

## Optimization of Electrocoagulation on Removal of Wastewater Pollutants

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### Abstract

In this work the performance of electrocoagulation (EC) process was investigated and optimized under different operational conditions for the best removal of wastewater contaminants such as Nitrate, total hardness, Calcium and Magnesium. Samples were obtained from Gaza Wastewater Treatment Plant (GWWTP). Under the optimal condition (pH=7.45, inter-electrode spacing=1 cm, operating time=40 minutes and current density=3.18 mA/cm<sup>2</sup>), the results showed that the removal efficiency of total hardness, Calcium, Magnesium, Nitrate were 94.6%, 93.3%, 95.2% and 70.9% respectively by using stainless steel electrodes, while for aluminum electrodes the results were 92.83%, 93.33%, 92.30%, 50.43% respectively. For Iron electrodes, the removal efficiencies of contaminants were 87.84%, 88%, 87.64%, 57.26% respectively. In addition, the experimental results also showed that the effluent wastewater was very clear, odorless and its quality is fit for reuse.

**Keywords** Electrocoagulation; Removal efficiency; Wastewater treatment; Electrodes types; Total hardness; Calcium; Magnesium; Nitrate

### Introduction

The reuse of wastewater has become an absolute necessity. Demands to the cleaning industrial and domestic wastewater to avoid environmental pollution and especially contamination of pure water resources are becoming national and international issues. The Gaza Strip is described as one of the most exploited places in the world where the level of demand on water and land resources exceed the capacity of the environment due to water shortage, contamination of water resources, densely populated area and highly intensive irrigated agriculture characterize. The water balance records revealed a water deficit of  $80 \times 10^6$  m<sup>3</sup> in 2012 [1]. The use of wastewater as a supplemental source of irrigation is inevitable for increased agricultural production in the Gaza Strip, where irrigation supplies are insufficient to meet crop water needs. Moreover, irrigation with treated wastewater is considered a promising practice that helps in minimizing the pollution of the ecosystem subjected to contamination by direct disposal of wastewater into surface or groundwater. The treated wastewater has several advantages over other sources of water, it minimizes pollution, augments groundwater resources by artificial recharge and it is a good nutrient source for landscape and farm irrigation [2]. Different technologies have been reported for the treatment of water and wastewater such as: nanotechnology [3-7], Photocatalytic [8], Advanced Oxidation Processes [9] and Solar Energy [10]. In the last years, there is an increasing interest in the development of environmentally friendly electrochemical methods to treat of water. Electrocoagulation (EC) is one of the novel methods for wastewater treatment. The EC process possesses several advantages such as easy operation, short treatment time, low sludge production and no chemical requirement [11,12]. A literature survey indicates that EC is an efficient treatment process for different wastes, e.g. soluble oils,

liquid from the food, textile industries and effluents from the paper industry, aquacultural wastewater, textile wastewater, herbicide and polymer [13-21].

In this study, electrocoagulation has been suggested as an advanced alternative in pollutant removal from wastewaters to improve the effluent quality prior to wastewater reuse for agriculture purposes. This research is mainly focused on the capability of EC technology to improve wastewater quality by removal of Nitrate, Total hardness, Calcium, Magnesium and other pollutants from the Gaza wastewater treatment plant under different operational conditions.

