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Research Article

ELECTROCOAGULATION USING STAINLESS STEEL ANODES: THE REMOVAL OF NITRATE FROM AGRO-BASED GROUNDWATER

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Abstract: *This study covers groundwater treatment for nitrate (NO_3^-) removal by the electrocoagulation (EC) method for March (pre-irrigation) and October (post-irrigation) of 2023 for ten observation points in an arid and semi-arid region. Stainless steel (SS) plate electrodes were used in the process, and energy consumption and cost assessment were also carried out. The study area is covered with fertile agricultural lands, and the people of the region make their living from agricultural activities. Therefore, the actual groundwater samples used in the study contain high concentrations of NO_3^-. The effectiveness of the main operational parameters, such as initial pH, electrical conductivity, and current density, was analyzed. It was observed that nitrate removal at the highest electrical conductivity value (2239 µS/cm) had the lowest energy consumption value (0.81 kWh/m^3). The SS values measured at the end of the EC process were < 0.4. In other words, the SS value passing into the water is very low and has no adverse effect on human health. In this study specific to the Harran Plain, a cost of \$1.06 per m^3 was calculated for nitrate removal using current electricity costs.*

***Keywords**: Electrocoagulation, energy consumption, nitrate removal, stainless steel.*

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1. Introduction

The electrocoagulation (EC) process utilizing stainless steel electrodes has emerged as a promising method for the treatment of nitrate-contaminated groundwater. This technique leverages the electrochemical reactions that occur when an electric current is applied to electrodes submerged in water, leading to the destabilization and aggregation of pollutants, including nitrates (NO_3^-). The efficiency of this process is influenced by several factors, including electrode material, current density, pH, and operational parameters. Electrocoagulation operates by generating metal hydroxides through the dissolution of the electrodes, which then interact with dissolved contaminants. Stainless steel (SS) electrodes are particularly advantageous due to their durability and resistance to corrosion, which can enhance the longevity of the treatment system [1,2]. Studies have shown that stainless steel electrodes can achieve significant nitrate removal efficiencies, often exceeding 90% under optimal conditions [3,4]. For instance, research indicates that the electrocoagulation process can effectively reduce nitrate levels in groundwater, with operational parameters such as current density and electrode spacing playing critical roles in optimizing removal rates [5,6]. The mechanism of nitrate removal via EC involves both adsorption and electrochemical reduction. The hydroxide ions generated at the cathode facilitate the formation of flocs that capture nitrate ions, while the applied electric current promotes the reduction of nitrates to nitrogen gas or ammonia [7,8]. This dual mechanism not only enhances the removal efficiency but also allows for the potential recovery of valuable by-products, such as ammonia, which can be utilized in various applications [9,10]. Various traditional techniques such as adsorption, ion exchange, reverse osmosis, electrochemical, chemical, and biological methods have been developed to remove nitrate from water [11,12]. Furthermore, the integration of electrocoagulation with other treatment methods, such as adsorption, has been explored to further enhance nitrate removal efficiencies [13].

