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Electrocoagulation as an efficient technology for nitrate removal from water

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ABSTRACT

High levels of nitrate in groundwater used for drinking are of major concern due to their adverse impacts on human health. According to the World Health Organization "WHO", nitrate concentration in drinking water should not exceed 15 and 50 mg-NO_3/L for infants and adults respectively. These limits were set to protect against the blue-baby syndrome, abortion, and stomach cancer. The main purpose of this study was to investigate the efficiency and the optimum operation parameters of the electrocoagulation (EC) process for nitrate removal from water. A laboratory continuous flow EC reactor was used to investigate the effects of different parameters on the nitrate removal from water. Influence of process parameters such as initial nitrate concentration (C_i), hydraulic retention time (HRT), distance between electrodes (D), applied voltage (V) and effective area of anode (A_{eff}) were investigated. It was found that the highest nitrate removal efficiency (91.6%) was achieved at the following conditions: C_i = 200 mg-NO_3/L, V = 40 V, HRT = 3.5 h, D = 3 cm, and A_{eff} = 61.4 cm^2 when the parameters: initial total dissolved solid (TDS_i), pH, and T_i were kept constant at 1,000 mg/L, 7 ± 0.1, and 25°C ± 2°C, respectively. The effectiveness of nitrate removal from groundwater in the Northern Gaza Governorate using EC showed that nitrate removal efficiency was 80% at the optimal conditions: V = 40 V, HRT = 2.5 h, D = 3 cm and A_{eff} = 61.4 cm^2, when TDS_i and C_i were 1,000 mg/L and 200 mg-NO_3/L, respectively. The study concluded that it was possible to reduce nitrate from groundwater to less than 50 mg-NO_3^-/L using EC. The results of this research indicated that the EC process is an efficient alternative technique for nitrate removal from groundwater.

Keywords: Nitrate; Electrocoagulation; Groundwater; Electrolytic oxidation; Removal efficiency

1. Introduction

Nitrate (NO_3^-) and nitrite (NO_2^-) are naturally occurring inorganic ions, which are part of the nitrogen (N) cycle. Although being stable and chemically unreactive, nitrate can be reduced by microbial action [1]. Microbial action decomposes wastes containing organic nitrogen first into ammonia, which is then oxidized to NO_2^- and NO_3^- [2]. Nitrate pollution of surface and groundwater has become a major problem in areas with agricultural activity nearby where there is excessive use of fertilizers [3]. Other sources of nitrate are animal waste, sewer lines, land discharges from wastewater, and atmospheric deposition [1,4,5]. Degradation of urine containing urea ($(NH_2)_2CO$) results in ammonia formation which can be oxidized to nitrite and nitrate by nitrification. At the same time, nitrite and nitrate can be converted to nitrogen gas and nitrogen monoxide by denitrification [6]. Different sources of the combined organic or ammonia nitrogen can be considered as the main source of nitrate, as most nitrogenous

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compounds in water tend to be transformed with certain means to nitrate [7].

High levels of NO_3^- and NO_2^- can cause dangerous illness due to acute exposure, while high concentrations of NO_3^- in drinking water cause both environmental and health concerns due to its toxicity. Methemoglobinemia or known as blue-baby syndrome attacks infants that are bottle fed with formula prepared with drinking water containing high levels of nitrate [8–10]. It results in breathing difficulty and turning the skin into blue due to the absence of oxygen or sometimes leads to death [8,9]. In drinking water, nitrate may cause different types of cancer in humans who are exposed to high amounts [1]. Other studies indicated that water contaminated with nitrate has been related to outbreaks of infectious diseases, childhood diabetes and decreased iodine uptake [6].

The World Health Organization (WHO) states that the concentration of nitrate in drinking water for adults should be <50 mg/L [8,9,11]. WHO limitation for infants is 15 mg/L [12]. Other international standards of nitrate concentration in drinking water can be summarized as follows: Canada, 45 mg/L [13]; Ontario, 10 mg/L [14]; Australia, 50 mg/L [15]; Malaysia, 50 mg/L [16]; USEPA, 10 mg/L [17].

Electrocoagulation (EC) is a popular water and wastewater treatment that has been studied extensively and widely applied as a wastewater treatment technology [18–20]. It is an evolving technology characterized by its versatility, ease of setup, simplicity of equipment used, low footprint, eco-friendly nature, less production of greenhouse gases, ability to provide active cations required for coagulation without increasing the salinity of the water and capability of removing many pollutants. EC can be applied effectively for treating various types of wastewaters, such as metal plating wastewater, baker's yeast wastewater, and municipal wastewater [21–23]. EC includes a high degree of automation [24] and doesn't require additional chemicals hence no neutralizing excess chemicals are needed [25–27]. Moreover, sludge formed by EC tends to be readily settable and easy to dewater. EC produced effluent with less total dissolved solid (TDS) content compared with chemical treatments and can remove the smallest colloidal particles. Other advantages of EC are electrically controlling the electrolytic processes with non-moving parts, thus requiring less maintenance, and it can be conveniently used in rural areas where electricity is unavailable hence a solar panel attached to the unit can sufficiently run the system. However, EC has some disadvantages: the sacrificial electrodes need regular replacement since they are dissolved in wastewater streams due to oxidation, using electricity may be expensive in many places; an impermeable oxide film may be formed on the cathode leading to efficiency loss of the EC unit, high conductivity of the wastewater suspension is required, and gelatinous hydroxide may tend to solubilize in some cases [25]. Despite the reduced production of sludge in EC [28], one inherent disadvantage is that the sludge needs further treatment and correct disposal [29].

