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Evaluation of the efficiency of electrocoagulation process in removing cyanide, nitrate, turbidity, and chemical oxygen demand from landfill leachate

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Abstract

Background: Leachate contains toxic and non-biodegradable substances that are not easily treated by conventional treatment methods. This study investigated the effect of pH, current density, and reaction time parameters on the removal of cyanide (CN⁻), nitrate (NO3⁻), turbidity, and chemical oxygen demand (COD) from leachate by electrocoagulation process.

Methods: This study was an experimental one with direct current using four parallel bipolar aluminum electrodes with 90% purity. The length, width, and thickness of the electrodes were 5 cm, 10 cm, and 2 mm, respectively. There were 6 holes with a diameter of 0.7 cm on each of the electrodes. The samples were prepared from the old leachate of solid waste landfill in Ghaemshahr, Iran.

Results: In this study, at a current density of 33 mA/cm² and a time of 60 minutes, the optimum removal efficiency of cyanide (100 %) was obtained at pH 5.5 and pH 10. Moreover, the maximum removal of nitrate (99.65%) and turbidity (86.41%) were at pH 5.5 and pH 8.3, respectively and the highest removal efficiency of COD (83.14 %) was obtained at pH 10.

Conclusion: The results showed that the removal of cyanide, nitrate, turbidity, and COD increases with increasing current density and reaction time. Due to the proper removal of nitrate and cyanide from leachate by electrocoagulation, nitrate and cyanide amounts were less than the allowable contamination level. Based on the results, electrocoagulation is considered an efficient and effective method for removing nitrate and cyanide from old leachate of municipal solid wastes.

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Introduction

Sanitary landfilling is the most common method of solid waste disposal due to its simplicity and low investment and operation costs (1). This method is usually used for the final disposal of municipal solid waste. Sanitary landfilling is a complex and heterogeneous physical, chemical, and biological system in which waste materials are decomposed under the influence of compaction, humidity, temperature, and other environmental parameters (1,2). In this biological decomposition system, a malodorous dark liquid with distinctive quantitative and qualitative properties is produced which is called

leachate. This special liquid contains various groups of toxic and dangerous organic and inorganic compounds that threaten the life of soil organisms and cause pollution of surface and groundwater (3). Table 1 presents some compounds and chemical parameters of landfill leachate.

Cyanide is a singly-charged anion containing unimolar amounts of carbon and nitrogen atoms triply-bonded together C=N. It is a strong ligand that can react with all heavy metals at low concentration. Cyanide is highly toxic and the lethal dose for human adults is between 50 and 200 mg (5). This anion exists in different forms such as salts, ions, and metal complexes in municipal and industrial

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Table	1.	Organic	and	inorganic	chemical	components	in	the
leacha	ite d	of municip	oal sc	olid waste la	andfills (4)			

Parameter	Range
COD (mg/L)	150-100000
BOD ₅ (mg/L)	100-90 000
Benzene (µg/L)	1-1630
Ethyl benzene (µg/L)	1-1680
Carbon tetrachloride (µg/L)	3-995
Chloroform (µg/L)	4.4-16
Vinyl chloride (µg/L)	10-3000
рН	5.3-8.5
NH4+ (mg/L)	1-1500
NO3 ⁻ (mg/L)	0.1-50
PO4 ²⁻ (mg/L)	0.3-25
SO4 ³⁻ (mg/L)	10-1200
Cl- (mg/L)	30-4000
CN- (mg/L)	0.04-0.9
Hg^{2+} (µg/L)	0.2-50
As^{2+} (µg/L)	5-1600
Cd^{2+} (µg/L)	0.5-140
Pb^{2+} (µg/L)	8-1020
Cr^{2+} (µg/L)	30-1600
Ni ²⁺ (µg /L)	20 - 2050

wastewaters which is discharged into water resources and other environments (6). The U.S. EPA standard for the pollutants in drinking water is 50 ppb (4). According to Resource Conservation and Recovery Act, all cyanide species are considered acute hazardous materials (5). Cyanide is found in water, soil, and air and enters water through industrial processes such as plating and dyeing as well as pharmaceuticals, insecticides, and photographic films (6). This toxin is present in the household solid wastes such as chemical solvents, dyes, cosmetics, and household pesticides.