The EC process has emerged as a promising technology for the removal of nitrates from groundwater, particularly when utilizing SS electrodes. This method leverages the electrochemical generation of coagulants, primarily metal hydroxides, which destabilize and aggregate contaminants, facilitating their removal from aqueous solutions. The efficiency of EC in treating nitrate-laden water is influenced by various operational parameters, including electrode material, current density, pH, and contact time, which collectively dictate the effectiveness of the process. Stainless steel electrodes have garnered attention due to their durability and effectiveness in electrocoagulation applications. Studies have shown that stainless steel electrodes can effectively generate the necessary coagulants while exhibiting resistance to corrosion, which is a significant advantage over more traditional materials like aluminum or iron [3,14]. The use of stainless steel also minimizes the risk of introducing additional contaminants into the treated water, which is a critical consideration in groundwater remediation efforts [15]. The operational parameters of the electrocoagulation process are crucial for optimizing nitrate removal. Research indicates that the optimal contact time for effective nitrate removal typically ranges from 15 to 60 minutes, with a peak efficiency observed at around 30 minutes [16]. Additionally, the current density applied during the electrocoagulation process plays a pivotal role in determining the rate of nitrate removal. Higher current densities have been associated with increased production of hydroxides, thereby enhancing the coagulation process [1]. However, it is essential to balance the current density with energy consumption, as excessively high values may lead to diminishing returns in nitrate removal efficiency and increased operational costs [8]. The pH of the solution is another critical factor influencing the electrocoagulation process. The removal efficiency of nitrates tends to be higher at acidic pH levels, where the presence of hydrogen ions can facilitate the reduction of nitrate ions [17]. In addition to the operational parameters, the configuration of the electrocoagulation system, including the distance between electrodes and the arrangement of the electrodes, can significantly impact the treatment efficiency. Studies have shown that maintaining an optimal gap between stainless steel electrodes can enhance the electric field strength, thereby improving the coagulation process [14]. Furthermore, the design of the electrocoagulation reactor, such as the use of batch versus continuous flow systems, can also influence the overall effectiveness of nitrate removal [18]. The integration of electrocoagulation with other treatment methods can further enhance nitrate removal efficiencies. For instance, coupling electrocoagulation with activated carbon filtration has been shown to improve the overall quality of treated water by not only removing nitrates but also adsorbing residual contaminants [19]. This two-step approach allows for a more comprehensive treatment strategy, addressing multiple contaminants simultaneously and improving the sustainability of groundwater treatment practices. The environmental implications of nitrate contamination in groundwater are significant, as elevated nitrate levels can lead to adverse health effects and ecological disturbances. The electrocoagulation process offers a viable solution to mitigate these risks by providing an efficient and environmentally friendly method for nitrate removal [20,21].

The central and southern parts of the Harran plain basin have good groundwater potential. While very good groundwater potential is found regionally towards the north, other parts of the basin are generally at moderate levels. In some regions, groundwater potential is almost non-existent and therefore, it is characterized as poor. Unfortunately, the fact that agro-based nitrate pollution is encountered in the waters in the Harran Plain region, where groundwater potential is characterized as good, negatively affects the quality of the waters here. The point of this study was to look at the quality of groundwater in the Harran plain, which is mostly used for farming, and see how well the electrocoagulation process worked at getting rid of nitrates in water samples that were taken from severely nitrate-contaminated groundwater before and after irrigation. This strategy is anticipated to enhance its applicability in other places exhibiting similar features to the study area.

2. Materials and Methods

2.1. Description of study area

The Harran Plain is situated in the Sanliurfa-Harran irrigation area. The plain, about 30 km in breadth and 50 km in length, is situated in the Southeastern Anatolia Region (36° 43' - 37° 10' N and 38° 47' - 39° 10' E). The region, the largest component of the GAP Project, encompasses 141,855 hectares of irrigable land, a drainage area of 3,700 square kilometers, and the plain area of 1,500 square kilometers. In the area characterized by a semi-arid climate, precipitation is virtually absent from June to September. The long-term annual precipitation is 284.2 mm, the temperature is 18 °C, and the evaporation is 1848 mm. The overall gradient orientation is from north to south, ranging from 0% to 2%. The terrain gradient is nearly level in the vicinity of Harran and Akçakale districts. The level terrain contributes to issues related to agricultural water runoff and drainage complications. The soils are clay-rich, with pH values ranging from 7.5 to 8.0. The minimum permeability is 0.22 m/day, while the maximum permeability is 3.51 m/day. Seventy-seven percent of the land has soil profiles above 150 cm in depth [22].

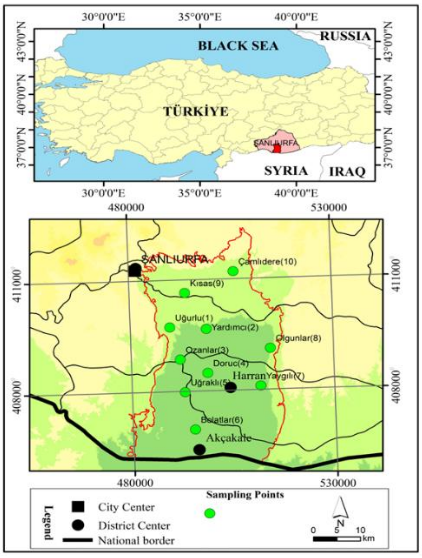

Figure 1. The map of the study area. *Sampling Points: Uğurlu (1), Yardımcı (2), Ozanlar (3), Doruç (4), Uğraklı (5), Bolatlar (6), Yaygılı (7), Olgunlar (8), Kısas (9) and Çamlıdere (10).