A study to remove cephalexin antibiotics from water using aluminum-based EC unit was performed by Arab et al. [30]. Computational and statistical models were used to optimize the effects of key parameters on the electrochemical removal of cephalexin, including the initial cephalexin concentration (15–55 mg/L), initial pH (3–11), electrolysis time (20–40 min), and electrode type. The response surface methodology-central composite design (RSM-CCD) was used to investigate the dependency of the studied variables, while the artificial neural network (ANN) and adaptive neuro-fuzzy inference system (ANFIS) methods were applied for predicting the experimental training data. The results showed that the best experimental and predicted removals of cephalexin (CEX) were 88.21% and 93.87%, respectively, which were obtained at a pH of 6.14 and electrolysis time of 34.26 min [30]. A research aimed to develop EC unit design using drilled electrodes to mix the solution being treated without using external mixers – to minimize power consumption – was conducted by Abdulhadi et al. [31]. The performance of the EC unit was validated by applying it to remove iron from water taking into account the effects of applied current density (ACD), pH of water (PoW), iron concentration (IC) and treatment time (TT). The effects of these parameters were optimized using the Box–Behnken model. Synthetic water samples containing different concentrations of iron (10–30 mg/L) were treated in a continuous flow, using the EC reactor at different ACD (1.5–4.5 mA/cm^2), PoW (4–10) and TT (10–50 min). The results revealed that the removal of 99.9% of iron was achieved by keeping PoW, ACD, IC and TT at 7, 3 mA/cm^2, 10 mg/L and 50 min, respectively. The effects of ACD, PoW, IC and TT on iron removal could be successfully simulated with $R^2 = 0.9788$ [31]. A study conducted by Hashim et al. [32] combined electromagnetic radiation (microwave) and electrocoagulation technologies to purify water from OM-heavy metal complexes created using iron and ethylenediaminetetraacetic acid (EDTA). The organic matter-iron solution was introduced to a microwave field to break down the complex, followed by electrolysis of the solution using an aluminum-base EC cell. Microwave power (50–300 W), temperature (50°C–150°C) and irradiation period (5–15 min) were measured. During the electrolyzing stage, initial pH (4–8), current density (1–2 mA/cm^2) and space between electrodes (5–20 mm), were examined. The results showed that the electromagnetic radiation-electrocoagulation technology removed up to 92% of the organic matter-iron complex in comparison to 69.6% removal using a traditional electrocoagulation method. The best operational conditions were established as follows: 10 min of microwave irradiation at 100 W and temperature of 100°C, followed by 20 min of electrolyzing at an initial pH of 6, the space between electrodes 5 mm and current density of 1.5 mA/cm^2 [32].

The removal of nitrate from tap water by electrocoagulation–flotation (ECF) process was investigated by Moradi and Ashrafizadeh [33]. In their study, an ECF reactor made of Plexiglass with a working volume of 8 L was used. The effect of different arrangements of anode/cathode electrodes (Al/Fe, Fe/Al, and SS/Fe) and the effects of operating parameters were evaluated. The results revealed that at the optimum conditions, Al/Fe electrodes, up to 93% of the nitrate was removed.

Apshankar and Goel [34], determined optimum operating conditions for maximizing nitrate removal from drinking water using EC followed by settling and filtration.

Batch experiments were carried out using iron electrodes and four types of water were tested. In 22 DC power experiments with all waters tested, maximum removal efficiencies were 37% (after EC) and 38.21% (after EC and settling). Majlesi et al. [35], investigated nitrate removal from water by the ECF in continuous operation under different conditions. The results indicated that the maximum nitrate removal was achieved by Al/Al electrode arrangement. Increasing the current density from 0.4 to 3.2 mA/cm^2 increased the nitrate removal efficiency from 55 to 96% under optimum conditions of time and pH. Continuous operation of the ECF reactor increased nitrate removal from 37% at a detention time of 10 min to 96% at a detention time of 30 min. Dehghani et al. [36], determined nitrate removal efficiency from water by EC using Al/Fe electrodes. Nitrate removal was determined at pH levels of 3, 7, and 11, different voltages (15, 20, and 30 V), and operation times (30, 60, and 75 min). Results showed that the removal efficiency increased from 27% to 86% as pH increased from 3 to 11 at the optimal condition of 30 V and 75 min operation time. Moreover, increasing the reaction time from 30 to 75 min (at 30 V and pH = 11) increases the removal efficiency from 63% to 86% respectively. Hooshyar and Abbas [37], studied nitrate removal from an aqueous solution by EC using aluminum/graphite electrodes. The results showed that by applying an electric current of 0.14 Å for 120 min, the nitrate content would be reduced to 97% [37]. Malakootian et al. [38], studied the removal efficiency of nitrate from aqueous solutions using EC. In the experiment, the concentrations of nitrate ranged from 100 to 200 mg/L and the experimental set-up was a batch reactor. The results showed that EC can result in nitrate levels lower than the standard limit. Moreover, pH, electrical potential difference, TDS and number of electrodes have direct effects on nitrate removal, while initial nitrate concentration has a reverse effect. An experiment was conducted by Kumar and Goel [27], to evaluate some factors influencing arsenic and nitrate removal from drinking water in a continuous flow EC process. A bench-scale simulation of drinking water treatment was performed by adding a filtration column after a rectangular EC reactor. The contaminant removal efficiency was determined for applied voltages ranging from 10 to 25 V and a comparative study was done with distilled water and tap water for nitrate and arsenic (*V*). The maximum removal efficiency was 84% for nitrate at 25 V and 75% for arsenic (*V*) at 20 V. Emamjomeh and Sivakumar [28], investigated the effects of electrolysis time, electrolyte pH, initial nitrate concentration, and current rate on nitrate removal efficiency. The optimum nitrate removal was observed in the pH range 9–11. It appeared that the nitrate removal rate was 93% when the initial nitrate concentration and electrolysis time were 100 mg-NO_3/L and 40 min respectively. Koparal and Öğütveren [39], investigated the feasibility of nitrate removal from water by electroreduction (ER) and EC. In the ER, removal of nitrate to an allowable concentration has been accomplished in the pH range of 5–7 with an energy consumption value of 1×10^{-3} kWh/g. In EC, an allowable concentration of nitrate has been achieved in the pH range of 9–11 with energy consumption value of 0.5×10^{-4} kWh/g. The full removal of nitrate was also possible, but with higher energy consumption for the two methods [39].

This research investigated the efficiency and the optimum operating parameters of the EC process for nitrate removal from water using a continuous flow reactor. The specific objectives were: (1) to investigate the combined effects of applied voltage (V), initial nitrate concentration (C_i), distance between electrodes (D), effective area of anode (A_{eff}) and hydraulic retention time (HRT) on the efficiency of EC for nitrate removal from aqueous solutions using continuous flow reactor, (2) to study the effect of EC on the other chemicals existing in the groundwater, of the northern area – Gaza Strip, during nitrate removal and the possible production of other chemicals during the process, that is, the matrix effect.