### Experiment

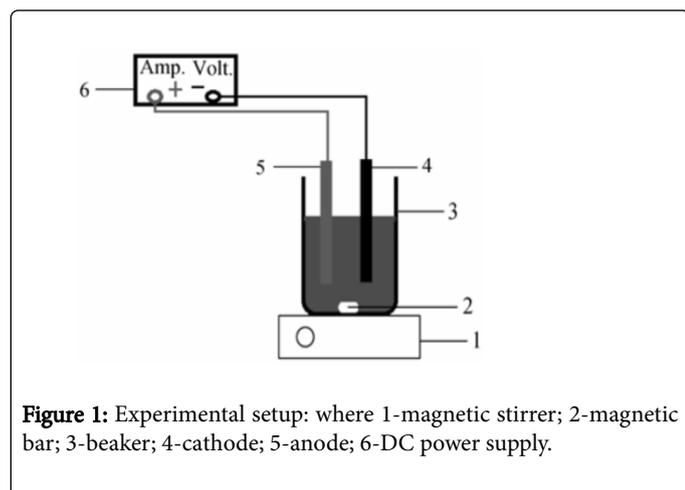
#### Chemicals

Sodium chloride, sodium sulfate, sodium carbonate, potassium nitrate, sodium hydroxide, sulfuric acid and potassium dichromate, were of analytical grade and purchased for the preparation of standard solution from Merck. Standard solutions of potassium dichromate (K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) reagent with silver sulfate (Ag<sub>2</sub>SO<sub>4</sub>) and Mercury sulfate (HgSO<sub>4</sub>) were prepared to measure the COD.

#### Equipment and procedure

For electrochemical tests in this work, different electrodes types were used such as stainless steel (Ss), iron (Fe) plate and aluminium (Al) electrodes. The total effective electrode area and the distance between electrodes, electrode type, electrode size, pH, current intensity and time were used as variable factors may affect the pollutants removal efficiencies. Before each run, electrodes were washed with Potassium Chloride solution to remove surface grease. At the end of each run, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces and dried. The experiments were conducted in a 1000 ml glass beaker in batch mode of operation. The volume of wastewater sample was 600 ml. Aluminium; iron and

stainless-steel sheets with dimensions of 270 mm × 40 mm × 1.8 mm (length×width×thickness) were used as electrodes. The area of electrodes dipped into the solution was 50.88 cm<sup>2</sup>. Power supply was started at time zero (t=0) and it was the starting time of the EC process. After 30 min, 40 min, 1 hour, the samples were withdrawn, filtered using filter paper (0.45 μm), and analyzed. The schematic diagram of the experimental set up is shown in Figure 1.



### Analysis

The equation used to calculate the wastewater removal efficiency and COD in the treatment experiments was [22,23]:

$$CR = \frac{C_0 - C_1}{C_0} \times 100\% \quad (1)$$

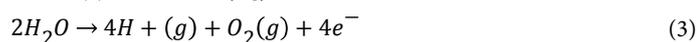
Where,

CR is the removal efficiency percentage; C<sub>0</sub> is initial concentration of wastewater before electrocoagulation in mg L<sup>-1</sup>.

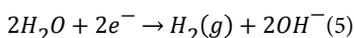
### Results and Discussion

In an EC experiment the electrode or electrode assembly is usually connected to an external DC source. The amount of metal dissolved or deposited is dependent on the quantity of electricity passed through the electrolytic solution [24]. If in this process M is considered as anode, the following reactions will occur [25]:

At anode:



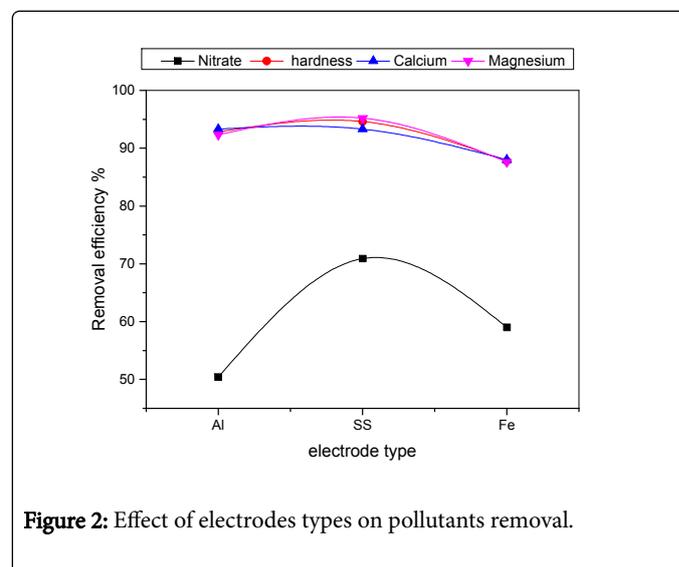
At cathode:



Where M is the material used as electrode and n is the number of electrons.

