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Investigating the Electrocoagulation Treatment of Landfill Leachate by Iron/Graphite Electrodes: Process Parameters and Efficacy Assessment

Tahereh Rookesh [1], Mohammad Reza Samaei [2,*], Saeed Yousefinejad [3], Hassan Hashemi [1], Zahra Derakhshan [2], Fariba Abbasi [1], Mahrokh Jalili [4], Stefanos Giannakis [5,*] and Muhammad Bilal [6]

[1] Department of Environmental Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz 7153675541, Iran; Trookesh68@gmail.com (T.R.); h_hashemi@sums.ac.ir (H.H.); Faabbasi@sums.ac.ir (F.A.)
[2] Research Center for Health Sciences, Department of Environmental Health, School of Health, Shiraz 7153675541, Iran; derakhshz@sums.ac.ir
[3] Department of Occupational Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz 7153675541, Iran; yousefisa@sums.ac.ir
[4] Department of Environmental Health Engineering, School of Public Health, Shahid Sadoughi University of Medical Sciences, Yazd 8915173160, Iran; mahrokh.jalili@ssu.ac.ir
[5] Departamento de Ingeniería Civil: Hidráulica, Energía y Medio Ambiente, E.T.S. de Ingenieros de Caminos, Canales y Puertos, Universidad Politécnica de Madrid, Unidad Docente Ingeniería Sanitaria, c/Profesor Aranguren, s/n, ES-28040 Madrid, Spain
[6] School of Life Science and Food Engineering, Huaiyin Institute of Technology, Huai'an 223003, China; bilaluaf@hotmail.com
* Correspondence: mrsamaei@sums.ac.ir (M.R.S.); stefanos.giannakis@upm.es (S.G.)

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Abstract: Electrocoagulation is a widely used method for treating leachate since it is cost effective and eco-friendly. In the present study, the electrocoagulation process was employed to remove chemical oxygen demand (COD), NH_4^+, total dissolved solids (TDS), total suspended solids (TSS), turbidity, and color from landfill leachate. At first, lime was used as a pretreatment, then the Fe/Gr and Ti/PbO_2/steel electrodes were used, and the optimum electrode was selected. Afterwards, the effects of some variables, including pH, current density, temperature, the inter-electrode distance, and the type of electrolyte were investigated. Results showed that COD, NH_4^+, TSS, TDS, electrical conductivity (EC), turbidity, color, and pH of effluent pretreatment chemical reached 22,371, 385, 884, 21,820 (mg/L), 13.8 (ms/cm^3), 1355 (NTU), 8500 (TCU) and 10, respectively (the removal efficiency was 0, 20.37, 32.4, 61.99, 59.18, and 56.6 percent). With the Fe/Gr electrode, the optimal condition was observed as follows: pH of 7.5, current density of 64 mA/cm^2, inter-electrode distance was equal to 1.5 cm, temperature at 20 °C, and retention time 2–4 h. Overall, the electrocoagulation with the Fe/Gr electrode was a suitable technology for landfill leachate treatment due to its effectiveness for the removal of both COD and NH_4^+, with advantageous performance indicators.

Keywords: electrocoagulation; leachate; landfill; treatment optimization; contaminant degradation

1. Introduction

Solid waste generation is increasing worldwide due to the rampant population and economic growth of its inhabitants [1]. Sanitary landfilling is the most common method and approaches that are used for managing municipal solid wastes (MSWs) [2]. Although landfilling is applied as a cheap and effective solution for countries with available space, this application does contain serious environmental risks due to the generation of landfill leachate (LL). Leakage of leachate to the environment can damage the water bodies [3]. The MSW leachate is a dark liquid containing soluble organic compounds (e.g., volatile fatty acids and resistant compounds), inorganic ions, such as ammonia (NH_4^+), chloride

(Cl^-), potassium (K^+), and sodium (Na^+), heavy metals, and xenobiotic organic compounds (such as aromatic hydrocarbons, phenol, chlorinated aliphatic, and pesticides) [4–6]. This composition is greatly variable and depends on MSW composition, landfill age, and the climatic conditions of the region [7]. Some factors such as precipitation, evapotranspiration, infiltration, and degree of the landfill compaction are related to the amounts of generated LL [8,9].

Iran is located in a warm and arid region of the Middle East and the Persian Gulf. The MSWs have been increasing gradually, mainly because of population growth over the last decades and the shift in lifestyle changes. A recent analysis showed that 60 to 80 percent of the Iranian MSW composition is organic materials (OMs), while the humidity is high (around 40 percent) [10,11]. These conditions lead to the intense leachate formation, as high as 70 to 100 L per metric ton of MSWs, approximately due to annual high precipitation (823 mm) and mean lower temperature (min = 5.8 °C and max = 40.4 °C) compared to other parts in Iran [12]. Therefore, there is a high volume of LL generation, so use of an effective and fast technology is essential for this area.

Although many attempts have been made to manage and alleviate the problem of leachate, complete elimination of the LL is not feasible. The first choice in handling LL is the use of the impermeable evaporation ponds. According to Moraes Costa et al. (2019), the most popular techniques in Brazil are conventional treatment methods, such as lagoon systems, biological filters, activated sludge, and wetlands, but this application is connected with notable issues in public health and the environment [8,13]. Therefore, this method needs to be replaced with a treatment rather than a management solution.