2.2. Sample collection

The primary concern to Harran Plain groundwater is the pollutant characteristics transported by agricultural return irrigation flows. Sampling was conducted in the Harran Plain over two intervals: pre-irrigation in March 2023 and post-irrigation in October 2023. Initially, samples were collected from 10 wells at specified ratios (Figure 1). Harran Plain contains two types of aquifers: shallow and deep. The selected ten sampling locations illustrate the shallow aquifer of the plain [23]. Groundwater sampling was conducted in the shallow aquifer. Water samples collected from the sampling locations were processed in compliance with the laboratory conditions established according to the norms at Harran University. In-situ and laboratory analyses were performed on these samples. This study conducted nitrate analyses in October and March utilizing the ICP-MS technique in accordance with Standard Methods. During sampling, parameters such as groundwater level, temperature, conductivity, pH were measured in-situ. The groundwater level of the wells was determined by a well meter. pH measurements were made with a WTW Multi 340i device and conductivity and temperature measurements were made with a Hach HQ40D device.

2.3. Design of Electrocoagulation Units for Nitrate Removal

Glass with dimensions of 100 x 100 x 250 mm served as the EC reactor in the experimental studies. The study used seven plate electrodes measuring 96 × 180 × 1.5 mm (purity P 99.5%). A batch reactor carried out the EC process to remove nitrate from groundwater using SS electrodes. Figure 2 illustrates the process, while Table 1 displays its characteristics. The minimum distance between the electrodes with monopolar connection was determined to be 10 mm. The Rigol DP832A Programmable 3-Channel DC Power Supply provided current and voltage control. Mixing was carried out with the IKA RH Basic 2 brand magnetic stirrer.

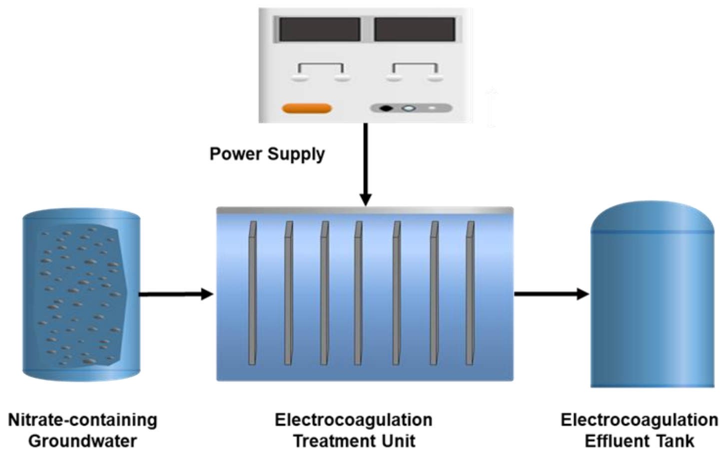

Figure 2. Experimental setup used in the current batch EC process studies.

The pH, conductivity, and temperature of the samples taken at regular intervals during the reactor operation period were measured according to standard methods for examination of water and wastewater. The experiments were carried out in three replicates.

3. Results and Discussion

The Harran Plain saw a rapid level rise from the start of the irrigation period in 1995 until 1998. A rise of about 25 m can cause damage to groundwater quality, such as increased conductivity values due to increased salinity. After 2006, the increase in water level continued (about 15 m), and in 2016, the groundwater levels in the wells reached alarming levels (up to a depth of about 2 m below the ground). According to the data obtained within the scope of this study, this level gradually increased and reached approximately 1.9 meters.

Examining the pre-irrigation and post-irrigation periods reveals that many sampling points have nitrate concentrations exceeding the permitted 50 mg/L. The conductivity values showed little change before and after irrigation periods. Furthermore, the measurements at the same points revealed very similar results. Examining the pH values revealed similar results in the pre- and post-irrigation periods. This indicates that there is a direct proportional relationship between nitrate and electrical conductivity.