Nitrate is a mineral anion that results from the oxidation of nitrogenous compounds. Nitrite is produced by nitrate reduction and rapidly enters the bloodstream and converts the divalent iron in hemoglobin to trivalent iron causing diseases such as methemoglobinemia, headache, and gastrointestinal and respiratory disorders (7).

chemical oxygen demand (COD) is the amount of oxygen required for the chemical oxidation of organic materials in a sample, which is used to measure organic matter contents in wastewater and other environments. This parameter is one of the most common methods for determining the pollution loading of domestic and industrial wastewaters. During the COD test, organic carbonaceous materials are converted to carbon dioxide and water, and the organic nitrogenous compounds are first converted to ammonia and then to nitrite and nitrate (8-10).

One of the effective processes in removing various contaminants from leachate is electrocoagulation. Electrocoagulation is an effective and rapid method for the removal of pollutants and toxic compounds from water or wastewater (11-13). In this process, no coagulant chemical is added to the water and the volume of sludge produced in this technology is less than the one produced by the chemical coagulation method (14-16). Other advantages of electrocoagulation process include absence of toxic residual compounds, removal of various compounds from soluble and insoluble organic pollutants (pesticides, detergents, solvents), soluble inorganic pollutants (heavy metals, nitrate, phosphate, calcium and magnesium), and microbial pathogens elimination (such as bacteria, fungi, viruses) from aqueous environments (17-20). In this technology, the metal ions produced at the anode electrode (iron or aluminum) react with the hydroxide ions formed at the cathode electrode to form the metal hydroxides. These metal hydroxides react and precipitate with ionic compounds, colloids, and suspended solids (15, 16).

The following equations show the mechanism of the electrocoagulation process by the aluminum electrode:

1.	Anodic reaction:	
	$Al_{(s)} \rightarrow Al^{+3}_{(aq)} + 3e^{-1}$	(1)
2.	Cathodic reaction:	

	$3e^++3H_2O \rightarrow H_2(g)^+ 2OH_{(aq)}$	(2)
3	Chemical reaction that takes place in the aqueous	2

5.	Chemical reaction that takes place in the aqueou	.5
	medium:	
	$Al^{+3}_{(aq)} + OH^{-}_{(aq)} \rightarrow Al(OH)_{3(s)}$	(3)

4. Overall reaction: $Al^{+3}(aq)+3H_2O(1) \rightarrow Al(OH)_{3(s)}+3H^+_{(aq)}$ (4)

Based on the literature review, most of the previous studies focused on the performance of electrocoagulation process for removal of biological oxygen demand (BOD), COD, total organic carbon (TOC), and other organic and inorganic compounds from the contaminated water and wastewater. This study aimed to evaluate the effectiveness of electrocoagulation process in removing cyanide, nitrate, turbidity, and COD from the old leachate produced from municipal solid waste to reduce the risks of these pollutants on human and environmental health.

Materials and Methods Sampling method

This study was an experimental one conduced as a continuous flow in Water and Wastewater Chemistry Laboratory in Babol University of Medical Sciences. The leachate sample was prepared from an old solid waste landfill in Ghaemshahr (Iran). Table 2 shows the characteristics of this leachate.

Considering the studied parameters (contact time, pH, current density, electrode type) and optimization

 Table 2. Characteristics of the old leachate of solid waste landfill
 in Ghaemshahr

Parameter	Unit	(Mean ± Standard deviation; n =3)
Odor	-	Medium
Color	-	Dark black
рН	-	8.3 ± 0.2
Temperature	°C	24 ± 1.6
COD	mg/L	19420 ± 1250
Nitrate	mg/L	498.8 ± 50
Cyanide	mg/L	0.1 ± 0.1
Conductivity	µS/cm	1960 ± 100
Total dissolved solids	mg/L	624 ± 35
Total suspended solids	mg/L	440 ± 20
Turbidity	NTU	780 ± 25
Acidity	mg/L	9600 ± 150
Alkalinity	mg/L	11200 ± 215

of operational parameters, the total number of samples required was determined as 162. The samples were taken almost from the middle of the reactor and then passed through Whatman filter 42.

Pilot setup

In this research, a corrosion-resistant Plexiglas reactor (10 cm length, 5 cm width, and 2 mm thickness) was used with a volume of 1 liter and four anode (aluminum with a coating of lead oxide) and cathode (stainless steel) electrodes. The electrodes were in contact with each other in a bipolar arrangement parallel to the leachate sample inside the container at a distance of 5 cm from each other (Figure 1). At each test, the reactor with 0.6 L of leachate was placed in a container containing cold water in order to maintain the system temperature at room temperature. After preparing the reactor and placing the magnet inside the container, the reactor was placed on the heater-shaker. The samples were mixed in the reactor at 100 rpm to prevent clots from settling. Direct current (DC) power supply was used and alternative current (AC) was converted to direct current using MEGATEK model



Figure 1. Laboratory pilot system of electrical coagulation process.

transformer made in Taiwan.

