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Full Paper

Optimized Cost-Effective Electrocoagulation Reactor Cell Designed with Effective Parameters for The Removal of Cadmium from Actual Landfill Leachate Samples

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Abstract- In this research, an innovative cost-effective electrocoagulation (EC) reactor cell was designed with optimized effective parameters for effectiveness of EC technique in the removal of cadmium (Cd) from landfill leachate samples. This was achieved by employing cheap aluminum and stainless steel plates as suitable electrodes, and using solar cells for power, which not only made the system more sustainable but also aided in saving operational costs. The study also discovered that the efficiency of Cd removal from a landfill leachate sample was improved by controlling the current density, adjusting the pH of the solution, optimizing the concentration of supporting electrolyte, and optimum fixing the distance between the electrodes. The results suggested that the optimal operating conditions for the removal of Cd from the raw leachate were current density of 6.0 A/m2, duration time of 40 min, concentration of supporting electrolyte of 2000.0 mg/L NaCl, and the initial pH 8.5. The laboratory experiments showed that the treated samples were free of Cd in all trials, with a removal rate of about 99%. This approach provides a scalable, relatively low-cost method for tackling Cd pollution, an issue that is a big deal given Cd's toxicity and the strict regulatory limits on its discharge.

Keywords- Cadmium; Electrocoagulation; Wastewater; Landfill leachate; Pollution

1. INTRODUCTION

The escalating growth of the global population and the demand for energy and food have rendered wastewater recycling and resource recovery a pressing global necessity [1]. The provision of clean water to the majority of individuals worldwide has become a formidable challenge for humanity, particularly in underdeveloped nations [2]. The majority of industrial wastewaters are contaminated with heavy metal species, which, due to their toxicity, can have a severe impact on the environment through bioaccumulation, even in minute quantities [3,4].

In general, heavy metals are classified as elements with atomic weights ranging from 63.5 to 200.6 and a gravity value greater than 5.0 [5]. The pollution caused by these metals is currently considered to be one of the most serious environmental problems. Unlike organic pollutants, heavy metals are not biodegradable and tend to accumulate in living organisms. The ions of these metals are highly toxic and can lead to fatal diseases such as cancer. Heavy metals, including copper (Cu), nickel (Ni), zinc (Zn), lead (Pb), cadmium (Cd), and chromium (Cr) or their compounds, are commonly utilized by many mining, metal finishing, and chemical industries, resulting in an increase in water pollution. The presence of any of these toxic metals in water bodies can have undesirable consequences [5,6].

Cd, a widely spread toxic pollutant for humans, animals, and plants, is primarily released into the environment from various industrial emissions and human activities such as the application of chemical fertilizers, herbicides, and pesticides or irrigation with polluted groundwater, and eventually enters the food chain [7]. Exposure to Cd can seriously affect the function of the nervous system, leading to health issues such as Parkinson's disease and learning disabilities [8,9]. Cd exposure can also damage the kidneys, which is generally manifested as a tubular disfunction detected by increased excretion of molecules with low weight proteins such as β_2-microglobulin and α_1-microglobulin. This heavy metal has been reported as one of the causes of prostate cancer. The Cd(II) ion has a relatively long half-life ranging from a few years to tens of years and has serious impacts on human bone. Chronic exposure to Cd wastewater can target organs [10,11].

The treatments of industrial effluents involve various techniques, including ion exchange [12], membrane processes [13], reverse osmosis [14], physical-chemical and biological [15], and electrochemical techniques [16]. Conventional biological treatment processes are time-consuming, require significant operational space, and are ineffective in treating effluent containing toxic elements. Advanced oxidation techniques, which are typically utilized to obtain high-purity water, are expensive. Chemical coagulation is a method that is not expeditious and produces an excessive amount of sludge. Recently, EC has garnered much attention as a versatile and environmentally-friendly technique for treating industrial effluent [16–19]. EC is an electrochemical technique that removes various contaminants, particularly metal cations, from wastewater or groundwater without requiring additional chemical supplements. Moreover, this method reduces the amount of produced sludge. Some studies

have demonstrated the financial advantage of this technique over conventional methods [20, 21].

