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# TREATMENT OF LEACHATE FROM ERBIL LANDFILL SITE BY ELECTRO- AND CHEMI-CAL COAGULATION METHODS

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# **ABSTRACT:**

Landfill leachate is commonly heavily contaminated wastewater. and consists of a high number of organic compounds, inorganic salts, toxic gases and heavy metals that exert a serious threat to the environment and public health. Thus, it requires treatments before direct release into receiving waters. This paper presents the results of electrocoagulation (EC) and chemical coagulation (CC) treatment of leachate from the Erbil landfill site. The removal efficiency of chemical oxygen demand (COD), phosphate (PO43-), total suspended solids (TSS), total organic compound (TOC), and color of leachate was studied using iron and aluminum electrodes. The removal percentages were also compared to those produced by electrochemically generated Fe2+ and Al3+ dosages. The effect of different pH values on the removal efficiency of these parameters was evaluated at optimal conditions. The removal percentages for chemically added coagulants were lower than those for electrochemically generated Fe2+ and Al3+. In EC, the highest COD removal efficiency of 92% and 87% was achieved at the original concentration (C1) for iron and aluminum electrodes, respectively. The iron and aluminum electrodes also showed a maximum color removal of 90% and 95%, respectively, for the original undiluted leachate solution. Both Fe and Al electrocoagulation methods were not effective in removing TOC from the leachate of municipal solid waste. The highest removal efficiency of 78% was achieved at a 1:4 diluted solution (C2) using the Al-electrocoagulation method. The maximum removal percentage for PO43- was 94% at C1 using the Fe-electrocoagulation system. However, both systems were not very effective in removing TSS.

KEYWORDS: landfill leachate, ELS, Electrocoagulation, COD, TOC, TSS.

### 1. INTRODUCTION

As a result of the significant expansion of global population and resource consumption, in parallel with increasing the amount of solid waste, important environmental problems are experienced especially in the cities. Approximately one billion tons of waste are generated annually, and this number is expected to continue to increase over time, reaching roughly 2.2 billion tons by 2025, (Bhada-Tata & Hoornweg, 2012). Solid waste production could lead to a wide range of effects on human wellness and the environment (Zhang et al., 2013). When municipal solid waste (MSW) is not handled properly, it facilitates the rapid growth of vector insects, the release of hazardous substances, and the pollution of soil and water (Kjeldsen et al., 2002). Solid wastes have negative effects on our country. It gets thrown away by a process known as normal landfilling. Although landfill is preferable because of being less expensive, it results in the creation of leachate (He et al., 2019).

Landfill leachate is a wastewater with very high pollution parameters, and one of the most challenging wastewater to clean nowadays (Peng, 2017). In many parts of the world, there are storage areas without leachate accumulation systems (Luo et al., 2020). Leachate contains organic and inorganic pollutants. The untreated leachate has the potential to penetrate and pollute groundwater and soil. Consequently, effective leachate treatment is a critical concern. There are many different methods for the treatment of leachates such as advanced oxidation techniques, aerobic and anaerobic processes, membrane processes, and coagulation–flocculation (Nawaz et al., 2020).

Erbil landfill site (henceforth, ELS) was opened in 2001 and covers a total of 37 hectares, located near the Kani-Qrzhala Subdistrict on the Erbil-Mosul highway about 15 kilometers from Erbil city center. The coordinates are  $36^{\circ}10'23''N$  and  $43^{\circ}35'32''E$  (Aziz & Maulood, 2015). According to statistics obtained from the ELS administration employees, the facility gets more than 2000 tons of MSW every day. MSW is mixed without sufficient component separation.

Residual organic materials cannot be significantly removed by biological treatment methods (Deng & Englehardt, 2007). For this reason, different treatment processes are required to treat the leachate, the most widely used being physical and chemical processes (Lebron et al., 2021). The initial characteristics of leachate are critical in determining the treatment technique. Some effective and low-cost treatment methods need to be developed to fix this problem.

Electrochemical treatment method is a promising technology for wastewater treatment (Yu et al., 2020). They offer advantages over traditional methods, including reduced equipment requirements, and shorter treatment times. less need for chemical reagents. However, there are also some ordinary problems in electrochemical treatment that reducethe removal efficiency, such as corrodes of anode with time (Othman, 2018). Despite these disadvantages, electrochemical processes are still a promising technology for water and wastewater treatment (Bagastyo et al., 2020). As the technology continues to develop, the cost of operation is expected to decrease, making it a more economical option (Ghanbari et al., 2020).