3.1. Nitrate concentrations were obtained in water analyses

The graph in Figure 3 displays the nitrate concentration values pre-irrigation (March) and post-irrigation (October). Fertilizers used in agricultural applications cause nitrate pollution, which belongs to the highest risk class of pollutants. The accumulation of nitrate in the soil is a result of increased agricultural production activities and fertilizer use. Different environmental conditions wash out accumulated nitrate, causing it to move deeper into the soil. Nitrate-reducing bacteria in the soil convert fertilizer to nitrate through nitrification, and because nitrate has a negative charge, it washes away and reaches the groundwater. Even under normal conditions, it is stated that only 50% of nitrogen fertilizers applied to the soil are used by plants, 2-20% is lost through evaporation, 15-25% is combined with organic compounds in clay soil, and the remaining 2-10% is mixed with surface and groundwater [23].

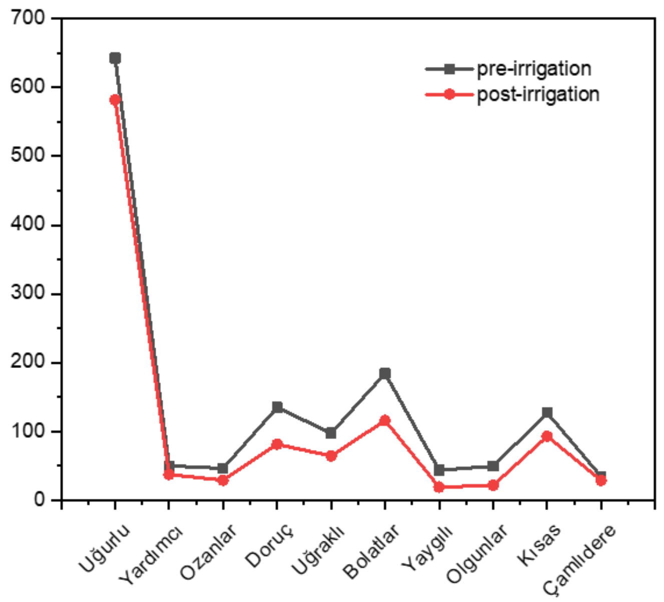

Figure 3. Nitrate values measured in the samples taken from the sampling points in pre- and post-irrigation periods.

In the Harran Plain region, farmers typically plan for cereal cultivation in the autumn season, while they plan for cotton and corn cultivation as a secondary crop between spring and summer. The first planting stages of products grown in dry or irrigated conditions undergo fertilization. Therefore, we apply intensive fertilizer to the soils both during the autumn-winter pre-irrigation period and the spring-summer irrigation period. In this scenario, nitrate leaching from the soils could potentially occur throughout the year. The graph displays very close nitrate values before and after irrigation at all sampling points in the Harran Plain. According to the results of March, which is the pre-irrigation period, the nitrate concentration in the well in Uğurlu town was 642.78 mg/L, which is an alarming level. The nitrate concentration in this well in October was 582.22 mg/L. We observed that the nitrate levels in this case did not significantly change seasonally or before and after irrigation. The region's intensive agricultural activities for many years have led to nitrate pollution in groundwater. However, experts believe that nitrate pollution in the sampling points stems from a multi-year process that began during the irrigation period (1995) in the Harran Plain. It is assumed that the nitrate values in the wells increased by adding each year and reached today's values, especially as the groundwater levels increased over the years.

3.2. Investigation of nitrate pollution in batch reactor

Excessive use of artificial fertilizers, particularly in intensive agricultural practices, is the primary cause of nitrate levels exceeding the limit values in the groundwater in the study area. Especially in regions with semi-arid-arid climates, the high level of irrigation needs and unconscious irrigation techniques cause nitrate washing in soils and nitrate mixing into groundwater. Various traditional techniques such as adsorption, ion exchange, reverse osmosis, electrochemical, chemical, and biological methods have been developed to remove nitrate from water. However, these have various limitations such as post-treatment re-treatment, less efficiency and high installation costs. Among conventional techniques, the electrocoagulation process is an effective technology for nitrate removal. This is because nitrate anions prefer to stick to the surfaces of metal-hydroxide precipitates that are growing, and the higher the current density, the higher the operational cell potential, which improves the removal efficiency [22].