In this research, we plan to develop a basic system that uses very little energy to effectively remove Cd(II) from wastewater and other polluted water sources. This led us to create an electrochemical cell system based on the electro-coagulation (EC) method. By using solar cells for power, along with a straightforward design and cost-effective reactor cell components, this project includes several innovative features. Utilizing solar energy not only makes the system more sustainable but also helps in reducing operational costs. The EC method utilizing inexpensive stainless steel and aluminum electrodes effectively coagulates and eliminates Cd(II) metal ions, thereby serving as a viable strategy for purifying polluted water. The following sections will provide detailed information on the technical aspects, experimental setup, and expected environmental advantages of this method.

2. EXPERIMENTAL SECTION

2.1. Real Leachate sampling

Tehran, the capital city of Iran, is home to a population of 16 million individuals who produce a daily output of 8000 tons of waste. The waste is collected from designated stations and subsequently transported to the Kahrizak waste disposal site located in the southern region of Tehran. The Kahrizak landfill site generates approximately 670 m^3 of leachate on a daily basis. Landfill leachate, which is produced by municipal solid waste landfill sites, is widely recognized as a hazardous and heavily polluted substance. The effluent lake area of Kahrizak, which was established 63 years ago, spans nearly 12 hectares. In the present study, leachate samples were collected from the Kahrizak landfill site and stored in four clean plastic jars, each filled with 2.5 liters of leachate. The samples were then transported to the laboratory for analysis. The initial tests were conducted using a required amount of the samples, while the remainder was stored at a temperature of 5.0 ℃ in a refrigerator. Figure 1 provides a visual representation of Kahrizak Lake.

Figure 1. Kahrizak Lake, the landfill leachate collection site of Tehran Iran

Primarily, some physico-chemical properties of the leachate were tested to find more knowledge about the sample contents that may affect the EC process of Cd(II) removal. The results are outlined in Table 1.

Table 1. Some properties of the leachate from the Kahrizak landfill in Tehran province

Parameter	Value (mg/L)
Cd	2.36
Cr	2.17
Pb	2.07
Zn	1.29
pH	6.7
BOD5	5014
COD	55400
BOD	640
Cyanide	0.01<
Color	black
Electrical conductivity	38.3 (DC/m), 25 ºC

BOD5: Biochemical Oxygen Demand over 5 days; COD: Chemical Oxygen Demand

2.2. Synthetic wastewater

To generate a synthetic wastewater, $CdCl_2$ (Merck, Germany) was dissolved in tap water, with a concentration of 100.0 mg/L of Cd utilized in all experiments. Prior to conducting the experiments, the pH of the electrolyte was adjusted by adding solutions of either NaOH (0.1 M) or HCl (0.1 M) (Merck, Germany) solutions, depending on the desired pH value. Additionally, NaCl (Merck, 99.9%, 2000.0 mg/L) was added as a supporting electrolyte. Prior to each trial, one liter of the solution was introduced into the cell and the current density was adjusted as necessary.

2.3. Materials and equipment

The present study employed an EC reactor cell, which was constructed using cubic glass measuring 30×25×20 cm^3. The reactor cell was equipped with three tanks and two outputs, one for the discharge of sludge and the other for the outflow of refined water. The electrodes utilized in the reactor cell were made of aluminum and stainless-steel plates, with dimensions of 15×10×0.2 cm^3 and 15×10×0.1 cm^3, respectively. The stainless-steel plates were procured from a reputable local market in Tehran, while the aluminum plates, with a total purity of 98%, were manufactured by Arak Alopars company (Iran). A battery BS1245 as the electricity

energy storage from Taiwan, a solar panel Yingli solar of China as the electricity power source, an aeration Pump device SPL-181-3 from Iran for aeration of the reactor cell, a DC/AC inverter NO.TBE 36177120 (China) as the electricity inversion, were employed. Furthermore, a DC power supply Ryi3005-3D from China (0 - 10 A) and (0 - 60 V) was also applied to the set as the regulating power supply. A comprehensive list of all the pertinent characteristics of the EC reactor cell is provided in Table 2. An atomic absorption spectrometer (1570 Shimadzu, Japan) (AAS) was utilized to measure metal contents of the samples.