Experimental methods and process optimization

In this study, different parameters such as contact time (10, 20, 30, 40, 50, 60 minutes), pH (5.5, 8.3, 10), current density (11, 22, 33 mA/cm²), type of electrode (aluminum with lead oxide coating) and 3 replications were investigated. In order to reduce the number of samples and optimize the process, at first the removal efficiency of the process in constant current density with different pH and contact times was investigated. After determining the appropriate pH and time, the efficiency of this method was evaluated to determine the optimum density. In this way, the best operating conditions of the desired parameters were obtained and the number of required samples was reduced to 60. The standard method for measuring the parameters as well as device names and models is shown in Table 3.

The removal efficiency of the studied contaminants was obtained by means of equation (5).

$$RE(\%) = \frac{C_0}{(C_0 - C_t)} *100$$
(5)

In this equation, C_t is the concentration of the contaminant at time t, C_0 is the initial concentration of the contaminant at the initial reaction time, and RE is the removal efficiency of a particular contaminant.

Statistical analysis was performed using SPSS statistical software package (version 22.0). All measurements were repeated 3 times and data were expressed as means \pm standard deviation.

Results Effect of pH and reaction times

 Table 3. Standard methods for measurement of the parameters and other characteristics (21)

Parameter	Method	Device name and model
COD	Spectroscopy method	DR5000 (HACH model); Germany
NO3 ⁻	Spectroscopy method	DR5000 (HACH model); Germany
CN⁻	Spectroscopy method	DR5000 (HACH model); Germany
EC	Electrode method	HANNA, Hi 8733; Romany
TDS	Electrode method	TDS meter (C.C.K.); Taiwan
TSS	Gravimetric method	-
рН	Electrode method	pH meter (Aqualytic- pH200); Germany
Turbidity	Nephelometric method	Turbidimeter (HANNA, Hi 93,703); Portugal
Acidity	Titration method	-
Alkalinity	Titration method	-

Amouei et al

Cyanide removal efficiency at different pH and reaction times

The highest CN removal efficiency values during 60 minutes at raw pH (pH = 8.3), alkaline pH (pH = 10), and acidic pH (pH = 5.5) were 96.43%, 100%, and 100%, respectively (Figure 2).

Nitrate removal efficiency at different pH and reaction times

The highest NO3⁻ removal efficiency values after 60 minutes at raw pH (pH=8.3), alkaline pH (pH=10), and acidic pH (pH=5.5) were equal to 60.12%, 93.91%, and 99.65%, respectively (Figure 3).

Turbidity removal efficiency at different pH and reaction times

The highest turbidity removal efficiency values after contact time (60 minutes) at raw pH (pH=3.8), alkaline pH (pH=10), and acidic pH (pH=5.5) were equal to 86.41%, 76.92%, and 64.29%, respectively (Figure 4).

COD removal efficiency at different pH and reaction times

The highest COD removal efficiency values after 60



Figure 2. Cyanide removal efficiency at different pH (current density= 33 mA/cm², cyanide concentration = 0.1 mg/L) (mean \pm standard deviation, n =3)



Figure 3. Nitrate removal efficiency at different pH (current density= 33 mA/cm^2 , nitrate concentration = 499 mg/L) (mean \pm standard deviation, n =3).

minutes of contact at raw pH (pH=3.8), alkaline pH (pH=10) and acidic pH (pH=5.5) were equal to 77%, 83.14%, and 77.94%, respectively (Figure 5).

Current densities and reaction times

Cyanide removal efficiency at different current densities and reaction times

The highest CN removal efficiency values after contact time (60 minutes) at current densities of 11, 22, and 33 mA/cm² were 64.29%, 77.86%, and 100%, respectively (Figure 6).

Nitrate removal efficiency at different current densities and reaction times

The highest nitrate removal efficiency values after 60 minutes of contact at current densities of 11, 22, and 33 mA/cm² were 31.24%, 52.12%, and 93.91%, respectively (Figure 7).

Turbidity removal efficiency at different current densities and reaction times

The highest turbidity removal efficiency values after 60 minutes of contact at current densities of 11, 22, and 33 mA/cm² were equal to 76.92%, 77.24%, and 53.97%,



Figure 4. Turbidity removal efficiency at different pH (current density= 33 mA/cm², turbidity=780 NTU) (mean \pm standard deviation, n =3)



Figure 5. COD removal efficiency at different pH (current density= 33mA/cm², COD=35000 mg/L) (mean ± standard deviation, n =3)

respectively (Figure 8).