Considering its complex mixtures and high contamination, as well as the stricter discharge regulations, LL cannot be treated thoroughly using conventional and biological techniques [14–16]; so, the removal efficiency of COD obtained was 65% [17]. Several treatment methods have been practiced for leachate treatment, such as adsorption, precipitation, and coagulation–flocculation [18,19]. However, most of these methods do not eliminate the contaminants; therefore, it is preferred to apply destructive methods, such as the advanced oxidation processes (AOPa). The Fenton, photo-Fenton, membrane processes, coagulation and flocculation, flotation process, and membrane electrochemical reactor were previously used for leachate treatment [20–27].

Although some of the methods have shown notable efficiency in removal of COD and nitrogen, they are associated with complex treatments or excessive costs [28]. For instance, membrane filtration would require coupling with powdered activated carbon (PAC) and carbon nanoarchitectures or reverse osmosis combined electrochemical [29–31]. Moreover, the Fenton process would require high amounts of Fe and H_2O_2 [32,33]; both of the issues would impact the operational cost of the corresponding processes. Moreover, the initial costs and operational conditions are the main limitations of the dissolved air flotation (DAF) process for leachate treatment [34], while processes that mainly involve phase change prior to LL recirculation do not really alleviate the problem; some of the studied processes available in literature are provided in Table 1.

Table 1. The summary of literature review for leachate treatment by different AOPs.

Process	Experimental Condition					Efficiency of Removal			Power Consumption ($kWhr/m^3$)	References
	Initial COD (ppm)	Catalyst	pH	Time	Current Density	COD	Color	NH_3		
Photo-electro-Fenton	3000–5000	H_2O_2	1.5–5		0.07–0.35 A/dm^2	97	100	-	3.1	Adapted from [35]
Electro-oxidation		Stainless steel	7	35 min	4.3 mA/cm^2	42	97			[36]
Electrochemical		Boron-doped diamond			64 mA/cm^2	100			127.5	[28]
Reverse osmosis with electrochemical	8750	$Ti\text{-}PbO_2$		3.5 h	141 A/m^2	96			28.7	[30]
Fe_2O_3 NPs+ Electroflotation	2200			120 min		96	100	99		[37]

Among the processes presented in Table 1, electrochemical oxidation is a cost-effective and affordable means of removing persistent organic pollutants from LL [38,39]. The major advantages of electrocoagulation are its easy handling, full automation, low cost of operation, high efficacy (which could lead to recycling and reuse of LL), low sludge volume, and lack of chemical additives [40–42].

Currently, various anodes have been used for the electrochemical treatment of LL. The results of a study by Rezaei et al. (2015) showed that the electrocoagulation process using platinum and graphite electrodes effectively reduced the biological oxygen demand (BOD), chemical oxygen demand (COD), and chromium content [12]. Nisha Priya et al. (2005) investigated the electrochemical treatment of leachate using Ti/TiO_2-RuO_2-FeO_2 and copper. They showed that these electrodes could reduce the COD, ammonia, nickel, lead, cadmium, zinc, and chlorine by 78, 100, 85, 85, 90, 90, and 40%, respectively [43]. Moreover, a previous work studied the anodic oxidation of LL with Ti/BD and found that the removal of COD, TOC, NH_4^+–N, and total nitrogen (TN) were 95.17%, 91.89%, 81.18%, and 63.54%, respectively, while, with Ti/RuO_2 anode, they were 92.57%, 78.76%, 97.85%, and 72.12% at 83 mA cm^2 and 8 h [44,45]. Although Panizza et al. (2005) reported complete removal of COD and NH_4^+, some researchers have reported these as 92 and 84%, respectively [46–48]. Most of the aforementioned works were applied for the stabilization and sedimentation of leachate with low initial pollutants. Moreover, several studies operated at extensive conditions, such as high rate of current density, and high level of coagulants and oxidizer agents [49,50], which shows the removal efficiency increased in the higher current density.

Since limited studies were investigated on the evaluation of different electrodes for leachate treatment and the applicability of electrodes, and the coagulation process was determined unclear, this study aimed to propose an optimal condition for leachate treatment. So, it can reuse the treated leachate for irrigation of green space. Then, the anode type effects, pH, current density, the distance between electrodes, and treatment time on the efficiency of the EC process for removal of COD, NH_4^+, TDS, TSS, color, and turbidity were investigated. Moreover, this study focused on cost-effectiveness and main indices in environmental pollution for selectivity of the optimal electrode. According to the previous researches, this is the first time that these criteria have been used in electrochemical coagulation.

2. Materials and Methods

2.1. Characterization of Yasuj Landfill Leachate

Landfill leachate was collected from an MSW sanitary landfill site in Yasuj, Iran. Yasuj is located in a mountainous region in southern Iran, 550 km away from Tehran. This landfill is of medium age and its area is 16.5 hectares. The medium-age landfills are 5 to 10 years old [9,16,51]. The weight of solid wastes that enter the landfill site are approximately 220 metric tons per day (ton/day) or about 80,000 tons/year, and their volumes are estimated to be 27.000 cubic meters per year (m^3/year) approximately.