### Effect of electrodes materials

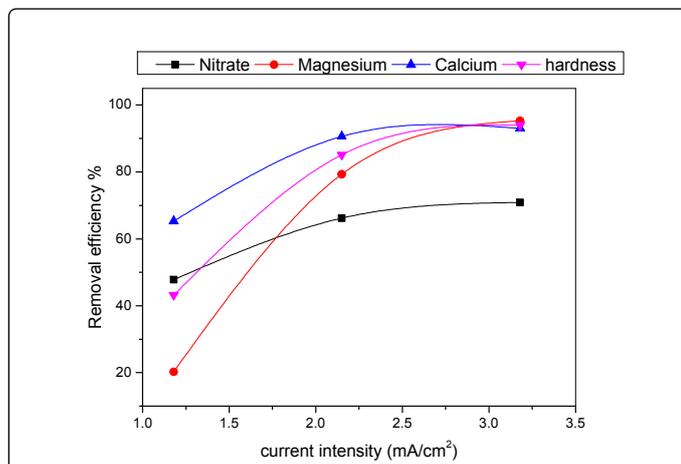
Electrode assembly is the heart of the present treatment facility. The most common electrode materials for electrocoagulation are iron, aluminum and stainless steel [26]. The experimental removal efficiencies for Total hardness, Calcium, Magnesium, and Nitrate were (92.83%, 93.33%, 92.30% and 50.43%) respectively for aluminum electrode as shown in Figure 2 and for stainless steel electrode removal efficiencies of the above mentioned water quality parameters (Total hardness, Calcium, Magnesium and Nitrate) were 94.26%, 93.33%, 95.19% and 70.94%, respectively and for iron electrode were 87.84%, 88%, 87.64% and 57.26%, respectively. The effluent treated with iron electrode, appeared firstly greenish color and then turned yellow color and turbid in the first minutes. This green and yellow color may be resulted from Fe<sup>2+</sup> and Fe<sup>3+</sup> ions generated during EC process. Fe<sup>2+</sup> is the common ion generated in situ of electrolysis of iron electrode. It has relatively high soluble at acidic or neutral conditions and can be oxidized easily into Fe<sup>3+</sup> by dissolved oxygen in water [27], where the effluent treated with aluminum electrode appeared as the first time white and stay white all the process, no sludge settled remarked, only white foam is formed as the electrode was eroded and liberated trivalent aluminum (Al<sup>3+</sup>). The (Al<sup>3+</sup>) formed an ionic pair with the pollutant of wastewater rich in magnesium and Calcium. There was formation of a strong coagulant. An excellent flocculation and coagulation was observed. The Stainless steel electrodes used to treat the effluent release in the first of process black color and after a few minutes it starts to appear clear and a little of sludge is formed. In general the electrocoagulation process increases due to the formation of metal hydroxide species which adsorbed the pollutants molecules, and this causes the increase of the removal efficiency (Figure 3) [28-29].



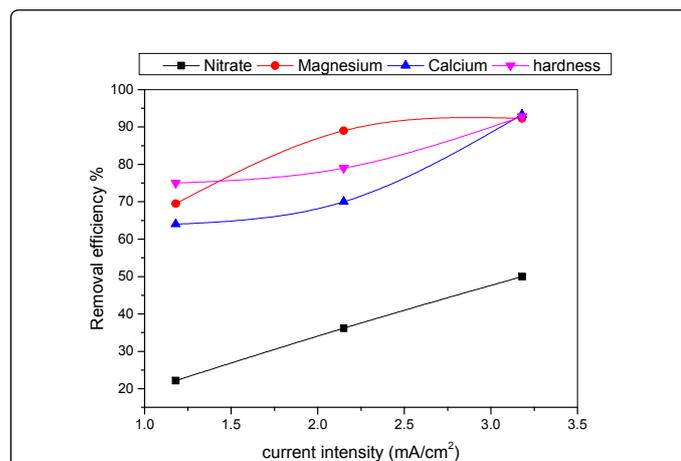
### Effect of current density (mA.cm<sup>-2</sup>)

The applied current controls the anode dissolution speed on one hand and the formation of hydrogen on the other hand [30]. The influence of the variation of this parameter between (1.18–3.18 mA.cm<sup>-2</sup>) has been examined on the removal efficiency for the total hardness, Calcium, Magnesium, and Nitrate at 40 minutes, electrode distance of 1cm and pH 7.45. Figures 3 and 4 indicate that a current of 3.18 mA/cm<sup>2</sup> give the maximum rates of pollutants removal for total hardness, calcium, magnesium and nitrate using stainless steel and

aluminum electrode. The increase of current density increases the number of metal cation dissolution from anode, formation of H<sub>2</sub> bubbles (Figures 3 and 4). However, the higher the current density the smaller the size of the hydrogen bubbles which lead to a faster removal of pollutant [30,31].