COD removal efficiency at different current densities and reaction times

The highest COD removal efficiency values after 60 minutes of contact at current densities of 11, 22, and 33 mA/cm² were 83.14%, 77.33%, and 68%, respectively (Figure 9).

Discussion

The highest COD removal efficiency occurred at alkaline pH (pH= 10) which was equal to 83.14% and the lowest COD removal efficiency was at acidic pH (pH= 5.5). This is because of the effects of pH on the system and the formation of insoluble compounds and flocs in alkaline pH. In equations 1, 2, and 3, Al (OH)₃ and Al (OH)₂ were precipitated and on the other hand, H₂ was gaseous and left the surface. Al (OH)₃ had higher density and specific gravity. When alkaline conditions prevailed in the system, larger and heavier flocs formed and more foam was produced on the surface. As a result, the COD removal efficiency increased. These results are consistent with those of the study by Malakootian et al (22).



Figure 6. Cyanide removal efficiency at different current densities and reaction times (pH=10, CN⁻ concentration= 0.1 mg/L) (mean \pm standard deviation, n =3)



Figure 7. Nitrate removal efficiency at different current densities and reaction times (pH=10, NO_3^- concentration= 499 mg/L) (mean ± standard deviation, n =3)

- 1. In natural pH conditions: $3Al_{(s)}+8H_2O_{(l)}\rightarrow Al(OH)_{2(s)}+2Al(OH)_3+4H_{2(g)}$
- 2. In acidic pH conditions: $2Al_{(s)}+6H_2O_2O_2 (g)+4H_{2(g)}+Al (OH)_{2(S)}$
- 3. In alkaline pH conditions: $2Al_{(s)}+6H_2O_{(I)} \rightarrow 2Al (OH)_{3(S)}+3H_2O_{(I)}$

The results of study by Jotin et al showed that the removal of COD by electrocoagulation was strongly influenced by the raw pH and it was found to be very effective when the raw pH range was between 4 and 8. The highest COD removal efficiency (74.08%) occurred at 10 volts, conductivity of 28 mS/cm², raw pH 4, and contact time of 100 minutes (15). In the study by Sivakumar et al on the removal of COD and TOC from landfill leachate by electrocoagulation process, the results showed that the highest removal efficiency values of TOC and COD were 94.7% and 98.2%, respectively. Temperature was 50°C, current density was 40 mA/cm², and stirring speed was 50 rpm (17).

The highest cyanide removal efficiency was related to alkaline pH (pH= 10) and acidic pH (pH= 5.5). The increase in cyanide removal efficiency under alkaline conditions is due to aluminum hydroxide precipitation



Figure 8. Turbidity removal efficiency at different current densities and reaction times (pH=10, turbidity=780 NTU) (mean \pm standard deviation, n =3)



Figure 9. COD removal efficiency at different current densities and reaction times (pH=10, COD=35000 mg/L) (mean \pm standard deviation, n =3)

and the production of the associated flocs. Under acidic conditions, the hydrogen ions produced complex compounds of sediment with cyanide ions (6). Therefore, the removal efficiency of cyanide in these conditions increased.

The highest NO₃ removal efficiency occurred at acidic pH (pH= 5.5) which was equal to 99.65%. At alkaline pH (pH=10) and natural pH (pH=8.3), the removal efficiency values of NO, were 93.91 and 60.5%, respectively. The high efficiency of NO₃ removal by the electrocoagulation process in alkaline pH, can be attributed to the reaction between the aluminum electrode and the hydroxide ions in the solution leading to insoluble aluminum hydroxide and floc formation, in which soluble nitrate can be removed by flocs through precipitation or adsorption processes (7,23). Moreover, with increasing pH of the solution, the inactive aluminum oxide on the electrodes disappears (24). At acidic pH, due to the combination of nitrate ions with positive hydrogen ions and the formation of complex compounds, nitrate removal occurred more rapidly as a result of the formation of very small bubbles with high density (8,25). Huang believed nitrate reduction at acidic pH is possible in two ways: 1) H⁺ ions participate directly in the nitrate reduction reaction as nitrate complex compounds. 2) H⁺ ions affect nitrate uptake at the reactive site (26,27). Lee et al investigated the removal of ammonia nitrogen and COD from landfill leachate by electrocoagulation. In this study, the highest removal efficiency values of COD and ammonia nitrogen were 49.8% and 38.6%, respectively, at current density of 4.96 mA/cm², normal pH, chloride concentration of 2319 mg/L with iron electrode, and the contact time of 90 minutes (16).