Table 2. Properties of the electrochemical reactor cell used

Characteristics of the reactor cell	**Description**
Material	Glass
Anode	Stainless steel
Cathode	Aluminum
Electrode distance	0.5 cm
Current density range	0.0–10.0 A
Voltage range	0.0–60.0 V
Volume	2.0 L
Size of the stainless-steel electrode	15×10×0.2 cm^3
Size of the aluminum electrode	15×10×0.1 cm^3

2.4. Electrocoagulation

Typically, an EC reactor cell comprises an electrolytic cell with one anode and one cathode. Upon connecting the reactor cell to an external power source, the anode electrode undergoes electrochemical corrosion due to the oxidation phenomenon, while the cathode is subjected to passivation. However, this electrode arrangement does not yield satisfactory results in wastewater treatment, as it necessitates the use of electrodes with a large surface area to achieve an acceptable metal dissolution rate. This issue can be addressed by utilizing cells with monopolar electrodes connected either in series or parallel. Figure 2 depicts a simple configuration of an EC cell with three anodes and three cathodes arranged in parallel.

The electrochemical reactions exhibited by metal M can be concisely summarized as follow [22]:

At the anode:

$$M_{(s)} \rightarrow M_{(aq)}^{n+} + ne^- \quad (1)$$

$$2H_2O_{(l)} \rightarrow O_{2(g)} + 4H^+_{(aq)} + 4e^- \quad (2)$$

At the cathode:

$$M_{(aq)}^{n+} + ne^- \rightarrow M_{(s)} \quad (3)$$

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^- \quad (4)$$

For stainless steel or iron electrodes, the cell occurred reactions are as follows:

Anode:

$$Fe_{(s)} \rightarrow Fe_{(aq)}^{2+} + 2e^- \quad (5)$$

Cathode:

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \quad (6)$$

Precipitation:

$$Fe^{2+}_{(aq)} + 2OH^-_{(aq)} \rightarrow Fe(OH)_{2(s)} \quad (7)$$

Overall:

$$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2\,(s)} + H_{2(g)} \quad (8)$$

In acidic conditions, the following reactions take place in the anode:

$$4Fe_{(s)} \rightarrow 4Fe^{2+}_{(aq)} + 8e^- \quad (9)$$

Precipitation:

$$4Fe^{2+} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)} \quad (10)$$

Cathode:

$$8H^+_{(aq)} + 8e^- \rightarrow 4H_{2(g)} \quad (11)$$

Overall:

$$4Fe_{(s)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)} \quad (12)$$

The EC process utilizing iron electrodes yields iron oxides with low concentration, which are nontoxic. Furthermore, the electrochemical dissolution of the electrode may result in the formation of hydroxide species in the aqueous medium [22,23].

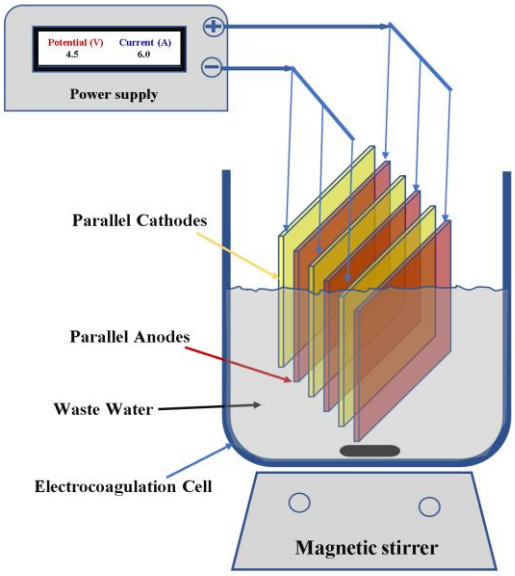

Figure 2. An EC reactor cell with monopolar electrodes connected in parallel arrangement

2.5. Method and procedure

An EC reactor cell with a volume of 2.0 L, equipped with four electrodes composed of aluminum and stainless steel, was used in this research. The anode and cathode poles were arranged vertically and in parallel to each other, with electrode distances of 5.0 cm. To clean the electrodes' surfaces, prior to each experiment, the electrodes were subjected to sandpapering and immersion in a 0.2 M dilute HCl solution for 15 min followed by rinsing with distilled water. Subsequently, all electrodes were heated at 100.0 ℃ for 15.0 min in an oven. During each experimental run, 1.0 L of the sample was introduced into the reactor cell, and the desired current density was achieved using the external DC power supply. The initial pH of the solution was adjusted using HCl (0.1 M) or NaOH (0.1 M), while the initial conductivity was fixed using NaCl (2000.0 mg/L). The experiment commenced once the simultaneous current density, aeration, and voltage had reached the desired levels (20.0 V), with the temperature maintained at a constant 25.0 ℃ throughout all stages of the experiments. The sample was subsequently filtered using Whatman filter paper, and the Cd concentration of the sample was measured by the AAS. To ensure the reliability of the results, all runs were conducted in triplicate. The removal efficiency percentage (%RE) of Cd was calculated as a function of EC at the end of the EC process (Eqn. 1).

$$\%RE = (C_0-C)/C_0 \times 100 \quad (1)$$

Where C_0 is the initial concentration and C is the final concentration of Cd, respectively. Figure 3 presents a diagram of the EC reactor cell employed in this study.