In recent years, EC has gained acceptance as an effective leachate treatment technology. EC includes the use of direct current that flows between electrodes submerged in polluted wastewater or sewage effluents. The method encourages coagulation and flocculation processes, resulting in pollutant agglomeration into bigger particles that can be readily removed (Hamid et al., 2020).

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This study focuses on making a comparisons of aluminum and iron electrodes for the electrocoagulation treatment of leachate. There are many studies used aluminum and iron electrodes for the treatment of other industrial waste- waters (Bektas et al., 2004; Xu et al., 2017; Nasrullah et al., 2017; Nawarkar & salkar, 2019) However, the literature for the treatment of leachate by electrocoagulation process is not enough (Ilhan et al., 2008; Mandal et al., 2017; Guo et al., 2022).

The removal effectiveness of leachate produced from Erbil City's residential landfill (Kani-Qirzhala) using EC and CC was investigated in the study. At first, the fundamental distinctive characteristics of landfill leachate were investigated. The removal efficiency of such a mentioned process was evaluated concerning some parameters ,such as COD, PO4, TSS, total organic compound, and color. Both results obtained from CC and EC were compared for the above parameters.

# 2. MATERIALS AND METHODS

### 2.1. Landfill Leachate Properties

The sample was taken from the Kani Qirzhala, sub-district municipal waste Landfill. The removal efficiency of leachate collected from ELS by EC and CC was investigated. Table 1. shows specific characteristic values of raw leachate.

Parameter	Levels
pH	7.34-8.8
COD mg/L	4854-6255
TOC mg/L	1320
Conductivity mS/cm	20-30
color (455 nm) (Pt-Co)	3782
PO4 <sup>3-</sup> mg/L	358.5
TSS mg/L	356

Table 1. Some initial characteristics of leachate.

#### 2.2. Characterization and Scheme of EC Reactor

The treatment cell consists of a 1L glass reactor used for the CC and EC process. In the CC and EC process, the dimension of the reactor is (10 cm x 15 cm). Six electrodes (3 anode and 3 cathode) were used, each electrode is 3mm thick with the dimensions 8 cm and 12 cm. The distance between each electrode is 3mm. Electrodes have been connected to a Statron 3262.3 digital DC power supply (0-8 A and 0-300 V). During the experiment, a magnetic stirrer with a stirring speed of 300 rpm was used. The electrochemical process was applied at room temperature and atmospheric pressure. The pH was set to 3. A low pH encourages extra conditions for metal dissolution. In the present study, the removal of COD, TOC, TSS, PO4, and color from leachate through EC and CC process is investigated. For this purpose, Fe and Al electrodes were used in EC, and FeCl2·6H2O and polyaluminum chloride (PAC) (from Sinopharm Chemical. China) was used in CC. (Figure 1).



Figure 1. Experimental setup of EC system (1. magnetic stirrer, 2. magnet, 3. cathodes, 4. anodes, 5. Insulator, 6. beaker, 7. polarity changer, 8. power supply)

### 2.3. Analytical Techniques

When conducting the present study, the samples were collected and measured promptly without using any filter. Chemical oxygen demand data were collected using the TS2789 standard (water quality-Determination of COD). The samples were titrated with previously standardized ferric acid ammonium sulfate (FAS) to measure the quantity of potassium dichromate consumed (Bilgin et al., 2023). COD removal percentages of obtained results were calculated as follows:

COD removal % =  $[(C0 - C) / C0] \times 100$ 

Where C0 denote to initial concentration of COD, and Ct is the concentration at any time.

(1)

The color is determined spectrophotometrically. Color measurements were made by a photometer (MERCK-SQ118). Furthermore, the pH was measured by a pH meter (pH610). The adjustment of pH was done by using 0.1N NaOH and 0.1N H<sub>2</sub>SO<sub>4</sub>. A TOC analyzer (Sievers-2244 AP) was used in the study to measure the total organic compounds.

Phosphate analysis was performed by the method entitled 4500-PC "Vanadomolybdo phosphoric Acid Colorimetric Method" given in Standard Methods. A Photometer (Merck-SQ 118) was used in the experiment.

The TSS analysis in the study was done by the method given by (Estefan, 2013). In the experiment, filter paper (Millipore-AP40), TSS apparatus (Sartorius), oven (Venticell-MMM), and precision balance (Sartorius-BA210S) were used.