The raw leachate samples were collected at the bottom of the pond feedline by a drainage system, stored in clean polypropylene containers, kept at 4 °C, and were transported to the laboratory, where they were stored at -20 °C until further analysis [52]. The raw leachate was initially treated by a chemical process (lime addition) to reduce suspended solids. Table 2 shows the physicochemical characteristics of the raw leachate from Yasuj, including COD, NH_4^+, TDS, TSS, electrical conductivity (EC), turbidity, and color.

2.2. Experimental Design

The leachate treatment process included two steps to increase the efficiency of the treatment system, including the chemical (pre)treatment process and electrocoagulation process. In the first step, lime was added to the reactor, and then coagulation was performed. In each step, the concentration of the parameters was measured. In all cases, control samples were considered. Next, the electrocoagulation process was performed. Stage 1 is shown in

Figure 1 and also stage 2 provided in Appendix A, while a description of the parameters tested is given in Table 3.

Table 2. The composition of raw leachate of Yasuj landfill site.

Parameter	**PH**	**Color (TCU)**	**Turbidity (NTU)**	**EC (ms/cm^3)**	**TDS (mg/L)**	**TSS (mg/L)**	**NH_4^+ (mg/L)**	**COD (mg/L)**
Raw	6.25	19,600	3320	14.62	32,280	2326	486	22,000
Blank	7	0	2	220	500	0	0	0
Sediment leachate (blank for sedimentation process)	6	13,200	1740	13.76	21,480	825	486	22,153
Leachate + Lime	10	8500	1355	13.8	21,820	884	385	22,371
* standards of Iran	<8.5		<50		<1000	<100	<2.5	<200

* Adapted from [53].

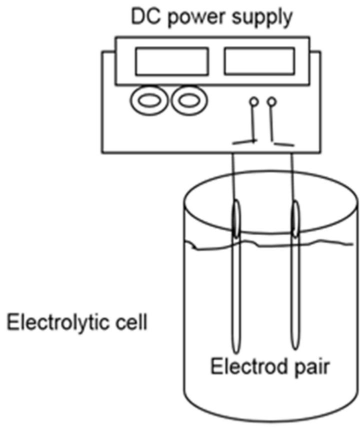

Figure 1. Experimental set-up (Stage 1).

Table 3. Variables related to the electrochemical process.

Parameters	**Values**
Electrolysis Time (min)	30, 120, 240
Electrolyte Type	NaCI, Na_2SO_4, No electrolyte
Ph	3, 7.5, 10.2
Current density (mA/cm^2)	16, 48, 64
Electrode distance (mm)	10, 15, 20
Temperature (°C)	Room temperature 20, 30, 40

2.3. Sample Preparation for Analysis

Samples were pretreated to remove suspended solids (SS) and greases. Firstly, 10 mL of the sample was diluted with distilled water to a volume of 1000 mL. Then, 800 mL of this sample was filtered through a 0.45 micrometers (µm) pore size filter [54] using a vacuum pump. The sample was used to measure various parameters, such as COD, NH_4^+, TDS, TSS, EC, turbidity, and color with the mentioned procedures in *Standard Methods for the Examination of Water and Wastewater* [55].

2.4. Physical and Chemical Analytical Details

The used chemicals were manufactured by Merck (Darmstadt, Germany) and Sigma (Ronkonkoma, NY, USA). After the operation of the electrocoagulation reactor, the physical and chemical parameters, including turbidity, electrical conductivity (EC), pH, color, TSS, NH_4^+, and COD, were analyzed according to the corresponding standard method: turbidity was measured by a turbidimeter (2100Q Laboratory Turbidimeter, HACH Company, Loveland, CO, USA). pH was measured by a portable pH meter (SANXIN comb.pH, San-Xin Instrumentation Inc., Shanghai, China). COD was determined using the closed reflux titrimetric method certified by the APHA. NH_4^+ was analyzed by the Nessler method [55]. The color was measured using a standard platinum cobalt method (8025 HACH) by spectrophotometer (HACH DR5000, HACH Company, Loveland, CO, USA). TSS and TDS were measured using the gravimetric method [15].

2.5. Electrocoagulation Monitoring

The EC was measured with a standard WTW325 device based on the standard gravimetric method. The current supply voltage of 3A/5V (MEGATEK, MP3003 D, Villeneuve le Roi, France) was used as the current density supply in the electrochemical cell so that the voltage and then the current density were adjusted.

2.6. Statistical Analysis

Results were compared by the nonparametric statistic Kruskal–Wallis test, followed by Dunn's comparison test ($p < 0.05$). Correlations between data were analyzed through Spearman's rank correlation coefficient and linear regression using Infostat. Each experimental condition was performed in triplicate and the average standard deviation was approximately 0.05.

3. Results and Discussion

3.1. Characteristics of the Yasuj Leachate

According to Table 2, the leachate had slightly acidic properties, dark brown to black color, high COD, turbidity, and total solids (TSS and TDS). These results can be attributed to leachate's nature, age, and regional climatic parameters [54]. Because of the investigated leachate is classified as medium age and acidic (pH = 6.25), the SS is easily dissolved in the LL. Hence, SS converts to dissolved matter, and TDS (32,280 mg/L) was higher than TSS (2326 mg/L). As a comparison, in Hamadan, a region which has different characteristics to Yasuj, the amount of BOD5, COD, and TS in the raw leachate was 20,000, 85,000, and 200,000 mg/L, respectively [56], while, compared to other medium-age LL (e.g., Qazvin) [57], our values are lower.