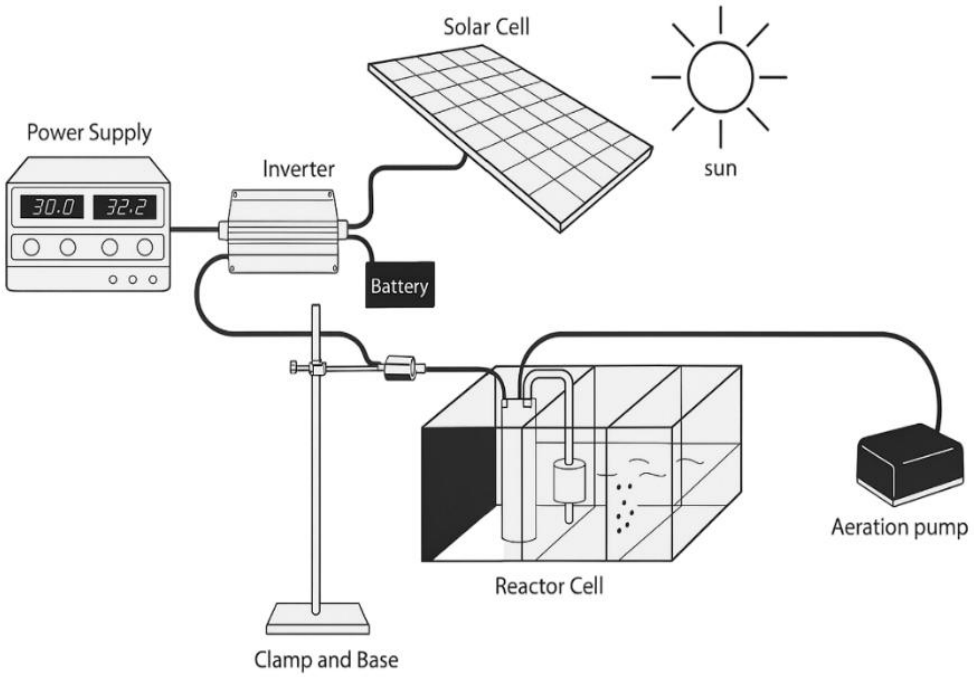

Figure 3. A diagram of the EC reactor cell employed in this study

3. RESULTS AND DISCUSSION

Electrochemical processes are known to be influenced by the current density, which has a significant impact on the production rate of coagulants, as well as the size and rate of bubbles. This parameter is also known to affect the growth of flocs on the EC, as reported in previous studies [24, 25].

In an initial study of the reactor cell's efficiency for Cd removal, a synthetic solution containing a consistent concentration of 100.0 mg/L of Cd was evaluated over varying removal durations at current densities of 3.0, 4.5, and 6.0 A/m^2 within the specially designed reactor cell. The findings are illustrated in Figure 4, which indicates that the reactor cell effectively functions as a viable apparatus for the removal of Cd from the solution, particularly under the conditions of a current density of 6.0 A/m^2 and a treatment duration of 40 min. The results presented in Figure 4 demonstrate that an increase in electrical current was associated with a higher %RE of Cd. It is postulated that this effect may be attributed to the removal of protective oxide layers from the surface of the electrodes through higher electro-dissolution [26]. The optimal treatment, resulting in about 100% removal of Cd within 40.0 min, was achieved at the highest electrical current of 6.0 A/m^2.