#### 3. RESULTS AND DISCUSSION

The removal mechanisms in EC involve the releasing of positive metal ions from an anode and reacting with hydroxide ions (OH<sup>-</sup>) that are formed by the cathode electrode to form metal hydroxide that can bind to and remove positively charged pollutants from wastewater

There is a linear relationship between the concentration of dissolved metal and current density at different time intervals. Thus, different concentrations of metals can be calculated from the current density (i) value (mA/cm<sup>2</sup>) (Table 2). The calculation was used in the conversion of current Density to metal dissolved quantities (DM) to make a comparison with CC.

Table 2. The linear equation for different values of current den-

sity (1) and electrolysis duration (min) in an EC reactor.								
Fe electrode	10 min	20 min	30 min					
а	4.907	2.076	1.933					
b	8.989	4.939	2.863					
R	0.955	0.965	0.950					
Al electrode								
а	1.426	2.189	4.262					
b	2.833	4.366	8.711					
R	0.998	0.960	0.973					

\*linear equation DM(g/L) = a+b+i; R:correlation coefficient

In the first part of the study, the treatability of solid waste leachate by the EC method, which is based on electrolytic oxidation of the anode, was investigated. COD, TOC, TSS, PO4, and color parameters were investigated, which was carried out by using Fe and Al anodes. In the CC method, which is the second part of the study, the salts of metals (FeCl<sub>2</sub>·6H<sub>2</sub>O and PAC) were added as coagulants. A CC study was also carried out in two steps by using different salts and by monitoring the same above parameter.

#### 3.1. Effect of Ferrous Coagulant Dosage

According to Table 3. which shows the relationship of Fe<sup>2+</sup> on COD, TOC, TSS, PO4<sup>3-</sup>, and color, it is clear that the removal capacity increases by increasing the dosage of Fe<sup>2+</sup> concentration. The optimal removal efficiency was observed for COD. So,

the maximum removal efficiency for the original leachate solution (C1) and (C2) was 92% and 82% respectively. such increase of the removal efficiency with additional dosage is observed in literature (Ilhan et al., 2008). The lowest removal efficiency was 55-60% observed for TOC in (C1) and (C2). The best color removal noticed for both concentrations (C1, C2) was 93%-89%. The results for leachate that was treated chemically by adding (FeCl<sub>2</sub>·6H<sub>2</sub>O), showed poor removal capacity when compared to electrically generated Fe<sup>2+</sup> (Table 4) in which the maximum removal efficiency recorded for COD, TOC, and color for both concentrations were 74%, 46%, and 67% at (C2) respectively. Table 3. The percentage of COD, TOC, and Color removal as the

function of electrocoagulation ( $Fe^{2+}$ ) for the undiluted original leachate (C1) and the 1:4 diluted solution (C2).

Fe <sup>+2</sup>		C1			C2	
Conc.	COD	TOC	Color	COD	TOC	Color
g/L	%	%	%	%	%	%
0.4	56	42	70	56	50	65
0.8	72	46	72	68	52	68
1.1	76	49	80	76	56	70
1.5	86	52	84	80	56	78
1.8	92	55	93	82	60	89

Table 4. The percentage of COD, TOC, and Color removal as the function of chemical coagulation ( $Fe^{2+}$ ) for the undiluted original leachate (C1) and the 1:4 diluted solution (C2).

			(	- / .			
Fe <sup>+2</sup>		C1				C2	
Conc.	COD	TOC	Color	-	COD	TOC	Color
g/L	%	%	%	_	%	%	%
0.4	19	15	46		50	11	44
0.8	33	25	61		57	38	52
1.1	52	38	64		61	43	60
1.5	55	44	66		73	46	69
1.8	60	46	67		74	46	75

Both electrically produced and chemically added  $Fe^{2+}$  show approximately similar removal efficiency for PO4<sup>3-</sup>, in which the maximum removal capacity was 94% and 93% respectively at C2 (Figure 2a,b).

As seen from (Figure 3a,b), it does not show the significant removal efficiency for suspended solids. The highest removal percentage recorded for EC was 73% at C1. This is due to the size and nature of the suspended solid that affect the performance of EC. In general, EC shows superiority in the removal efficiency when compared with chemically added (FeCl<sub>2</sub>·6H<sub>2</sub>O).



Figure 2. The percentage of  $PO4^{3-}$  removal as the function of  $(Fe^{+2})$  for a) EC, b) CC.