As expected, there was a high concentration of NH_4^+, COD, EC, color, and turbidity in the investigated raw leachate. Therefore, the use of pretreatment was necessary. In this study, lime was used as a typical cost-effective chemical pretreatment. TSS in the leachate was consistent with that of typical MSW leachate [58]. The leachate filtration could remarkably reduce the TSS in such leachates. Moreover, the value of COD = 22,000 mg/L and BOD/COD = 0.1 indicates that the biodegradability of the leachate was negligible.

3.2. Leachate Treatment

Today, there are various technologies for the treatment of leachates. However, the local context can significantly affect the choice of the process; Yasuj is a small, cold city and the application of biological treatment methods is neither feasible nor cost-effective. Currently, the most common landfill leachate managing method is evaporation. In many cases, there is not any management and the untreated LL is released to the environment. Hence, studying and proposing an effective method is of utmost importance.

3.2.1. Chemical Flocculation

For chemical pretreatment, the lime was added to the reactor and stirred. Then, the efficiency of the process on the removal of COD, NH_4^+, TDS, TSS, turbidity, and the color was calculated at 0, 20.37, 32.4, 61.99, 59.18, and 50.63 percent, respectively. The highest efficiency of this process was associated with TSS and turbidity. During the complexion process of particles and the generation of suspended flocs from dissolved matters, TSS and TDS were decreased. Turbidity and color removal can be related to mixing during pretreatment. However, lime addition as a pretreatment was not efficient for COD and

NH_4^+ removal. The reactor solution was not filtered before analysis, since the particulate COD was measured; the level of COD was increased slightly after the liming process.

The concentration of COD (22,371 mg/L) and ammonia (385 mg/L) and other parameters in the effluent from the chemical process exceeded the disposal limits. Therefore, the chemical process was proved to be inefficient. On the other hand, the effluent of the chemical process had a high conductivity value due to the high concentration of ions. Therefore, the electrochemical oxidation could be feasible, even without the addition of electrolytes.

3.2.2. Electrocoagulation Process

Since the use of the electrode in the electrochemical process is debatable, at first, the efficiency of two electrodes of different components and structure was investigated for the removal of COD, NH_4^+, TDS, TSS, turbidity, and color from LL. An Fe/Gr and a Ti/PbO_2 electrode were tested and their removal performance is summarized in Figure 1.

As shown in Figure 2, the highest efficiency for COD and NH_4^+ removal was found to be achieved by the Fe/Gr electrode, whereas the efficiency performance of Ti/PbO_2 for removal of TSS, TDS, turbidity, and color was higher than that of the Fe/Gr.

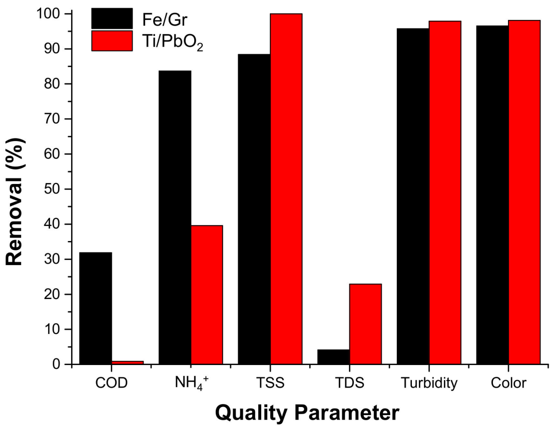

Figure 2. The efficiency of Fe/Gr and Ti/PbO_2 electrodes for leachate treatment (current density = 64 mA/cm^2, inter-electrode distance = 1.5 cm, pH = 7.5, electrolysis time = 120 min, temperature = 20 °C, and without electrolyte addition).

As also shown in Figure 2, the decrease in the parameters in question was divided among the two electrodes. For instance, the Fe/Gr electrode had a higher performance in COD and ammonium removal. This may be due to generated Fe^{3+} during the electrocoagulation process leading to a high level of sediments with good structure in comparison to the other flocks. In some studies, the highest COD removal rate obtained by iron electrodes was about 65 [54] and 35% [59], whereas we measured 32.4%.

The removal capacity of COD by Fe/Gr electrodes was higher than Ti/PbO_2, which is similar to the other studies, such as Li et al. [60]. However, the removal rate of TSS by the Fe/Gr electrode was 88%, compared to the 100% by the Ti/PbO_2 electrode, because of the generated hydroxyl radicals adsorbed firmly on the surface of the electrodes. Dissolved OMs, such as organometallic compounds, are precipitated by covalent compounds and decreased TSS [39,61]. That may be related to the structure of the electrode, in which the PbO2 was coated on the surface of Ti.

The removal rate of TSS, TDS, turbidity, and color by Ti/PbO_2 electrodes were moderately to significantly higher than the corresponding rates of Fe/Gr. In general, the most important factors in the selection of optimal electrode are cost, availability, and operation [38,60,62]. Hence, a cost estimation was performed to select the appropriate electrode.