2. Theory of EC

EC processes a direct current source between metal electrodes immersed in water. The electrical current destabilizes the charges of the pollutants and causes the dissolution of metal electrodes, commonly iron and aluminum, into water. The dissolved metal ions, at an appropriate pH, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants that can be separated from the electrolytic mixture [40,41]. Simultaneous evolution of hydrogen gas at the cathode also helps pollutants removal by flotation and generation of precipitates and flocs capable of removing heavy metal ions. A range of coagulant species and hydroxides are formed to destabilize and coagulate the suspended particles and to adsorb dissolved contaminants [42,43]. EC is based on the in-situ formation of coagulants as the sacrificial anode corrodes due to the applied current as shown in Fig. 1.

Three successive stages occur during EC: (1) formation of coagulants by electrolytic oxidation of the sacrificial anode, (2) destabilization of the pollutants, particulate suspension and breaking of emulsions, and (3) aggregation of the destabilized phases to form flocs. The pollutants can be in the form of large particles easy to separate from water by settling, colloids and dissolved mineral salt and organic molecules [44].

3. Mechanisms of EC

The mechanisms of EC for water and wastewater treatment are very complex. There are three other possible mechanisms involved besides EC, namely, electroflotation, electrochemical oxidation and adsorption [45]. During EC, an electric potential difference is applied between a soluble Fe anode and a cathode, then ferrous ions form at the anode followed by in situ oxidation to the ferric state and hydroxyl ions are then generated at the cathode. Then, hydroxides precipitate with ferric ions as ferric hydroxide, which are responsible for the effects observed during particle aggregation [46–48].

The main electrochemical reactions at the electrodes during the EC process are:

At the cathode, the metal (M) may be chemically attacked by OH^- especially at high pH values and H_2 gas is liberated [40]:

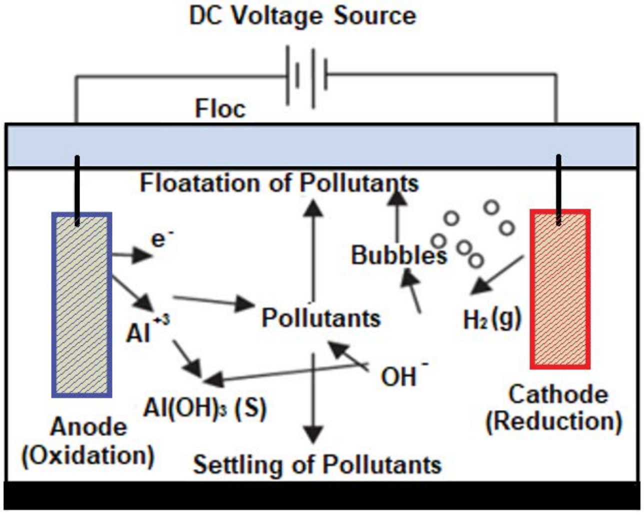

Fig. 1. Schematic diagram of two electrode EC cell.

$$3H_2O + 2e^- \rightarrow \left(\frac{3}{2}\right)H_2 + 3OH^- \tag{1}$$

$$2M + 6H_2O + OH^- \rightarrow 3H_2 + 2M\left(OH\right)_4 \tag{2}$$

At the anode, sacrificial metal (M), Al or Fe, is dissolved:

$$M \rightarrow 3e^- + M^{+3} \tag{3}$$

In the case of Fe electrode, the anodic reaction also occurs:

$$\text{Fe} \rightarrow 2e^- + M^{+2} \tag{4}$$

4. Materials and methods

4.1. Apparatus

The apparatus used in the EC experiments include: DC power supply (a laboratory digital DC power supply – type: GWINSTEK GPS 3303 – with a voltage range of 0–30 V and current range of 0–3 Å), 2 iron plate electrodes 13 cm × 3 cm × 0.3 cm, water pump (peristaltic pump), magnetic stirrer, flexible PVC pipe, stop watch, CT-2600 Spectrophotometer, pH meter (pH/ORP/ISE Graphic LCD pH Bench top Meter, HANNA instruments), turbidity meter (HI 93703, portable microprocessor turbidity meter, HANNA instruments), 20 L tank 20 containing synthetic polluted water, glass ware (pipettes, beakers, volumetric flasks and others), pH adjustment (HCl 1 mol/L and NaOH 1 mol/L), electronic balance, multi-meter (type: GWINSTEK GDM-8135), and EC cell (a continuous flow EC reactor was fabricated in the lab from Perspex sheet with dimensions of 24 cm length × 8 cm width × 12 cm height and a length to width ratio of 3). The experiments were conducted according to the Standard methods for the Examination of Water and wastewater, method No. 4500 – NO_3 B [49]. Figs. 2 and 3 give schematic and photographic presentations of the experimental setup used in this work, respectively.

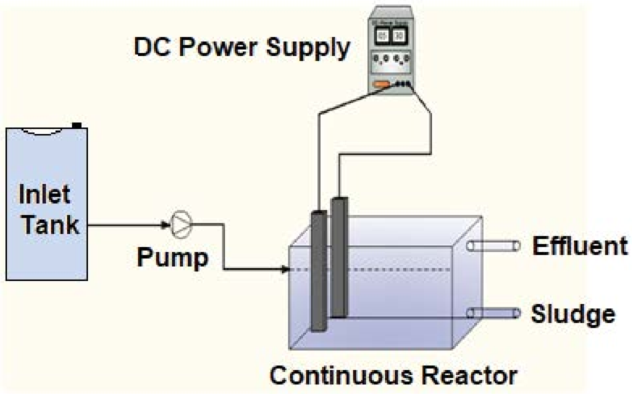

Fig. 2. Schematic illustration of the experimental setup for the EC reactor.

