear (a) Langmuir adsorption isotherm and (b) Freundlich adsorption isotherm

happens in numerous physical adsorption layers (Syafiqah Imla & Yussof, 2018).

Numerous research efforts have employed kinetic modeling to examine the adsorption of heavy metals during the electrocoagulation (EC) process. For example, Vasudevan et al. (2010) explored the removal of cadmium (Cd) through EC and evaluated the applicability of first- and second-order kinetic models, finding that the second-order model produced higher correlation coefficients. In a related study focusing on the removal of Hg, Pb, and Ni using a magnesium anode, Shaker et al. (2023) indicated that the adsorption mechanism for these three metal ions onto $Mg(OH)_2$ was more accurately represented by the pseudo-second-order model. This kinetic model also exhibited a strong fit in another study involving EC with iron electrodes for the removal of heavy metals. Similarly, multiple studies (Zhang et al., 2020; Chen et al., 2018) have assessed the relevance and applicability of these models. Consequently, it is possible to use a range of models to explain the heavy metal adsorption phase in EC. This could be explained by the fact that the nature, characteristics, and operating conditions of the contaminants and coagulant ions all affect their interactions.

CONCLUSIONS

Time, voltage, and distance affect the concentration and efficiency of mercury removal by electrocoagulation. The mercury removal efficiency from the leachate was 86.1%. The adsorption of mercury (Hg) on the aluminum plate adheres to a pseudo-second-order kinetic model, while the Freundlich isotherm effectively characterizes the adsorption equilibrium. This study revealed that the straightforward EC process design, in conjunction with Al electrodes and carefully chosen voltage, time, and electrode distance, effectively removes mercury from wastewater. It is crucial to take into account the amount of mercury that

remains in the effluent after treatment, even if a high removal efficiency is attained. The examination of concepts and discoveries uncovers a knowledge gap that underscores the need for continuous research to advance the field. Furthermore, the practical aspects of electrocoagulation and other pertinent factors require scrutiny to assess their operational efficiency.

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