For the consideration of the economic aspects of the treatment, the energy cost (EC) for the tests was calculated by the equation of current density (Equation (1)):

$$\text{Energy cost (kWh g}^{-1}\text{ COD)} = (V \times I \times t)/(\Delta(\text{COD})_{\text{exp}} \times V_S) \quad (1)$$

in which, V is the cell voltage, Vs is the solution volume (L), Δ(COD)exp is the experimental COD decay (mg L^{-1}), and I is the applied current intensity. From the application of the above formula, the energy cost of the Fe/Gr electrode is 35.9 and 2.1 times lower than that of Ti/PbO_2 for the removal of COD and NH_4^+, respectively.

The energy consumption of bio-treatment landfill leachate using a novel reactive electrochemical membrane (REM) technology was 3.6 kWh/m^3 [63].

Considering the removal rates of COD and NH_4^+, as well as the unitary cost of the treatment, the Fe/Gr electrodes were selected as the optimal electrode for further experimentation. Both COD and NH_4^+ have a significant effect on the ambient environment, such as eutrophication and the contamination of water supply [15]. Although the removal efficiency of turbidity and color by Fe/Gr was less than Ti/PbO_2, it was more than 95 percent. That is acceptable based on Iran standards for effluent of leachate. Moreover, the corrosion of the Ti/PbO_2 electrode was higher than Fe/Gr. Considering the above, the Fe/Gr electrode was chosen for the next experiments.

3.2.3. Effect of Operational Parameters on the Treatment Efficacy

After the determination of the optimal electrode, experimental parameters' effect on leachate treatment was investigated, namely the pH, the current density, electrolyte used, temperature of the matrix, and inter-electrode distance.

3.2.3.1. pH Variation

The removal percent of COD, NH_4^+, TDS, TSS, turbidity, and color are shown in Figure 2. The experiments were conducted by Fe/Gr electrodes and without the use of electrolytes.

In the electrocoagulation process, pH performs a key role in the treatment process [60,64]. According to Figure 3, pH had the highest effect on color, TSS, NH_4^+, and turbidity removal. Moreover, it seems that pH = 7.5 was the most effective condition and the stability of the Fe/Gr electrode in natural pH was higher than the others, corroborated by previous studies [60].

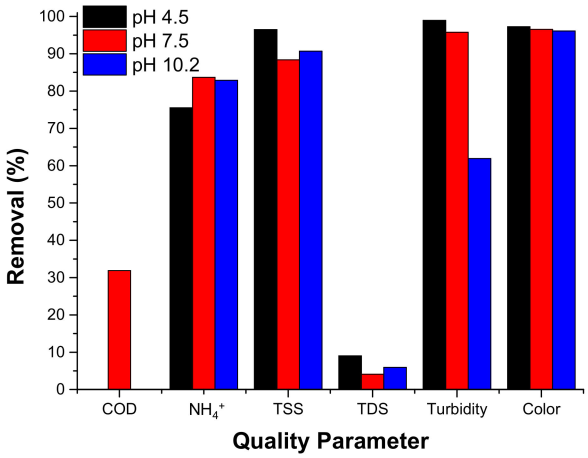

Figure 3. The effect of pH on the removal rate of COD, NH_4^+, TDS, TSS, turbidity, and color (current density = 64 mA/cm^2; inter-electrode distance= 1.5 cm; electrolysis time = 120 min; temperature = 20 °C).

Although the removal of TDS, TSS, turbidity, and color in acidic conditions was higher than neutral pH, this difference was not significant. The pH influences the hydrogen evolution at the cathode. Increasing the pH transfers CO_2 and exchanges OH^- to the other anions. In this situation, the powerful radicals are converted to weaker compounds, such as oxygen [10]. Moreover, the anodic discharge was decreased in acidic conditions due to a competition reaction [65]. In other studies, maximum removal of COD and ammonia had been reported at pH = 7.5 and 6.7 [50,65,66]. Likewise, solution conductivity is decreased in alkaline condition because of proton consumption [39], which explains the decrease in efficacy. Besides, water is decomposed into hydrogen and hydroxyl ions, and migration of ions is limited [67]. Therefore, pH = 7.5 was elected as optimal and other experiments were conducted with this pH.

3.2.3.2. Current Density Effect

The current density is an important operating factor that determines the energy consumption [60]. The investigated current densities were 12, 24, and 64 mA/cm^2, and the effect of these parameters on the removal rate is demonstrated in Figure 3.

As shown in Figure 4, the effect of current density = 64 mA/cm^2 was higher than the other densities tested, with a removal rate of COD, NH_4^+, TSS, turbidity, TDS, and color at 30.63, 83.67, 88.37, 95.59, 4.5, and 96.51%, respectively. In other studies, the enhanced electrochemical oxidation was observed in the higher current density [38,50,59,66,68] because, at a higher current density, the production of $HO^{\bullet}$ is increased remarkably, which leads to the reaction with the OMs [53,66]. Based on Faraday's law, the coagulant agent's generation (Fe^{2+}) is increased during an increase in the applied current density. The bubble-generation rate is increased and the bubble size is decreased; hence, the faster removal of pollutants by H_2 bubbles is observed [59,60]. The increase in electricity current flocculates colloids and removes it from the solution as a deposition. In high currents (at a small electrocoagulation unit), COD was not eliminated thoroughly. Since hydrophobic molecules are adsorbed on the surface of flocs and high molecular weight compounds, they can exert a weak charge density, which may be more easily neutralized by EC. Thus, OMs with low molecular weight may not be readily removed via an individual electrocoagulation process [53,60].