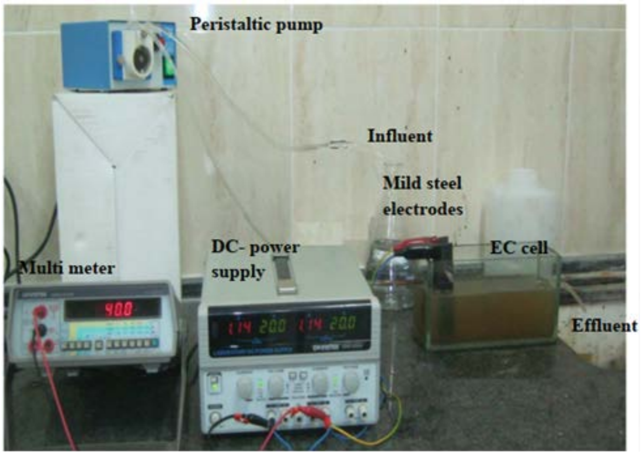

Fig. 3. Photographic presentation of the EC system.

4.2. Chemicals used

Chemicals used are potassium nitrate (KNO_3), purity of 99.5% by weight (produced by HiMedia Laboratories Pvt. Ltd., India), sodium chloride (NaCl), purity of 99.5% by weight (produced by HiMedia Laboratories Pvt. Ltd., India), sodium hydroxide (NaOH) pallets extra Pure, purity of 97.5% by weight (product of loba chemie, India), hydrochloric acid (HCl) of 1 M and 0.1 M, sodium hydroxide (NaOH) of 1 M and 0.1 M, buffer solutions with pH = 4, 7, and 11, conductivity standard – 1,412 µmhos/cm, and distilled water.

4.3. Water samples used

Two types of water samples were used in this work. The first group of samples were taken from a synthetic aqueous nitrate solution prepared in the laboratory and the second group were taken from a groundwater well in the north of Gaza Strip.

4.3.1. Synthetic aqueous nitrate solution water samples

A stock solution of 1,000 ppm nitrate was prepared by dissolving potassium nitrate (KNO_3) in distilled water.

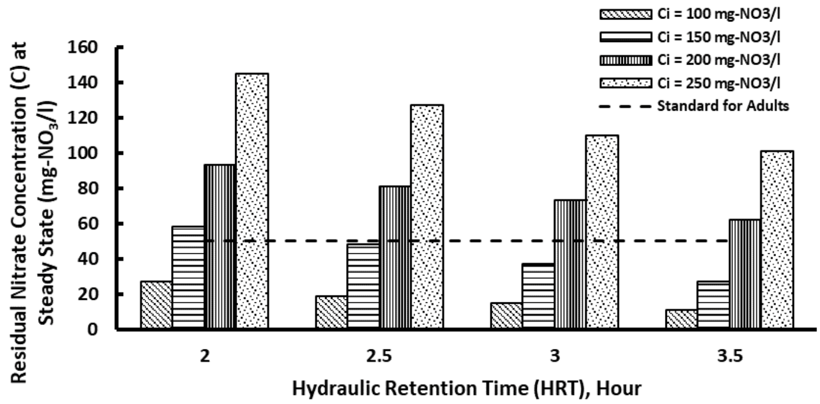

Fig. 4. HRT effect on residual NO_3^- concentration (C).

The amount of KNO_3 required to prepare the stock solution was calculated as [50]:

$$W = VC_{st}\left(\frac{M_{wt}}{At_{wt}}\right) \quad (5)$$

where W = weight of KNO_3 (g); V = volume of solution (L); C_{st} = nitrate concentrations in stock solution (g/L); M_{wt} = molecular weight of KNO_3 (101.1032 g/mole); At_{wt} = atomic weight of nitrate (62.0049 g/mole).

Based on Eq. (5), nitrate stock solution of 1,000 ppm was prepared by dissolving 1.639 g of potassium nitrate in 1 L of distilled water. A certain amount of the stock solution was mixed with distilled water to prepare the standard solution of each experiment to reach the required concentration.

4.3.2. Groundwater samples

The groundwater sample was taken from a water well located in the north of Gaza Strip, Palestine. Table 1 presents the main characteristic of this sample. As shown in the table, the initial nitrate concentration is 200 mg-NO_3/L, which is higher than the standard level specified by WHO for drinking water for adults (50 mg-NO_3/L).

4.4. Analytical methods

4.4.1. Measurement of NO_3^- as N by spectrophotometer instrumentation

Nitrate concentration was determined using the "Ultraviolet Spectrophotometric Screening Method" according to the Standard methods for the Examination of Water and wastewater, method No. 4500-NO_3 B [49]. The spectrophotometer used in this wok was a CT-2600 model. The wavelength settings of the spectrophotometer were 220 nm to obtain nitrate reading and 275 nm to determine interference due to dissolved organic matter. Two times the absorbance at 275 nm were subtracted from the absorbance at 220 nm to get the correct value of absorbance due to nitrate and to consequently get the correct concentration of nitrate.

Table 1
Characteristics of the groundwater sample

Parameter	Value
Nitrate, mg-NO_3/L	200
Chloride, mg/L	201
TDS, mg/L	700
pH	7.23
Electrical conductivity, µs/cm	1,417
Turbidity, FTU	1.2
Total hardness, mg/L	600
Alkalinity, mg/L $CaCO_3$	215
Sodium, mg/L	73

4.4.2. Evaluation of removal efficiency

Each sample taken from the EC cell was filtered and then analyzed using an atomic absorption spectrophotometric model (CT-2600). The removal efficiency of nitrate from the synthetic polluted water treated by EC is calculated as follows:

$$R(\%) = \left(\frac{C_o - C}{C_o}\right) \times 100 \quad (6)$$

where $R(\%)$ = nitrate removal efficiency; C_o = nitrate concentration at initial (mg/L as NO_3); C = nitrate concentration at any time (mg/L as NO_3) [51].

4.5. Experimental program

The experimental program throughout the work focused on treating the prepared aqueous solution as well as treating a groundwater sample. The operational conditions of the experimental program are presented in the following sections.