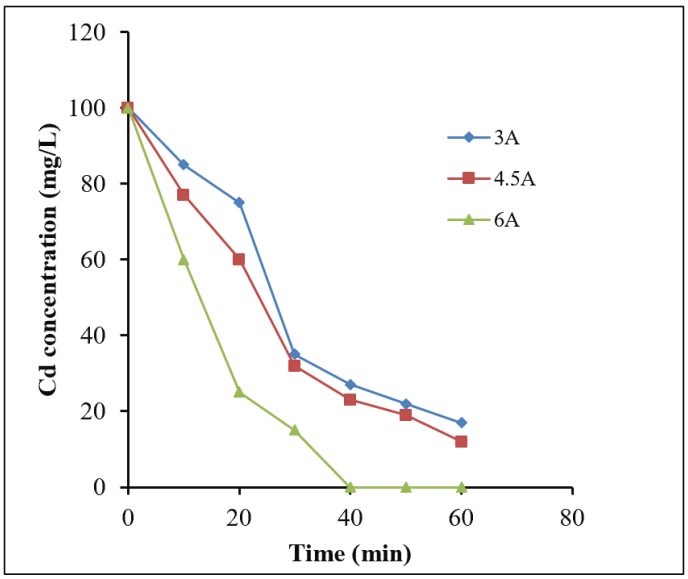

Figure 4. The reactor cell's efficacy for Cd removal from a synthetic solution 100.0 mg/L of Cd, over varying removal durations at current densities of 3.0, 4.5, and 6.0 A/m^2

Furthermore, to achieve optimal efficiency in the EC process, it is essential to carefully regulate several factors namely, the pH of the solution, current density, electrolyte concentration, contact time, and the spacing between electrodes. These parameters each play a significant role in process performance. In the sections that follow, each factor and its impact will be discussed in detail.

3.1. The solution pH

The pH value is a crucial factor in electrochemical or chemical separation processes, as it governs the formation of hydroxide metal species. Additionally, it plays a pivotal role in the removal mechanism of pollutants and ions [27]. In order to investigate the impact of pH, solutions of HCl and NaOH were utilized to achieve the desired pH levels. The dependence of the %RE on the initial pH was examined within the pH range of 3.0 to 12.5. Figure 5 illustrates the variation in %RE of Cd(II) ion with respect to pH after 40.0 min of electrolysis. These changes are observed at different current densities, specifically at 3.0, 4.5, and 6.0 A/m^2. The findings indicated that the initial pH had a significant effect on the %RE, particularly at high pH values. Previous studies have reported that Cd precipitation commences at pH= 8.2 [28], beyond which the RE begins to decline. At low pH values, H^+ ions compete with Cd^{2+} ions for adsorption sites on the adsorbent surface. Conversely, as the pH value increased, this competition diminished, allowing a greater number of Cd^{2+} ions to replace H^+ ions attached to the adsorbent surface. As depicted in Figure 5, the %RE of Cd increased up to 99% when the pH exceeded 8.5, while it was as low as 30% for pH= 3.0

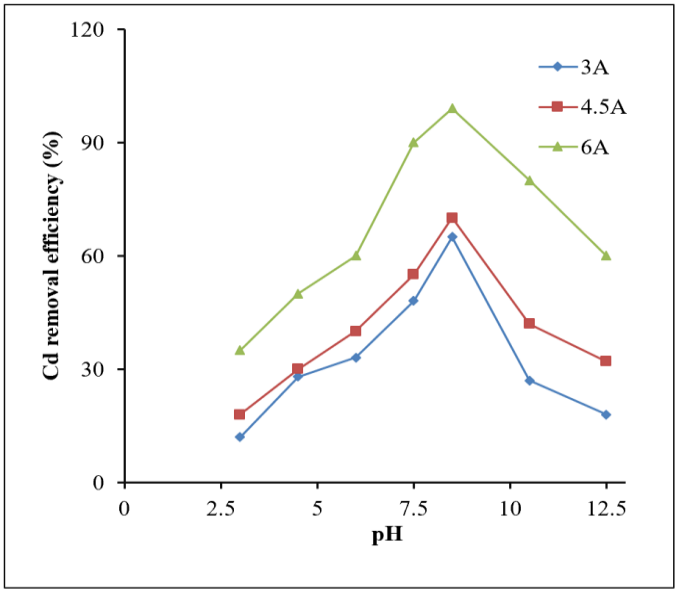

Figure 5. Effect of the initial pH on the %RE of Cd removal after 40.0 min of electrolysis, at current densities of 3.0, 4.5, 6.0 A/m^2 and electrodes spacing of 0.5 cm

As the results indicate, the optimal %RE of Cd was achieved at pH 8.5, where the Cd concentration decreased from 100 to zero (mg/L) within the 40.0 min reaction time, corresponding to a RE of 100%. Conversely, the lowest %RE was observed at pH 3.0, which can be attributed to the higher solubility of iron and aluminum species at this pH level compared to other pH levels. These soluble species are not suitable for water treatment with coagulating agents due to their ineffective adsorbing surface.