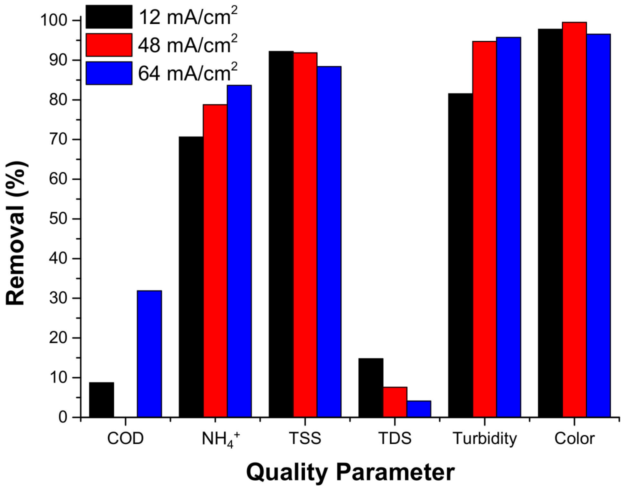

Figure 4. Removal rate of COD, NH_4^+, TDS, TSS, turbidity, and color during variation of current density (pH = 7.5; inter-electrode distance= 1.5 cm; electrolysis time = 120 min; temperature = 20 °C).

Finally, the current density is one of the most significant factors for controlling the reaction rate in the electrochemical process, and higher current densities are more economical [28]. Accordingly, the current density of 64 mA/cm^2 was selected as the optimal condition.

3.2.3.3. Effect of the Inter-Electrode Distance

In Figure 5, the effect of inter-distance between electrodes is presented. According to the results, for most parameters, the best removal rate was related to the inter-distance between electrodes of 1.5 cm (e.g., TDS, color). In other studies, the optimal condition for COD removal had been obtained in a 2 cm gap between aluminum electrodes [61]. Although anion evacuation at the anode electrode is increased in a lower distance, an electrical spark is a limiting factor. Therefore, it seems that the optimum condition for anion evacuation is 1.5 cm. Concerning the results, the inter-electrode distance of 1.5 cm was selected as an optimal distance, since COD and NH_4^+ removal, the two key parameters, were higher than the others at this distance, as well as the turbidity.

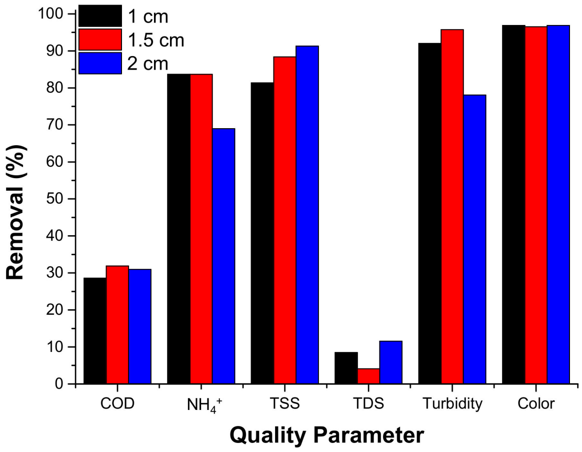

Figure 5. The removal rate of COD, NH_4^+, TDS, TSS, turbidity, and color in several distances between electrodes (pH = 7.5; current density = 64 mA/cm^2; electrolysis time = 120 min; temperature = 20 °C).

3.2.3.4. Electrolyte Type

The effect of electrolyte type on the removal rate of COD, NH_4^+, TDS, TSS, turbidity, and the color is demonstrated in Figure 6. While the efficiency of the electrochemical process was similar among the different electrolytes, the result for TDS was different. Although the higher amounts of ions may enhance the electrochemical process, the addition of NaCl and Na_2SO_4 had an inhibitory effect. Because SO_4^{2-} can contribute to the reactivity of oxygen, it may confine the generation of chlorites and hypochlorite. Moreover, the generation of active chlorine, including chlorine gas (Cl_2), hypochlorous acid ($HOCl^-$), hypochlorite ion (ClO^-), and peroxydisulfate ($S_2O_8^{2-}$), by both electrolytes causes indirect oxidation with beneficial effects [69], as detailed by the following equations Equations (2)–(7):

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{2}$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \tag{3}$$

$$HOCl \rightarrow H^+ + ClO^- \tag{4}$$

$$HSO_4^{2-} + 2OH^- \rightarrow SO_4^{2-} + 2e^- + H_2O \tag{5}$$

$$2NH_4^+ + HOCl \rightarrow N_2^+ + 2H_2O + 6H^+ + 2Cl^- \tag{6}$$

$$NH_4^+ + 4HOCl \rightarrow NO_3^- + H_2O + 6H^+ + 4Cl^- \tag{7}$$

Previous studies have shown that the effects of adding 100 mM NaCl as electrolytes on COD removal were negligible, but the ammonia removal was higher than normal conditions by the oxidizer products (according to Equations (1) and (2)). Moreover, the removal efficiency of TDS with Equation (6) was higher than that of 7, happening as a consequence of higher Cl content. Because the NH_3 and NH_4^+ convert to nitrogen gas (N_2), hydroxyl radical ($HO^{\cdot}$), and other reactive matter that participates in the oxidation of OMs, direct oxidation of OMs occurs on the surface of the anode. NaCl increases the removal of TSS due to the production of floated and suspended flocs. However, as a side effect, the turbidity and color removal were reduced to the presence of reactive oxidants on the electrode surface, compared to the tests without electrolyte addition. Nevertheless, since the electrochemical process diminished all variables, even without the addition of the electrolytes, this condition was selected as optimal.