4.5.1. Treatment of aqueous solution

Some parameters were kept constant throughout the experimental work including: the initial total dissolved

Table 2
Summary of operating parameters for test runs

Test run no.	Parameters' effect to be measured	Voltage (V)	Initial nitrate concentration (mg-NO_3/L)	Hydraulic retention time (HRT) (h)	Distance between electrodes (D) (cm)	Effective area (A_{eff}) (cm^2)
1	Hydraulic retention time	40	100, 150, 200, 250	2, 2.5, 3, 3.5	3	47.1
2	Applied voltage	35, 40, 45, 50	200	2, 2.5, 3	3	47.1
3	Initial nitrate concentration	40	100, 150, 200, 250	2, 2.5, 3, 3.5	3	47.1
4	Distance between electrodes	40	200	2, 2.5, 3	2, 3, 4, 5	47.1
5	Effective anode area	40	200	2, 2.5, 3, 3.5	3	47.1, 54, 61.4

solids "TDS_i" was kept at 1,000 mg/L, the pH was kept at 7 ± 0.1, and initial temperature "T_i" was kept at 25°C ± 2°C. The operating conditions for each test run are summarized in Table 2.

4.5.2. Treatment of groundwater

A groundwater sample with nitrate and TDS_i concentrations of 200 mg-NO_3/L and 700 mg-TDS/L, respectively, was used in the experiments. The conductivity of the solution was increased by adding electrolyte [100–300 mg of NaCl in 1 L of the groundwater sample]. The best operating conditions were determined from the result of the aqueous solution. After that, the best conditions were tested on groundwater samples from a well located in the north of Gaza Strip.

4.5.3. Experimental work

During the experiments, as shown in the experimental program, there was a need to vary some parameters, such as the HRT, the voltage (V), the initial nitrate concentration (C_i), the distance between electrodes, and the effective area of the anode. The hydraulic retention time (HRT) was varied based on the following equation:

$$\text{HRT} = \frac{V_r}{Q} \tag{7}$$

where V_r is the reactor volume (m^3) and Q is the flow rate (m^3/h) [52].

In this work, V_r was kept constant; 1,500 mL, while the flow rate was changed as 750, 600, 500, and 428 mL/h to vary the HRT to 2,2.5,3, and 3.5 h, respectively. The applied voltage (V) was varied to 35, 40, 45, and 50 V using a transformer that converts AC to DC. The current was measured using a multi-meter during each experimental run. The initial nitrate concentration was varied to 100, 150, 200, and 250 mg-NO_3/L by adding appropriate mass of potassium nitrate KNO_3.

Each experimental run was started by switching on the DC power supply. During the experiments, anodic dissolution occurred, and hydrogen gas was produced at the cathode. The turbidity and pH were monitored during each experiment in the influent and effluent. Effluent samples of 30 mL were taken at different times during the experiment, filtered, and then analyzed. The experiment continued until reaching steady-state concentrations. Generally, each experimental run lasted 4 h. All effluent samples were analyzed for pH, turbidity, temperature, current rate, total dissolved solids (TDS) and nitrate. After each run, the electrodes were washed, brushed and cleaned by ethanol to remove any accumulating solids on their surfaces.

5. Results and discussion

Nitrate removal efficiency was investigated by studying many parameters affecting the EC process. These parameters include the hydraulic retention time (HRT), the applied voltage (V), the initial nitrate concentration (C_i) in the solution, the distance between electrodes (D), and the effective area of the anode (A_{eff}). The following sections illustrate the effect of each parameter on nitrate removal using the EC process.

5.1. Effect of HRT on nitrate removal efficiency

The effect of the HRT on nitrate removal was examined by setting the initial nitrate concentration (C_i) at fixed values (100, 150, 200, and 250 mg-NO_3/L) while changing the HRT to values of 2, 2.5, 3, and 3.5 h in each experimental run (4 experimental runs for each C_i concentration). Fig. 4 illustrates the effect of HRT on the steady state residual nitrate concentration (C). During the experiments, the voltage (V), the distance between the electrodes (D), the initial total dissolved solids (TDS_i), the temperature (T_i), the pH, and the effective area of the anode (A_{eff}) were kept constant at 40 V, 3 cm, 1,000 mg-TDS/L, 25°C ± 2°C, 7, and 47.1 cm^2, respectively. As observed from Fig. 4, at low C_i, the increase in HRT resulted in a lower steady state nitrate residual concentration (C) (higher removal efficiency). For example, at a C_i of 100 mg-NO_3/L, when the HRT was 2 h, the final nitrate concentration was 27.8 mg-NO_3/L (R = 72%), while when the HRT was 3.5 h, the final nitrate concentration decreased to 13.14 mg-NO_3/L, and the removal efficiency increased to 86.9%. The same trend was observed for the other initial nitrate concentrations. The removal efficiency decreases with increasing the solution flow rate. This reduction in the efficiency is attributed to the decrease in HRT in the EC cell as the flow rate increases. With reference to Fig. 4, it can be interpreted that the standard concentration limit of nitrate in drinking water for adults (50 mg-NO_3/L) can be reached at HRTs of 2 h and 2.5 h for initial nitrate concentrations (C_i) of 100 and 150 mg-NO_3/L, respectively. However, for the C_i concentrations of 200 and 250 mg-NO_3/L, the HRT should be >3.5 h to achieve the mentioned standard concentration limit.

5.2. Effect of applied voltage

In electrochemical processes, the applied voltage is a very important parameter for controlling the reaction rate within the electrochemical reactor [25]. This variable determines the production rate of the coagulant, adjusts bubble production, and hence affects the growth of the formed flocs [53].

To investigate the effect of the applied voltage (V), the HRT was set at fixed values (2, 2.5, and 3 h) while changing the voltage (V) to values of 35, 40, 45, and 50 V (4 experimental runs for each HRT value). During the experiments, D, C_i, pH, TDS_i, T_i, and A_{eff}, were kept constant at 3 cm, 200 mg-NO_3/L, 7, 1,000 mg-TDS/L, 25°C ± 2°C, 47.1 cm^2, respectively. Fig. 5 illustrates the effect of the applied voltage (V) on the steady state residual nitrate concentration (C). It was observed that when the applied voltage (V) is increased, the removal percentage (R) also increased, and the steady state residual nitrate concentration (C) decreased. For example, at HRT of 2 h, when V was 35 V, the final nitrate concentration was 124 mg-NO_3/L (R = 38%), while when V was increased to 50 V, the final nitrate concentration decreased to 53.3 mg-NO_3/L, and the removal percentage increased to 73.33%. The same trend was observed for the other HRTs. It was also observed from Fig. 5 that the standard limit of nitrate for adults (50 mg-NO_3/L) can be reached at HRTs of 2.5 and 3 h for applied voltage ≥45 and 48 V, respectively, while for HRT of 2 h the applied voltage should be >50 V. From the above observations, it was concluded that for a given HRT, increasing the applied voltage resulted in better nitrate removal efficiency.