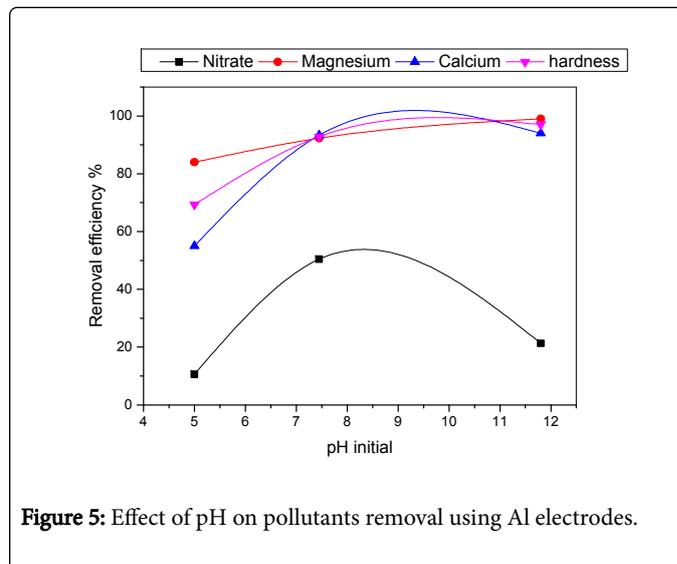
**Figure 3:** Effect of current density on pollutants removal using SS electrodes.



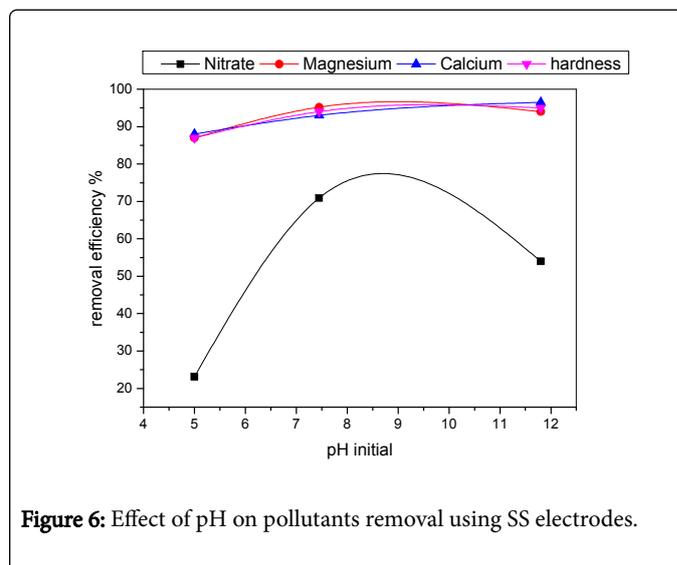
**Figure 4:** Effect of current density on pollutants removal using Al electrodes.