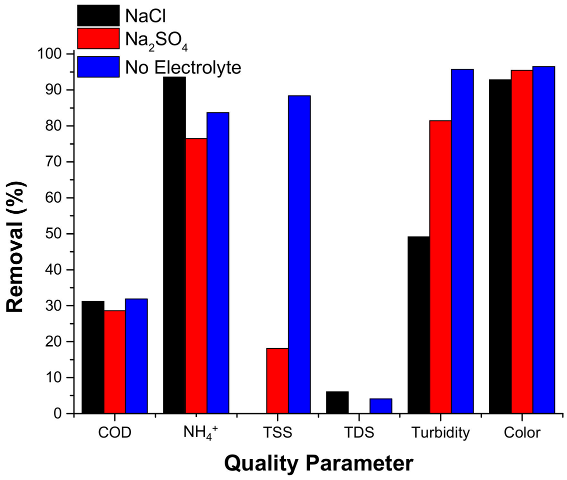

Figure 6. Removal rate of COD, NH_4^+, TDS, TSS, turbidity, and color without electrolyte, with sodium chloride (NaCl), or sodium sulfate (Na_2SO_4) (pH = 7.5; current density = 64 mA/cm^2; electrolysis time = 120 min; temperature = 20 °C).

3.2.3.5. Temperature Effect

In the current study, the effect of temperatures (20, 30, and 40 °C) was probed on the leachate treatment. The highest removal rates of COD, NH_4^+, TSS, turbidity, and color are attained at 40 °C (Figure 7). In other studies, COD had been notably removed at 25–50 °C [66]. The inhibitory effect when temperature increased in our work can be related to the nature of the coagulation reaction and the exothermic reaction of the electrochemical process. During this process, some of the compounds are converted to persistent intermediates [70]. Although, generally, the adsorption behavior of anion and discharge capacities of the electrode are increased at higher temperatures, our use of electrocoagulation instead of simple oxidation leads to 20 °C as the operational temperature of our choice.

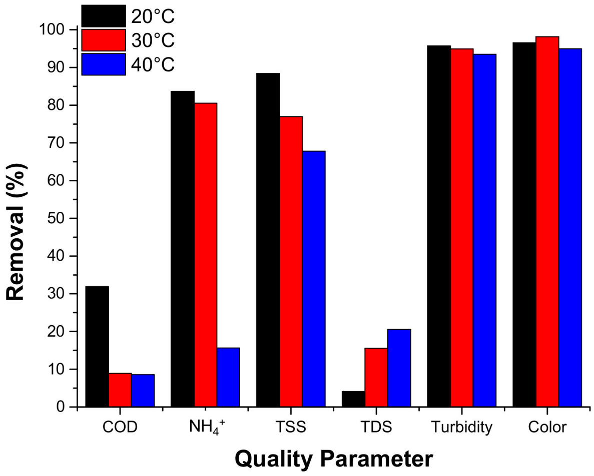

Figure 7. The effect of temperature on the removal rate of COD, NH_4^+, TDS, TSS, turbidity, and color (pH = 7.5; current density = 64 mA/cm^2; inter-electrode distance= 1.5 cm; electrolysis time = 120 min).

3.2.3.6. The Effect of Electrolysis Time

In the present study, the removal of LL contamination parameters during electrolysis time was investigated. The results are shown in Figure 8. The highest removal rate of all variables was after 2 h of electrolysis. In other studies, the optimal reaction time for COD and NH_4^+ removal was obtained at 20–90 min [38,53,58,59], because electrolysis time and the soluble metal level ions were increased. These results are the cause of the electrode poisoning, in which the catalytic activity of the anode is decreased and the polymer layer/film formed on the anode surface. The oxidation may be suppressed in the appropriate part where water is made to react with active oxygen or inorganic oxidizer, such as chloride ions [45]. Moreover, in our case, when the electrolysis time was raised, the hydroxide flocs were produced from iron ions and the rate of bubble generation was increased. Hence, the pollutants in leachate were removed by the co-effect of coagulation and flotation [60]. We note that this effect may differ according to the hydraulic parameters (mixing, size, etc.); hence, the difference noted in our tests. Based on Faraday law, the level of generated OH^- in electrochemical cells relates to current density. This rate is estimated from the following Equation (8):

$$\Delta m = (I \times t \times M)/(Z \times F) \quad (8)$$

where:

I: current density, t: reaction time, M: molecular weight, Z: the number of transmitted electrons during the reaction, F: Faraday coefficient = 96,486 c/mol.