5.3. Effect of initial nitrate concentration

The effect of the initial nitrate concentrations (C_i) was investigated by setting the HRT at fixed values (2, 2.5, 3, and 3.5 h) while changing C_i to the values of 100, 150, 200 and 250 mg-NO_3/L (4 experimental runs for each HRT value).

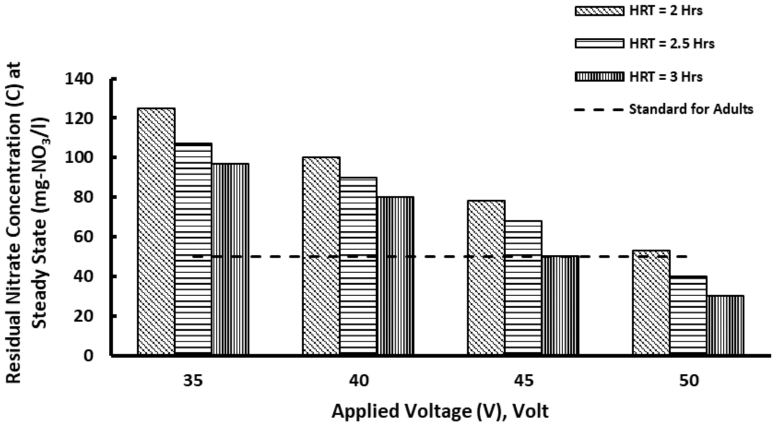

Fig. 5. Applied voltage effect on residual NO_3^- concentration (C).

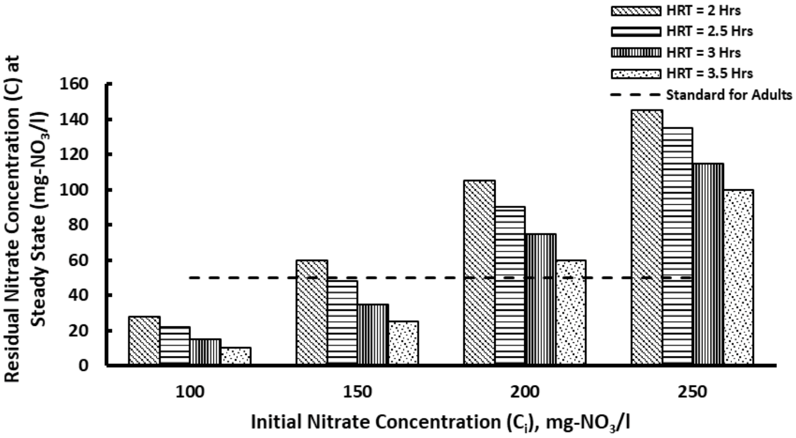

Fig. 6. Initial NO_3^- concentration effect on residual NO_3^- concentration.

During the experiments, V, D, pH, TDS_i, T_i and A_{eff}, were kept constant at 40 V, 3 cm, 7, 1,000 mg-TDS/L, 25°C ± 2°C, and 47.1 cm^2, respectively.

Fig. 6 illustrates the effect of initial nitrate concentration (C_i) on the steady state residual nitrate concentration (C). It was observed that when initial nitrate concentration (C_i) increased, the residual nitrate concentration (C) also increased, and the removal percentage (R) decreased. For example, at HRT of 2 h, when C_i was 100 mg-NO_3/L, the final nitrate concentration was 27.8 mg-NO_3/L (R = 72%). While when C_i was increased to 250 mg-NO_3/L, the final nitrate concentration increased to 145.5 mg-NO_3/L, and removal percentage decreased to 41.8%. The same trend was observed for the other HRTs. From Fig. 6 it can be observed that the standard concentration limit of nitrate for adults (50 mg-NO_3/L) can be reached at HRT 2, 2.5, 3 and 3.5 h for nitrate concentration that are ≤138, 153, 167 and 178 mg-NO_3/L, respectively.

In fact, the coagulation process sweeps and precipitates the NO_3^- ions by flocs formed. The NO_3^- ions were effectively removed from water owing to aggregation of the colloidal particles by means of Fe^{2+}/Fe^{3+} produced during electrolysis. These ions react with the colloids and form settable flocs that can be easily separated from solution. The decrease of nitrate removal efficiency when the initial nitrate concentration was increased is attributed to the lack of the formed iron oxide flocs in solution, more iron oxides were needed to decrease the dissolved nitrate concentrations. Nitrate removal is consequently limited by the production rate of iron oxides.

5.4. Effect of distance between electrodes

The effect of the distance between the electrodes (D) on nitrate removal was investigated by setting the HRT at a fixed value (2, 2.5, 3, and 3.5 h) while changing D to 100, 150, 200, and 250 mg-NO_3/L (4 experimental runs for each HRT value). During the experiments, V, C_i, pH, TDS_i, T_i and A_{eff}, were kept constant at 40 V, 200 mg-NO_3/L, 7, 1,000 mg-TDS/L, 25°C ± 2°C, and 47.1 cm^2, respectively.

Fig. 7 illustrates the effect of distance between electrodes (D) on the steady state residual nitrate concentration (C). It was observed that as the distance between electrodes (D) increased, the residual nitrate (C) concentration also increased, and the removal percentage (R) decreased. For example, at HRT of 2 h, and D of 2 cm, the final nitrate concentration was 71 mg-NO_3/L (R = 64.5%), while when D was increased to 5 cm, the final nitrate concentration increased to 132.75 mg-NO_3/L, and the removal percentage decreased to 33.6%; the same trend was observed for the other HRTs. For all HRT values, it was observed that C increased nonlinearly with a decreasing rate when the distance between the electrodes was increased.

With reference to Fig. 7, to achieve the standard nitrate concentration limit for adults (50 mg-NO_3/L) at the experimental conditions for HRT of 3 h, the distance between electrodes should be around 2 cm. While for other HRTs (2, 2.5 h) the distance between electrodes should be between 1 and 2 cm. The highest removal efficiency of 80% was obtained at a distance of 2 cm when the HRT was 3 h.