### Effect of initial pH

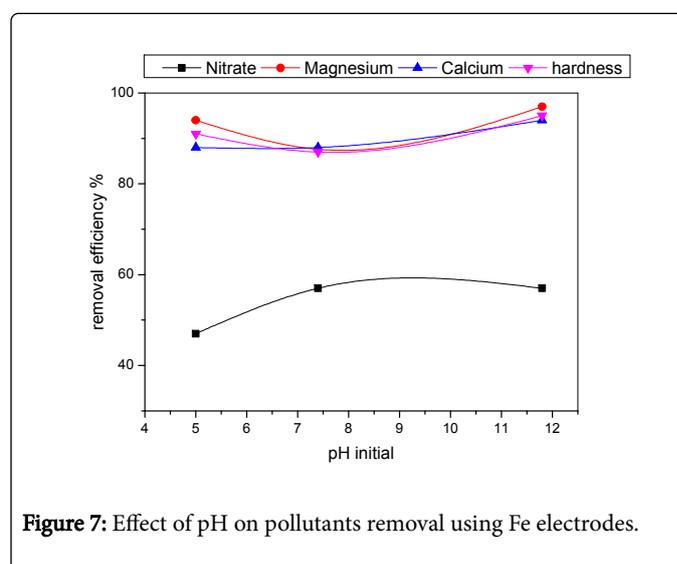
The process of electrocoagulation is proved to be highly pH dependent. Experiments were performed by applying an initial pH in the range of (5-11.8) at 40 minutes, electrode distance of 1cm and current density 3.18 mA/cm<sup>2</sup>. As shown Figures 5-7, total hardness, calcium and magnesium removal efficiencies were found to be the best near basic pH using the three types of electrodes as they were above 95%, but the highest one was by using aluminum electrode as it achieves 97.43%, 94.67, and 100% respectively. As the operating time of EC process increased the pH value increased. This is due to the OH<sup>-</sup> ion accumulation in aqueous solution during the process. From figure 5-7, it can be seen that the removal efficiency of waste water was increased by increasing the pH.



**Figure 5:** Effect of pH on pollutants removal using Al electrodes.



**Figure 6:** Effect of pH on pollutants removal using SS electrodes.



**Figure 7:** Effect of pH on pollutants removal using Fe electrodes.

### Effect of operating time

The EC time is a significant parameter which has a vital importance and influence on the performance of EC process. In this experiment the effect of time was studied at constant current density of 3.18 mA/cm<sup>2</sup> and constant distance 1 cm. Table 1 illustrates the effect of retention time on the removal of chemical and physical pollutants using three different electrodes (Al, Ss and Fe) at neutral pH. In this process, EC involves two stages which are destabilization and aggregation. The first stage is usually short, whereas the second stage is relatively long. Results show that the efficiency starts to be significant for some pollutant at the treatment time of 30 minutes but the maximum efficiency was obtained at a treatment time of 40 minutes. Treatment time has significant improvement in the removal efficiency for the studied pollutants. It is assumed that the more time consumed, especially above 30 minutes, the more production rate of hydroxyl and metal ions on the electrodes.

Time (min)	Electrode types	Pollutants Removal%					
		Hardness	Calcium	Magnesium	Turbidity	Nitrate	TDS
20	Al	79.05	70	88.33	72.21	27.35	20.45
	Ss	82.43	78.66	86.26	47.02	22.22	17.05
	Fe	80.4	73.33	77.88	71	23.08	11.36
30	Al	92.57	92	93.13	76.13	35.9	22.73
	Ss	89.86	92	87.64	81.57	26.5	20.45
	Fe	81.76	85.33	78.02	74.62	31.62	14.77
40	Al	92.83	93.33	92.3	95.2	50.43	22.95
	Ss	94.26	93.33	95.19	94.56	70.94	29.09
	Fe	87.84	88	87.64	93.35	57.26	22.05

**Table 1:** Influence of time on pollutants removal using three types of electrodes

### Effect of distance between electrodes

To investigate the influence of electrode distance on the electrocoagulation process for the removal of pollutants from effluent, different spaces between the electrodes were applied. According to Bukhari [32], increasing the electrodes spacing will reduce the capital cost of treatment but may reduce the treatment efficiency. Inter-electrode spacing of 1, 2 and 3 cm was studied to examine the effect of

electrode distance on the EC process for Stainless steel and Aluminum electrodes. Table 2 summarizes the influence of distance between electrodes on the removal of physical and chemical pollutants using three types of electrodes, Al, Fe and SS. The analysis reveals that the removal efficiency for EC process for total hardness, calcium, magnesium, turbidity, nitrate, TDS increased with the decrease in the inter-electrode spacing. For total hardness, calcium and magnesium, an inter-spacing electrode of 1 cm marked a significant percent of removal at 40 minutes for all type of electrodes.