Figure 3. The percentage of TSS removal as the function of  $(Fe^{+2})$  for a) EC, b) CC.

#### 3.2. Effect of Aluminum Coagulant Dosage

In the same manner, as  $Fe^{2+}$ , the  $AL^{3+}$  was generated electrically by the electrode with the two different solutions of leachate (C1, C2), and its removal efficiency was examined for the mentioned parameter (Table 5). The obtained results depend on the concentrations of  $Al^{3+}$  formed by EC.

The COD removal efficiency increases from 57%-53% to its optimal treatment level of 87%-73% for C1 and C2 by increasing the dose of dissolved metal from (0.15 mg/L to 0.6 mg/L). However, after such a point, any increase in dissolved metal dosage shows a non-significant change in the removal efficiency. At this specific point, a further increase in dissolved metal may not cause significant removal due to factors, such as saturation of metals, formation of complexes, and interference from other ions (Tahreen et al., 2020).

The maximum removal efficiency observed for TOC was 67% and 78% for C1, C2 at 0.75 mg/L. This shows that the electrically produced  $Al^{3+}$  like Fe<sup>+2</sup> is not affectable in TOC removal. This may be due to the fact that the hydroxide complex that formed is not effective in the removal of all types of substances (AlJaberi, 2020). On the other hand, the results show the efficiency of a system in color removal. The highest color removal was 95% at 0.6 g/L dosage and 88% at 0.75 g/L was noticed for C1 and C2 at the end of the experiment. The use of a lower current density for optimal energy and color removal was also seen in a number of researches (AlJaberi, 2020; Rodrigues et al., 2020) Table 5. The percentage of COD, TOC, and Color removal as the function of electrocoagulation (Al<sup>3+</sup>) for the undiluted original leachate (C1) and the 1:4 diluted solution (C2).

eachate	(CI) and the III and		
	C1	C2	

Al <sup>+3</sup>	COD	TOC	Color	COD	TOC	Color
Conc.	%	%	%	%	%	%
g/L						
0.15	57	37	52	53	50	64
0.3	65	62	70	65	58	70
0.45	80	61	75	67	67	75
0.6	87	63	95	73	77	80
0.75	87	67	95	74	78	88

Table 6. The percentage of COD, TOC, and Color removal asthefunction of chemical coagulation (Al<sup>3+</sup>) for the undiluted original leachate (C1) and the 1:2 diluted solution (C2).

	0		( )	 		( - )
Al <sup>+3</sup>		C1			C2	
Conc.	COD	TOC	Color	COD	TOC	Color
g/L	%	%	%	%	%	%
0.15	33	18	36	58	37	39
0.3	47	26	56	64	42	60
0.45	49	35	60	67	45	71
0.6	55	45	67	70	47	76
0.75	56	46	68	73	49	74

The highest removal efficiency of PO<sub>4</sub> was 86% observed at C1 within a dissolved metal dosage of 0.75 mg/L (Figure 4a). The removal efficiency mainly depends on phosphate concentration in the solution (Abdulkhadher & j Jaeel, 2021). Similar to  $Fe^{2+}$  Al also shows poor performance in the removal of suspended solids. The highest removal percentage was 79% noticed at C2 for EC (Figure 5a). This is due to the small size of particles and the potential of electrode fouling with such particles (Elazzouzi et al., 2017).

Chemically adding dissolved metals (PAC) to both solutions C1 and C2 did not show any significant efficiency in the removal for all parameters (Table 6). The highest effective removal recorded at C2 was 73%,74% for COD, and color parameter with a dissolved dosage of 0.75 g/L. The phosphate removal potential is low when compared with chemically added FeCl<sub>2</sub>·6H<sub>2</sub>O (Figure 4b). The maximum removal percentage was 77% at C2. The highest removal percentage for TSS was 79 % noticed at C2 for EC (Figure 5b).

Figure 4. The percentage of  $PO4^{3-}$  removal as the function of  $(Al^{+3})$  for a) EC B) CC.