This is attributed to the decrease in the electric current resistance in the solution as the distance between electrodes decreases. As a result, the electric current in the solution increases and consequently the rate of iron dissolution and Fe^{2+} release increases leading to more nitrate removal from the solution.

The distance between electrodes plays a significant role in an EC operation, as it determines the electrostatic field between the anode and the cathode. The electrostatic field is the highest when the inter-electrode distance is at its minimum value. Therefore, the metal hydroxides, that aid in forming flocs to support coagulation, were degraded due to the strong collisions resulting from the high electrostatic attraction [54]. Consequently, the EC efficiency is low at minimum inter-electrode distance. On the other hand, larger inter-electrode gap defers the subsequent formation of metal hydroxide-flocs due to reduced electrostatic forces [55]. Electrode spacing beyond the optimum value largely reduces the EC efficiency, incurring higher power consumption to overcome the slower movement of the released ions between the anode and cathode [56,57]. Hence, it is vital to run EC at an optimum inter-electrode distance. Several studies had used a minimum distance

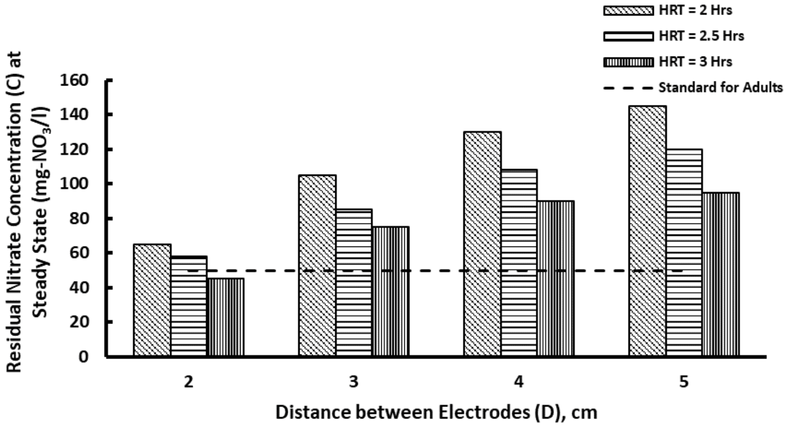

Fig. 7. Effect of distance between electrodes on residual NO_3^- concentration.

of no less than 10 mm [58–60]. Beside this, an inter-electrode distance of 20–30 mm had also been employed in many studies [61–64].

5.5. Effect of effective area of anode

Anode materials and their effective area have a significant effect on EC efficiency [24].

The effect of the effective area of anode (A_{eff}) on nitrate removal was investigated by setting the HRT at fixed values (2, 2.5, 3, and 3.5 h) while changing A_{eff} to 47.1, 54, and 61.4 cm². During the experiments, V, C_i, pH, TDS_i, T_i and D, were kept constant at 40 V, 200 mg-NO_3/L, 7, 1,000 mg-TDS/L, 25°C ± 2°C, and 3 cm, respectively. Fig. 8 illustrates the effect of anode effective area (A_{eff}) on the steady state residual nitrate concentration (C). From Fig. 8 it was noticed that when the effective area of the anode (A_{eff}) increased, the residual nitrate concentration (C) decreased and the removal percentage (R) increased. For example, at HRT of 2 h, when A_{eff} was 47.1 cm², the final nitrate concentration was 96.5 mg-NO_3/L (R = 51.8%), while as A_{eff} was increased to 61.4 cm, the final nitrate concentration decreased to 67 mg-NO_3/L, and the removal percentage decreased to 66.5%. The same trend was observed for all HRTs tested in this experiment. As observed from Fig. 8, the standard concentration limit of nitrate for adults (50 mg-NO_3/L) can be reached at HRTs of 2.5, 3 and 3.5 h for A_{eff} values that are ≥51, 54, and 58 cm², respectively. On the other hand, for HRT of 2 h, A_{eff} should be >61.4 cm², to achieve the standard nitrate limit.

The direct proportionality between A_{eff} and the nitrate removal efficiency as observed in this work is in line with the EC process principles. When A_{eff} is increased, the amount of oxidized iron and iron hydroxide flocs increase. The generated hydroxide flocs have a high adsorptive capacity rate that leads to the increase in the nitrate removal efficiency.

5.6. Treatment of groundwater sample

In order to confirm the results obtained from the experiments conducted on the synthetic aqueous solution, similar experiments were applied on a groundwater sample having the characteristics mentioned in Table 1. The experiments on this sample were conducted under the best operating conditions for nitrate removal using EC reactor that were determined in the experiments on the synthetic aqueous solution as presented earlier in this paper. These conditions are presented in Table 3. The temperature of the solution was kept at 25°C ± 2°C during the experiments.

Another important operating condition was the initial total dissolved solids (TDS_i) and its impact on the conductivity of the groundwater solution inside the EC reactor and consequently on nitrate removal efficiency (R). To examine the impact of TDS_i concentration on R, the experiments were conducted first on four TDS_i concentrations: the groundwater original TDS_i concentration of 700 mg-TDS/L, 800, 900 and 1,000 mg-TDS/L. The latter three concentrations of TDS_i were prepared by adding NaCl to the groundwater sample.

Fig. 9 indicates that the residual nitrate concentration (C) decreases with the increase of the initial total dissolved solid (TDS_i). The values of C were 96.50, 67.00, 48.43, and 41.57 mg-NO_3/L when the TDS_i was, 700, 800, 900, and 1,000 mg/L. For the same TDS_i concentrations, the removal efficiencies were 51.8%, 66.5%, 75.8%, and 79.2% respectively.