This study used electrocoagulation, a method that is both effective and easy to apply, for nitrate removal studies. Lacasa et al. [3] carried out nitrate removal studies with EC using Al and Fe electrodes from groundwater. They concluded that EC is an effective process for nitrate removal. Majlesi et al. [24] obtained nitrate removal efficiency of around 96% under optimum time and pH conditions using Al electrodes. The study was based on four different points (Uğurlu, Doruç, Bolatlar, Kısas) where nitrate concentrations were above the limit values as a result of the analyses. EC studies for nitrate removal were carried out on natural groundwater samples taken from four different sampling points in the Harran Plain region of Sanliurfa province.

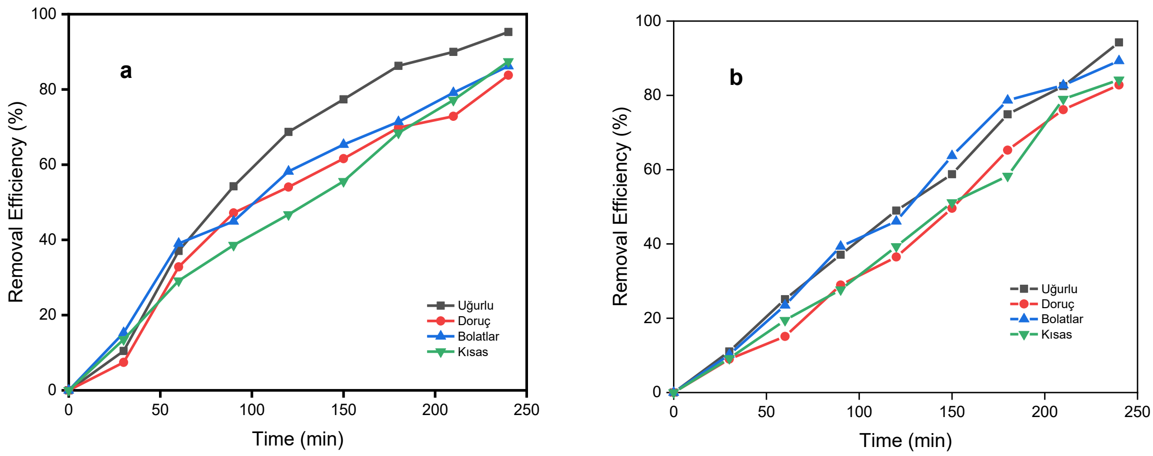

Figure 4. Variations of nitrate removal efficiencies for **(a)** pre-irrigation **(b)** post-irrigation periods during the EC process.

The EC process successfully reduced the nitrate concentration below the maximum pollutant level of 50 mg/L in the pre- and post-irrigation period. Nitrate removal efficiencies for Uğurlu, Doruç, Bolatlar and Kısas in the pre-irrigation period were 95.27%, 83.79%, 86.23% and 87.44%, respectively. In the post-irrigation period, nitrate removal efficiencies for Uğurlu, Doruç, Bolatlar and Kısas are 94.25%, 82.79%, 89.27%, 84.17%, respectively (Figure 4). EC operating conditions and energy consumption are shown in Table 1. According to the results, energy consumption increased as the conductivity decreased. The lowest energy consumption value (0.81 kWh/m^3) corresponded to the highest conductivity (2239 µS/cm).

Table 1. EC operating terms and conditions.

	Electrocoagulation Process							
	Pre-irrigation				Post-irrigation			
Parameter	Uğurlu	Doruç	Bolatlar	Kısas	Uğurlu	Doruç	Bolatlar	Kısas
Electrode material	SS	SS	SS	SS	SS	SS	SS	SS
Current density (mA/cm^2)	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Temperature (ºC)	18.1	17.8	20.4	18.8	23.1	22.2	24.8	24.6
Initial NO_3^- concentration (mg/L)	642.78	134.76	184.21	126.81	582.22	81.17	115.73	92.91
Final NO_3^- concentration (mg/L)	30.37	21.84	25.36	15.92	33.47	82.79	12.41	14.70
Initial pH	7.31	7.53	7.74	8.33	7.44	8.13	7.32	7.44
Final pH	7.14	7.38	7.45	8.13	7.17	7.76	7.12	7.29
Initial electrical conductivity (µS/cm)	2094	1982	1488	1253	2165	2239	1456	1136
Final electrical conductivity (µS/cm)	1387	1174	768	528	1247	1334	702	486
Operating time (min)	240	240	240	240	240	240	240	240
Nitrate removal (%)	95.27	83.79	86.23	87.44	94.25	82.79	89.27	84.17
Energy efficiency (kWh/m^3)	0.89	0.99	1.02	1.05	0.83	0.81	1.11	1.15