3.2. Current density

The impact of applied current on the %RE of Cd from an actual landfill leachate was investigated. The experiment was conducted under controlled conditions with an initial pH of 8.5, utilizing aluminum and stainless-steel electrodes spaced 0.5 cm apart, and varying applied currents of 3.0, 4.5 and 6.0 A/m^2 for a duration of 40.0 min. The results, as depicted in Figure 6, indicated that an increase in current density led to a corresponding increase in Cd %RE during the EC process. The findings presented in Figure 6 indicate that the %RE of Cd increases from 45% to 99% at a current density of 3.0 to 6.0 A/m^2, but no significant change is observed at higher current densities. Moreover, an elevation in current density results in the anodic dissolution of iron, which in turn produces a greater amount of hydroxide flocs for the purpose of pollutant removal. The current density of 6.0 A/m^2 was determined to be the optimal parameter for the subsequent experiments.

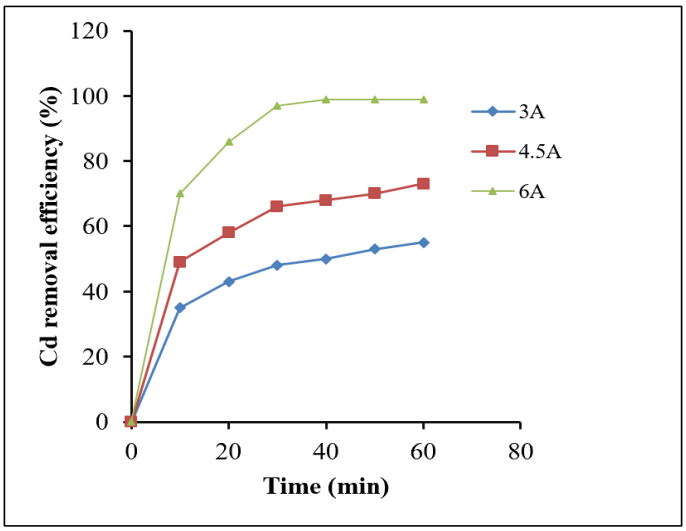

Figure 6. Effect of current density on %RE of Cd at pH 8.5, spacing electrodes of 0.5 cm, the contact time of 40.0 min, and 2000.0 mg/L of NaCl

3.3. Contact time

The duration of operation is a crucial determinant in the economic justification of the EC process. Faraday's law posits that the quantity of Fe or Al ions released to the system in EC systems utilizing Fe or Al electrodes may impact the residence time [29]. We examined the influence of electrolysis duration on the elimination of Cd from raw leachate up to 60.0 min, with an initial pH of 8.5 and a current density of 6.0 A/m^2. As depicted in Figure 6, the %RE of Cd raised to 99% as the duration was extended to 40.0 min, but it remains relatively constant beyond 40.0 min. This optimal contact time was determined to be the optimal parameter for the subsequent experiments.

3.4. Supporting electrolyte

The efficacy of the type of the suitable electrolyte, KCl, NaCl, and $NaNO_3$ as supporting electrolytes were examined. The objective was to compare the performance of each electrolyte under similar conditions in each batch, with a focus on achieving high %RE of Cd removal while minimizing energy consumption by reducing operating time. Figure 7 depicts the impact of NaCl, KCl, and $NaNO_3$ (2000.0 g/L) on the Cd removal efficiency at optimum pH of 8.5 and current density of 6.0 A/m^2 at stirring speed of 100.0 rpm, which were kept constant throughout the experiments. The results indicated that the use of different types of supporting electrolyte enhances the %RE of Cd. Specifically, KCl and NaCl yielded the highest %RE of Cd. An increase in the concentration of supporting electrolyte was found to be associated with a corresponding increase in the rate of Cd concentration reduction. For example, the %RE of Cd was approximately 99% at 0.6 kg/m^3 NaCl in the wastewater, while it was 85% at 0.41 kg/m^3 NaCl in the wastewater. The effect of $NaNO_3$ on the %RE of Cd was also examined, and the results suggested that $NaNO_3$ was not as effective as NaCl and KCl at the studied dosages. Overall, it can be concluded that KCl was as effective as NaCl in Cd removal, with a %RE of 99% and low energy consumption. However, NaCl is preferred due to its minimal toxicity, high conductivity and solubility, negligible impact on pH, and reasonable cost.