Figure 5. The percentage of TSS removal as the function of  $(A1^{3+})$ 

 $(Al^{3+})$  removal as the function of  $(Al^{3+})$  for a) EC B) CC The initial pH of the wastewater may have an impact on treatment efficiency since it impacts the stability of different hydroxide species generated in the process (Deghles & Kurt, 2016). To evaluate the impacts of the initial pH of leachate on the EC and CC process, laboratory experiments were carried out by varying the initial pH value. Taking the optimal metal ions dosage from the previous experimental stage, three different initial pH values were selected : 3, 6, and 9. Figure 6 reveals the varying levels of the COD removal efficiency for EC with beginning pH values. The COD removal efficiencies of 92%, 90.32%, and 68% have been obtained for pH 3, pH 5, and pH 9 respectively. Many previous studies related to EC using iron electrodes have shown that the maximum COD removals were observed at neutral pH6-7 (Bensadok et al. 2008; Ilhan et al., 2008). However, in this study original pH ( $\sim$ 3) was observed to be clearly more effective on the removal of COD.

According to Figure 7, the maximum removal efficiency for TOC removal was 78 % recorded at pH 3 and pH 5. While at pH 9 the removal efficiency decreases dramatically. A gel layer creates on the anode surface, particularly at elevated concentrations and pH, which is likely to interfere with aluminum hydrolysis (Izadi et al., 2018). The increase in pH during the experiment showed the reverse impact on color removal efficiency. The highest removal efficiency was 95 % and 90% at pH 3 and pH 5 respectively (Figure 8).

additional experiments were carried out for PO4<sup>3-</sup> and TSS under the previously optimized experimental conditions to investigate the effect of initial pH on the removal efficiency. Also, the increase in initial pH did not show any significant removal efficiency. The highest removal efficiency has been recorded at 94% (Fe-EC) and 79 % (Al-EC) at C2 for both  $PO_4^{3-}$  and TSS respectively (Figure 9) and (Figure 10). The obtained results are in accordance with the results of Bektas et al. (2004).



Figure 6. The effect of pH on COD removal efficiency (EC, C1,  $Fe^{+2}$  dosage =1.8 g/l, 30 min)



Figure 7. The effect of pH on TOC removal efficiency (EC, C2,



 $Al^{3+}$  dosage = 0.75 g/l, 30 min)

Figure 8. The effect of pH on color removal efficiency (EC, C1, Al<sup>3+</sup> dosage = 0.75 g/l, 30 min)



Figure 9. The effect of pH on PO<sub>4</sub> removal efficiency (EC, C2,  $Fe^{+2}$  dosage =1.8 g/l, 30 min)



Figure 10. The effect of pH on TSS removal efficiency (EC, C2,  $Fe^{+2}$  dosage =1.8 g/l, 30 min)

# CONCLUSIONS

The utilization of chemical coagulation (CC) and electrocoagulation (EC) processes demonstrates promise for the efficient treatment of leachate originating from municipal solid waste. A study found that EC using Fe and Al anode was effective in removing pollutants, such as COD, TSS, and color from leachate. The highest removal efficiency was achieved with undiluted original water (C1). The study also found that electrocoagulation using  $Fe^{2+}$  electrodes was more effective than electrocoagulation using  $Al^{3+}$  electrodes. The highest removal percentage of electrically produced  $Fe^{2+}$  was 92% at C1, while the highest removal percentage of electrically produced  $Al^{3+}$  was 87% at C1.

Both electrically formed ions were effective in removing color from leachate. Also, the highest removal efficiency was achieved with C1. The iron and aluminum electrode showed a maximum color removal of 90% and 95 % for the C1 solution respectively.

Both iron and aluminum electrodes were not effective in removing TOC from the leachate of municipal solid waste in EC. The highest removal efficiency noticed for TOC was 78% at C1 within Al-electrocoagulation methods.

The maximum removal percentage for  $PO_4^{3-}$  was 94% (1.8 g/l) at C1 within the Fe-electrocoagulation system. While both systems also were not so effective in TSS removal.

Taking the optimal metal ions dosage from the previous experimental stage for all parameters, the effect of initial pH showed that at acidic pH the system shows a significant removal efficiency.

As the landfill ages, the concentrations and characteristics of leachate also change and thus affect the coagulation performance. The presence of inorganic and organic loads in leachate may reduce the efficiency of EC due to the film formation around the anode. Moreover, the age of the landfill affects the pH and conductivity of the leachate and these two parameters highly affect the efficiency of coagulation as it affects the mobility and solubility of the coagulant. Therefore, it is so important to consider the age of the landfill when designing electrochemical treatment system . However, further study is needed to optimize both of these methods in terms of cost-effectiveness and performance under various conditions. It is critical to keep looking for creative methods to optimize pollutant removal efficiency while reducing environmental effects during leachate treatment operations.

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