The pH of a solution is one of the most important parameters in the EC process [25]. Especially in this study, pH affected nitrate removal and the performance of the electrochemical process. Throughout the experimental study, water pH was difficult to control due to the instability of the EC process. As shown in Figure 5, both pH and conductivity were measured at certain time intervals. Both decreased as the nitrate removal rate increased. At the end of the study period, pH and conductivity values decreased (Figure 6).

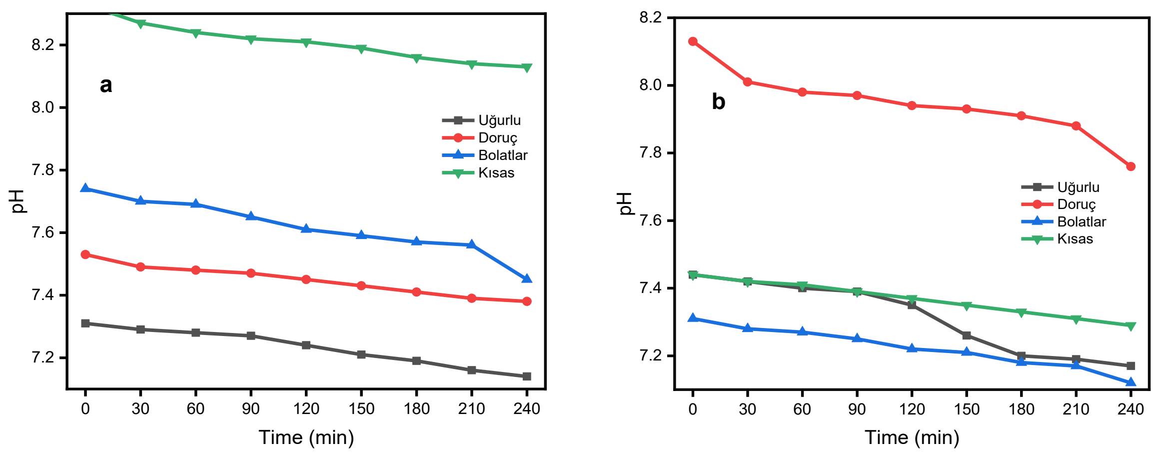

Figure 5. Variation of pH values for **(a)** pre-irrigation **(b)** post-irrigation periods during the EC process.

Operational parameters, including pH and current density, are crucial for optimizing the electrocoagulation process. Research has shown that maintaining a slightly acidic to neutral pH can improve nitrate reduction rates, as the presence of hydrogen ions can facilitate the electrochemical reactions involved in nitrate reduction [18]. Additionally, higher current densities have been associated with increased removal efficiencies, although they may also lead to higher energy consumption and potential electrode degradation [1,26]. Therefore, a balance must be struck between efficiency and operational costs.