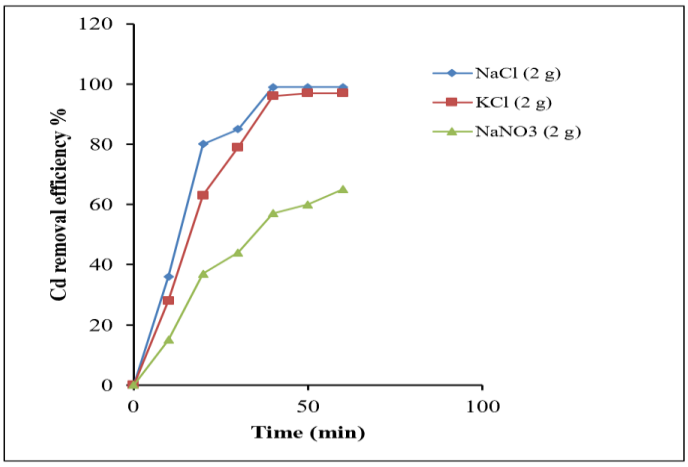

Figure 7. The impact type of supporting electrolyte, NaCl, KCl, and $NaNO_3$ (2000.0 mg/L) on the Cd removal efficiency at optimum conditions and at stirring speed of 100.0 rpm on the stainless steel-Al couple electrodes

To optimize the NaCl concentration as the best supporting electrolyte, the impact of NaCl concentration was examined as the electrolyte supporting agent on the %RE of Cd removal from the raw leachate. The results obtained indicated that electrolyte concentration was the most significant parameter affecting the %RE of Cd removal due to the role of active chlorine anions (ClO^- and/or HOCl) in the EC reaction [30]. NaCl addition to the EC system resulted in an increase in the conductivity level. Furthermore, it generated hypochlorite ions that acted

as an oxidizing agent in the degradation of the pollutant. The findings demonstrated that an increase in Cl^- concentration from 1000.0 to 2000.0 mg/L is accompanied by an increase in the %RE of Cd from 70.0 to 99.0%. Consequently, the optimal amount of NaCl was determined to be 2000.0 mg/L. This outcome can be explained by the fact that the presence of chlorides in the solution leads to anodic discharge, resulting in Cl_2 and ClO^- production. ClO^-, as a potent oxidant, can oxidize Cd ions present in the effluent. Therefore, the supporting electrolyte not only led to an increase in conductivity but also acted as a robust oxidizing agent. However, exceeding the optimal amount of electrolyte would result in the production of excess coagulant through the excessive dissolution of the anode.

3.5. Electrodes distance

The spacing of electrodes has a significant impact on the electrostatic field, and thus, the inter-electrode distance plays a crucial role in the EC process. Optimal Cd pollutant RE can be achieved by maintaining an appropriate distance between the cathode and anode [30, 31]. At the optimal inter-electrode distance, a portion of pollutants can be effectively eliminated from the aqueous phase through the flotation of H_2 bubbles. To investigate the effect of electrode distance on energy consumption and removal efficiencies, the EC process was conducted for 40.0 min at electrode distances ranging from 0.5 to 4.0 cm. As indicated in Table 3, an increase in the distance between stainless steel and aluminum electrodes from 0.5 to 4.0 cm resulted in a negative impact on the removal percentage of Cd. Conversely, when stainless steel and aluminum electrodes were combined, the total Cd removal percentage was only slightly increased. Table 3 provides additional information on energy consumption, EC efficiency, and removal rates. Overall, the best results in terms of the overall efficiency of the EC process were obtained at the smallest distance of 0.5 cm.