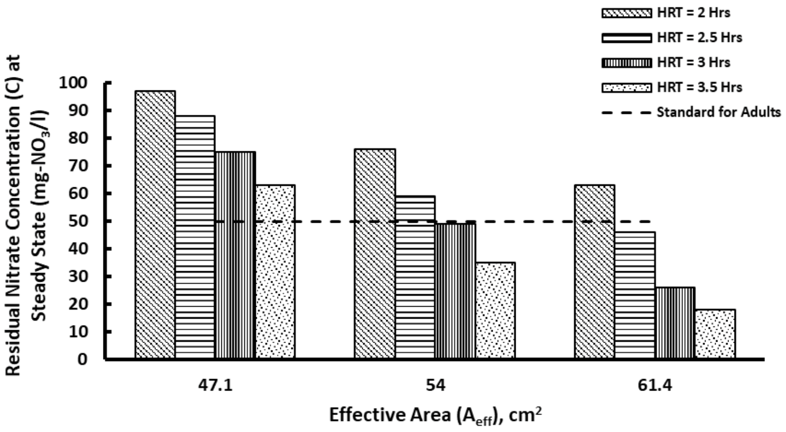

Fig. 8. Effect of effective area of anode (A_{eff}) on residual NO_3^- concentration.

Table 3
Best operating conditions for nitrate removal using EC

Initial concentration (mg NO_3^-/L)	Voltage (V)	Hydraulic retention time (Hr)	Distance between electrodes (cm)	Effective electrode area (cm²)
100	40	2	3	47.1
150	40	2.5	3	47.1
200	40	2.5	3	61.4

Table 4
Characteristic of groundwater sample after treatment

Parameter	Original sample	Adding 100 mg-NaCl/L	Adding 200 mg-NaCl/L	Adding 300 mg-NaCl/L
Initial TDS, mg/L	700	800	900	1,000
Nitrate residual, mg-NO_3/L	96.5	67	48.43	41.57
Chloride, mg/L	130	206	240	320
TDS, mg/L	505	596	656	892
pH	7.4	7.7	7.84	8.4
Electrical conductivity, µs/cm	790	1,150	1,292	1,550
Turbidity, FTU	4.3	4.45	4	3.5
Total hardness, mg/L	97	76	59.5	44
Alkalinity, mg/L $CaCO_3$	100	80	52	39
Sodium, mg/L	73	113	163	220

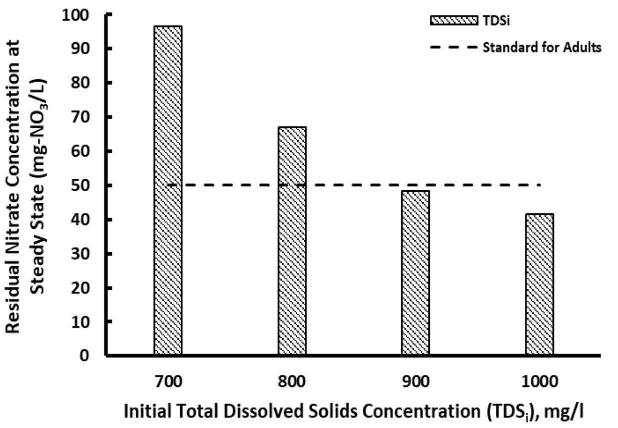

Fig. 9. Effect of TDS_i on residual NO_3^- concentration.

It is also observed from Fig. 9 that the standard nitrate concentration for adults (50 mg-NO_3/L) could be achieved at TDS_i of 900 and 1,000 mg/L. The characteristics of the groundwater after treatment is given in Table 4.

6. Conclusions

Continuous flow experiments were designed to investigate the effects of different parameters including applied voltage (V), initial nitrate concentration (C_i), hydraulic retention time (HRT), distance between electrodes (D), and effective area of anode (A_{eff}) on nitrate removal efficiency by EC reactor using synthetic aqueous solutions. Then the optimal operating conditions were determined and applied to groundwater samples obtained from a water well in the north of Gaza Strip. The main conclusions obtained from this work are:

- The EC process is successfully applied to remove nitrate from aqueous solution. Nitrate removal efficiency was dependent on the initial nitrate concentration, distance between electrodes, effective area of the anode and the applied voltage.
- The results showed that the applied voltage, the initial total dissolved solids, the effective area of anode and the hydraulic retention time are directly proportional to nitrate removal efficiency, while the initial nitrate concentration, and distance between electrodes are inversely proportional to nitrate removal efficiency.
- The best operating conditions for nitrate removal by EC under the conditions applied in this work are:
    - For C_i of 100 mg-NO_3/L: V = 40 V, HRT = 2 h, D = 3 cm, and A_{eff} = 47.1 cm^2.
    - For C_i of 150 mg-NO_3/L: V = 40 V, HRT = 2.5 h, D = 3 cm, and A_{eff} = 47.1 cm^2.
    - For C_i of 200 mg-NO_3/L: V = 40 V, HRT = 2.5 h, D = 3 cm, and A_{eff} = 61.4 cm^2.
- The highest nitrate removal efficiency from aqueous solutions was obtained for C_i = 200 mg-NO_3/L, V = 40 V, HRT = 3.5 h, D = 3 cm, and A_{eff} = 61.4 cm^2 when the parameters: TDS_i, pH, and T_i were kept constant at 1,000 mg/L, 7 ± 0.1, and 25°C ± 2°C. The nitrate removal efficiency reached 91.6%.
- The effectiveness of nitrate removal using EC from groundwater samples obtained from a well in the north of Gaza Strip showed that nitrate removal efficiency was 80% at the optimal conditions: V = 40 V, HRT = 2.5 h, D = 3 cm and A_{eff} = 61.4 cm^2, when TDS_i and C_i were 1,000 mg/L and 200 mg-NO_3/L, respectively. Therefore, EC is an efficient alternative technique for nitrate removal from aqueous solution. Moreover, it was possible to reduce nitrate concentration in groundwater to 50 mg-NO_3/L, the WHO maximum allowable nitrate concentration limit for drinking water for adults.
- This study generated important information on the best operating conditions of several variables that influence the EC process.

7. Recommendations

The following recommendations may be considered for further studies.

- Investigating the influence of the length to width ratio of the continuous flow reactor, energy consumption, the ratio of surface area of the electrode to volume of water in the reactor (A_s/V), and electrode passivation.

- Investigating the use of horizontal cathode (placed at the bottom of the cell) to enhance the flotation process by generating more distributed bubbles.
- Initiating a pilot project study in one of the north Gaza wells using an inline pipe reactor to investigate the feasibility of this method in nitrate removal from groundwater.

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