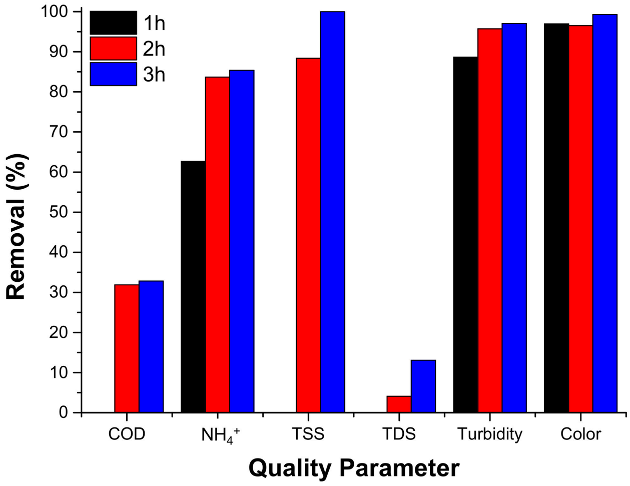

Figure 8. The effect of electrolysis time on removal rate of COD, NH_4^+, TDS, TSS, turbidity, and color (pH = 7.5; current density = 64 mA/cm^2; inter-electrode distance = 1.5 cm; temperature = 20 °C).

In low current density, a higher time is needed for similar efficiency. This fact results from the effects of loading on the treatment efficiency [71].

$$Q = I \times t \quad (9)$$

According to Equation (9), the level of generated ions requires higher loading. However, this usually is maintained at a low level because the cost is increased by increasing the loading.

After 2 h, COD removal increased insignificantly ($p > 0.05$) due to the generation of intermediates, corrosion of the electrode's surface, and conversion of SS to DS. The previous results were attained for NH_4^+, because NH_4^+ is stripped by the formed gases which approach the cathode [59,60,65].

4. Conclusions

Landfill leachate is categorized as a challenging effluent that should be mainly treated by advanced techniques. Therefore, in this study, conventional coagulation processes with the electrocoagulation process were combined for the removal of COD, NH_4^+, TSS, TDS, turbidity, and color from the municipal solid waste landfill leachate. This study applied two different electrode combinations, Ti/PbO_2 and Fe/Gr, in the ECO process with a pretreatment step, lime coagulation process. In this study, the chemical pretreatment by lime could remove the COD, NH_4^+, TDS, TSS, turbidity, and color about 0, 20.37, 32.4, 61.99, 59.18 and 56.6 percent, respectively. Since this process is inefficient, the effluent was entered into the electrochemical reactor. At first, the Fe/Gr was selected as an optimal electrode due to a higher capacity for removal of COD and NH_4^+. Both of these are critical parameters in environmental issues and are introduced for the determination of the electrochemical process efficiency. With the Fe/Gr electrode, the optimal condition for leachate treatment was observed in the current density of 64 mA/cm^2, the inter-electrode distance of 1.5 cm, pH of 7.5, electrolysis duration time of 2–4 h at lab temperature, and without adding of the electrolytes. In this condition, the removal of COD and NH_4^+ was higher than that of the others and the removal of color and turbidity was higher than 95%. Moreover, the cost and availability of the compound has a remarkable effect on the selection of optimal conditions. Improvement and modification of experimental conditions and further studies on other electrodes are proposed. Finally, our treatment data and the characteristics of the effluent suggest that the treated leachate should be further processed in an aerobic biological treatment facility before they are released into the environment because more biodegradable substances are expected to be formed.

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Appendix A

Two steps, including the chemical (pre)treatment process and electrocoagulation process. In the first step, lime was added to the reactor, and then coagulation was performed. Step 2 is shown in the following section.

Electrical Coagulation Stage (Stage 2)

The supernatant was transferred to another container in the previous step. The pH was adjusted to 7.5 using concentrated HCl solution. At this stage, a cylindrical reactor with a capacity of one liter was used. Then, 200 mL of the previous stage supernatant was increased to 800 mL using city water. A pair of anodes and cathodes were placed vertically with each other adjusted inside the reactor. The effective surface of each electrode was 30 cm and its thickness was 1 mm. Next, the following steps were performed. The electrical coagulation process was performed in two stages. In the first stage, complete anodes (Ti/PbO2.Al.Fe) and cathodes (steel, graphite, Cu) were used to complete the best anode and cathode in terms of factor removal (COD, NH3, TDS, TSS, turbidity, and color) to be selected. At this stage, other parameters of current intensity (64 mA/cm^2), distance between electrodes (1.5 cm), and pH (7.5) were considered constant and each experiment was performed for 120 min at laboratory temperature. In each of these laboratory conditions, no electrolyte was added to the solution because the concentration of leachate compounds remained unchanged. One of the problems during this stage could be the long duration of experiments due to the adhesion of clots on the surface of the electrode to prevent the production of coagulation ions. To eliminate this problem, the electrodes were superficially cleaned at 30-min intervals, and the foam was physically removed from the surface to avoid problems. In the second step, the time variables were examined using the one-factor method using optimal electrodes. Then the best values of electrolysis time, electrolyte type, current intensity, electrode distance, and temperature were determined.

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