In addition the odor was disappear by using stainless electrodes, and slightly smelt with iron and Aluminum electrodes, and the color of the sample had changed to colorless for all type of electrodes. Where removal of COD during the experiment in the range of 74% for stainless steel, 68.18% for aluminum and 67.27% for iron in an interval of 30-40 minutes at neutral medium. The COD removal by aluminum and iron electrodes was thought to be mainly adsorption of pollutants onto aluminum and iron hydroxide sludge.

Distance between electrodes (cm)	Electrode types	Pollutants Removal %					
		Hardness	Calcium	Magnesium	Turbidity	Nitrate	TDS
1	Al	92.83	93.33	92.3	95.2	50.43	23
	Ss	94.26	93.33	95.19	94.56	70.94	29.1
	Fe	87.84	88	87.64	93.35	59.26	22.1
2	Al	82.43	80	84.89	93.6	29.91	18.6
	Ss	89.86	90.67	89.01	93.6	62.82	29.6
3	Al	72.97	53.33	93.13	92.54	24.36	20.7
	Ss	88.51	89.33	87.64	89	48.29	20

**Table 2:** Influence of inter electrode distance on pollutants removal using different type of electrodes.

### Treated Wastewater Reuse

In this research the technology of EC is investigated to achieve the quality standard for wastewater irrigation. Recently the Environment Quality Authority with coordination of Palestinian ministries and universities has established specific wastewater reuse regulations. Table 3 presents the characteristics of the wastewater used in the experiments before EC where Table 4 presents the values of chemical, physical and biological contaminants after EC treatment and the Palestinian standards for wastewater reuse. According to the results of the EC experiments, the quality of treated wastewater in general is within the acceptable limits for all investigated parameters.

Parameters	Unit	Value
pH	-	7.45
COD	mg O <sub>2</sub> /l	550
Color	-	dark brown
Turbidity	NTU	33.1
Conductivity	μS/cm	4400
TDS	mg/l	2728
Hardness	mg/l	740
Calcium	mg/l	150.4
Magnesium	mg/l	88.34
Sodium	mg/l	575-600
SAR	-	9.18
Nitrate	mg/l	23.4
Odor	-	very bad
Fecal coliform	colonies/100 milli liters	2*10 <sup>5</sup>
Chloride	mg/l	920

**Table 3:** Characteristics of the wastewater used in the experiments

Pollutants	Type of electrodes			Palestinian Standards
	Al	Ss	Fe	
Calcium (mg/l)	10	10	18	400
Magnesium (mg/l)	6.79	4.25	19.92	60
SAR	7.01	7.68	7.52	9-10
Nitrate (mg/l)	11.6	6.8	10	15-50
Turbidity (NTU)	1.95	1.8	2.2	NA
EC (μS/cm)	3390	3120	3430	2400-3000
TDS (mg/l)	2102	1934	2127	1200-1500
pH	9	8.2	8.1	6-9
COD (mg/l)	175	139	180	150-200
Fecal coliform (colonies/100 ml)	0	0	1	200-1000
Colour	free	free	free	Free

**Table 4:** Comparison of chemical, physical and biological values of the Palestinian standards with effluent after EC treatment.

### Conclusion

The optimized conditions for this process were as follows: a current density of 3.18 mA/cm<sup>2</sup>, inter-electrode spacing of 1 cm, retention time of 40minutes and neutral pH (7.45).

The different electrode materials had an effect on the effectiveness of wastewater treatment because of its mechanisms. Aluminum electrodes marked the highest removal in turbidity (95.20%). Stainless steel electrodes marked highest removal of total hardness (94.26%), calcium (93.33%), and magnesium (95.19%), TDS (29.09%), nitrate (70.94%) and turbidity (94.56%). Iron electrodes have good removal efficiency and can also be applied for wastewater treatment as it is the cheapest one.

The applied current density has important effect on the removal efficiency of EC process. It was found that rising the current density from 1.15 to 3.18 mA/cm<sup>2</sup> increases the removal efficiency for studied pollutants.

Removal of COD during the experiment in the range of 74% for stainless steel, 68.18% for aluminum and 67.27% for iron electrode

The results obtained after EC meet the Palestinian standards for wastewater reuse.

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