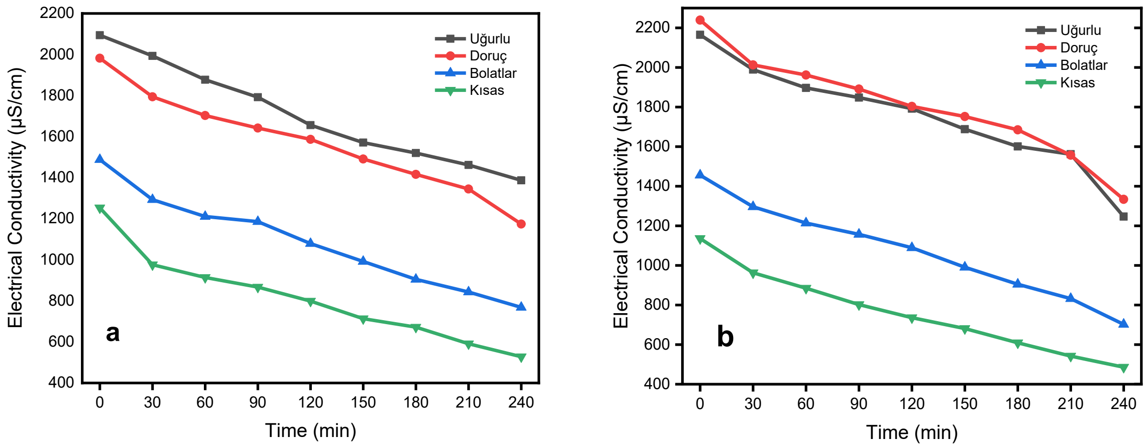

Figure 6. Variation of conductivity values for **(a)** pre-irrigation **(b)** post-irrigation periods during the EC process.

Furthermore, the economic feasibility of electrocoagulation using stainless steel electrodes is supported by its relatively low operational costs compared to traditional methods. The longevity of stainless steel electrodes reduces the frequency of replacements, and the energy consumption can be optimized through careful management of the operational parameters [27]. Studies have indicated that the use of stainless steel in electrocoagulation systems can lead to cost-effective solutions for treating nitrate-laden groundwater, making it an attractive option for municipalities and industries alike [8].

One of the important economic parameters in the EC process is electrical energy consumption. Electric energy consumption is a critical factor in various industrial processes, particularly in wastewater treatment technologies such as electrocoagulation. The energy consumption in electrocoagulation is primarily determined by the current density applied during the treatment process. Higher current densities can enhance the removal efficiency of contaminants but also lead to increased energy costs. For instance, studies have shown that optimizing current density can lead to significant reductions in energy consumption while maintaining effective pollutant removal [28,29]. Specifically, a current density of around 20 mA/cm² has been identified as effective for various applications, balancing energy use and treatment efficiency. The study reports energy consumptions of 0.143 kWh/kg of suspended solids (SS) for aluminum electrodes and 0.0571 kWh/kg SS for iron electrodes under optimized conditions [30]. This information is crucial for understanding the operational costs and energy efficiency of electrocoagulation as a drinking water treatment method.

4. Conclusion

The reactor established within the scope of the present study was operated as a batch. Samples were taken from 10 different sampling points and only 4 points (Uğurlu, Doruç, Bolatlar and Kısas) showed nitrate concentrations above the limit values. Therefore, EC studies were carried out using water samples from only these 4 sampling points. The study was carried out using SS electrodes. The SS values measured at the end of the EC process were < 0.4. In other words, the SS value passing into the water is very low and has no adverse effect on human health. In the pre-irrigation period, nitrate removal efficiencies for Uğurlu, Doruç, Bolatlar and Kısas were 95.27%, 83.79%, 86.23%, 87.44%, respectively.

In the post-irrigation period, nitrate removal efficiencies for Uğurlu, Doruç, Bolatlar and Kısas were 94.25%, 82.79%, 89.27%, 84.17%, respectively. According to the results, energy consumption increased as conductivity decreased. In this study, a cost of $1.06 per m^3 was calculated for SS electrode nitrate removal using current electricity costs for groundwater at 4 points taken from Harran Plain. It was observed that the SS-SS electrode combination used in the studies provided a good efficiency. The system can be developed and designed on a pilot scale in accordance with the needs of the region and on-site treatment can be carried out. It is thought that this method can also be applied to regions with the same characteristics as the arid/semi-arid study region. The laboratory-scale removal process that has been designed and implemented has created the foundation for future pilot-scale applications and portable EC system applications.

Ethical Statement

The author declares that this document does not require ethics committee approval or any special permission. This study does not cause any harm to the environment.

Conflict of Interest

The author declares no conflict of interest.

Author Contribution

B.Y.K. conducted the fieldworks and collected all data and wrote the manuscript.

Generative AI statement

The author declares that no Gen AI was used in the creation of this manuscript.

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