Table 3. The RE (%) of Cd removal at a pH of 8.5 and current density of 6.0 A/m^2

Operating time (min)	RE (%)				
	Electrode spacing (cm)				
	0.5	1	2	3	4
10	56.1	48.1	37.0	28.0	23.0
20	78.0	63.0	42.0	33.0	26.0
30	89.0	72.0	55.0	37.0	32.0
40	99.4	77.0	68.0	51.0	36.8

3.6. Scanning electron microscopy study

Figure 8 depicts the scanning electron microscopy (SEM) study of surface images of the stainless-steel plate in its original state prior to its utilization in the EC experiments. The electrode surface exhibited a uniform appearance, as shown in Figure 8A. Additionally, Figure

8B displays the SEM image of the same electrode after multiple uses in the EC tests. The electrode surface now displays a visibly rough texture, with indentations formed around the central regions of the active areas where electrode dissolution results in the formation of iron hydroxides. The emergence of a significant number of indentations may be attributed to the consumption of the anode material at the active sites, which is caused by the generation of oxygen at its surface.

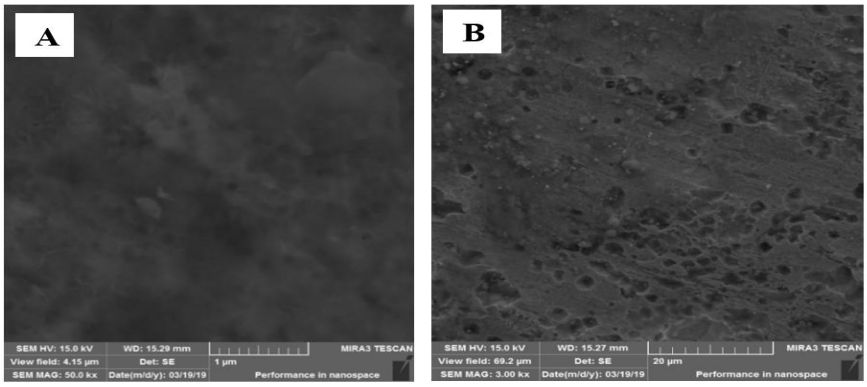

Figure 8. The SEM images of the stainless-steel plate in its original state (A) and prior to its utilization in the EC experiment (B), after multiple uses in the EC tests

To elucidate the efficiency of the constructed reactor cell in eliminating Cd from the actual Kahrizak sample, Figure 9 presents the authentic configuration of the designed reactor cell along with all necessary equipment. This figure depicts the examined solution of the Cd sample prior to treatment (A) and subsequent to treatment with the reactor cell (B). The images clearly demonstrate that the treatment was carried out successfully as expected under the optimized conditions.

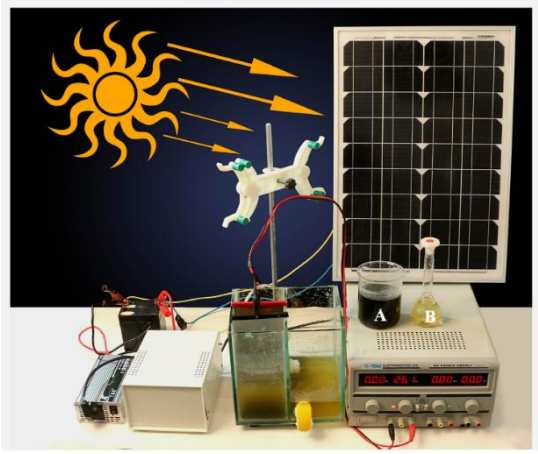

Figure 9. The configuration of the designed reactor cell along with all necessary equipment. The examined solution of the Cd sample before treatment (A) and after treatment with the reactor cell (B), under the optimized conditions

4. CONCLUSION

The present study aimed to investigate the efficacy of EC in removing Cd from both real landfill leachate and simulated wastewater, utilizing stainless steel and Al electrodes. The results obtained demonstrated that EC can be effectively employed for the treatment of both real landfill leachate and synthetic wastewater for Cd removal. The study also examined the impact of various parameters, including initial pH, current density, contact time, types of supporting electrolyte, and inter-electrode distance, on the performance of EC. The optimal operating conditions were determined to be an initial pH of 8.5, an inter-electrode spacing of 0.5 cm, a current density of 6.0 A/m^2, an electrolysis time of 40.0 min, and NaCl of 2000.0 mg/L as the supporting electrolyte. Under these conditions, the highest Cd removal efficiency of 99.0% was achieved for the leachate, and complete removal was achieved for the synthetic wastewater.

Declarations of interest

The authors declare no conflict of interest in this reported work.

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