The highest turbidity removal efficiency (86.41%) occurred at pH 8.3 and the lowest turbidity removal efficiency (67.5%) happened at acidic pH. The results indicated that at higher pH, the turbidity removal efficiency decreased. The reason for the decrease in turbidity, as mentioned earlier, was the production of Al (OH)₃ coagulants at alkaline pH as well as the production of the bubbles caused by H₂ gas, which was reduced by the two mechanisms of sweeping coagulation and flotation (28-30). In this regard, Kiliç and Hoşten et al estimated that the highest turbidity removal efficiency by electrocoagulation using aluminum electrode happens at pH 9 (31). On the other hand, in the study conducted by Marzouk et al, the optimal pH for turbidity removal from industrial wastewater was 8 (32). Solak et al showed that the pH 9 was the optimum pH for the turbidity removal using aluminum electrode in electrocoagulation process (33).

In this study, when the current density and reaction time increased, the removal efficiency values of cyanide and nitrate ions, turbidity, and COD increased. The reason for the direct effect of current density on the removal efficiency can be explained by several mechanisms that occur at the anode and cathode electrodes. The main products in the cathode electrode are hydrogen gas and OH⁻ ions and the main products in the anode electrode are Al³⁺, AL²⁺ and H⁺ ions. By connecting an electric current between the anode and cathode electrodes, aluminum ions (Al_{2}^{+}) at the anode and hydroxide ions (OH-) at the cathode electrodes combine together and produce Al (OH)₃. The large surface area of aluminum hydroxide flocs can trap organic and inorganic compounds in leachate through some mechanisms such as ionic compaction, electric charge neutralization, sweeping coagulation, and Interparticle bridging (34). In this study, the organic compound (COD) in leachate was removed by the complexation mechanisms and electrostatic adsorption. Cyanide and nitrate ions were removed by sweeping coagulation mechanism (5,7). Light flocs were also removed by the hydrogen gas bubbles produced at the cathode electrode (32). As a result of these mechanisms, the amount of turbidity was also significantly reduced. The released ions (Al $^{3+}$ H+, and OH) based on electric charge neutralization mechanism neutralized the electric charge of the particles and thus coagulation process was formed (35). The removal efficiency of any pollutant directly depends on the concentration of ions presented in the solution and reaction time. As the reaction time of the process increases, the concentration of ions produced from electrodes also increases and as a result, hydroxide flocs increase (27,36). Therefore, with increasing retention time, the removal efficiency of the pollutant increases. These results are consistent with those of the study by Takdastan et al in the removal of COD, turbidity, detergent, and phosphate (37).

Conclusion

This study investigated the removal of organic and inorganic parameters including COD, cyanide, nitrate, and turbidity from old leachate of municipal solid waste by electrocoagulation process. A total of 162 samples were prepared from the old leachate of solid waste landfill in Ghaemshahr, Iran. In order to reduce the number of samples in the electrocoagulation process, the parameters were optimized. Moreover to increase the contact surface of the electrodes with leachate samples and the removal efficiency of the evaluated parameters, 6 holes with a diameter of 7 mm were made on the aluminum electrode. Although in this study, the efficiency of cyanide removal in different pH was high, it was maximal (more than 95%) in acidic and alkaline conditions. The maximum removal of nitrate (99.65%) and turbidity (86.41%) were obtained at pH 5.5 and 8.3, respectively. The high removal efficiency of COD (83.14%) was at pH 10. Due to the proper removal efficiency of nitrate and cyanide pollutants in leachate by electrocoagulation technology, the nitrate and cyanide concentrations reached less than the allowable contamination level. However, due to the COD removal efficiency in this method and high contents of COD in

the old leachate, after electrocoagulation, the COD level (3200 mg/L) did not reach the allowable concentration of COD in the effluent standards. Therefore, after reducing the organic load of leachate by electrocoagulation process, it is necessary to design a final treatment unit for removal of landfill leachate. These results showed that the electrocoagulation process can be efficient for removal of the cyanide, nitrate, turbidity, and COD from a landfill leachate.

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Ethical issues

This research project was approved under the ethical code IR.MUBABOL.HRI.REC.1397.081. The authors certify this manuscript is the original work of the authors, all data collected during the study are presented in this manuscript, and no data from the study has been or will be published separately.

Competing interests

The authors declare that they have no conflict of interests.

Authors' contributions

All authors contributed equally to this work and participated in the data collection, analysis, and interpretation. All authors critically reviewed, refined, and approved the manuscript.

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