MODELING AND IMPROVEMENT OF AQUEOUS POLLUTANTS REMOVAL USING ELECTROCOAGULATION TECHNIQUE

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Abstract

This study investigates the possibility of using electrocoagulation to remove nitrate form water. An electrocoagulation reactor, with net dimensions of 0.21 m × 0.24 m × 0.3 m, has been used to remove nitrate considering the effects of different parameters, such as flow rate (Q), applied current (I), hydraulic detention time (HDT), conductivity ($\mho$), initially nitrate concentration (Ci), distance between electrodes (Dis), connection method, and electrodes material (aluminium, brass, stainless steel, and copper). During the experimental work, the initial pH, initial temperature (Ti), and surface area of electrodes ($Aeff$) were kept constant at 7±0.2 °C, 25±3.0 °C, and 0.16 m^2, respectively. The obtained results showed that the removal efficiency of nitrate increased with the increase of both I and HDT, while it decreased with the increase of the Ci, Dis, and Q. The optimum removal efficiency (85.2%) was obtained using aluminium (Al) electrodes with monopolar connection method at I of 5 A, Q of 0.042 L/min of 8 ms/cm, Dis of 1 cm, and Ci of 50 mg/L. Additionally, a nonlinear multiple regression models were developed using the program to simulate the experimental results. It is noteworthy to mention that the novelty of this work is linking between the effects of HDT and each one of the studied parameters.

Keywords: Aluminium electrode, Brass electrode, Copper electrode, Electrocoagulation, Nitrate .

1. Introduction

One of the main challenges nowadays is equipping clean water to a huge plurality of the population around the world. Rivers, canals, lakes and other water bodies are constantly polluted due to indiscriminate discharge of industrial effluents as well as other anthropogenic activities [1-4]. For example, significant amounts of nutrients (phosphate and nitrate) are discharged on daily basis for the surface water from industrial and domestic wastewaters [5-8].

Nitrate is naturally occurring ions that are part of the nitrogen cycle. The nitrate ion (NO_3^-) is the stable form of combined nitrogen for oxygenated systems. Although chemically unreactive, it can be reduced by microbial action. Chemical and biological processes can further reduce nitrite to various compounds or oxidize it to nitrate [9, 10]. Nitrates enter the aquatic environment by agricultural wastewater (inclusive the excessive utilize of inorganic fertilizers and nitrogen), domestic wastewater, surface runoff, and oxidation in groundwater. Nitrites can be chemically created in the distribution pipes via bacteria [7, 11]. Nitrate is hazardous to infants and pregnant women due to the risk of methemoglobinemia (also called blue-baby syndrome). Where it has been stated that the reduction of nitrate to nitrite in the stomach of infants results in the binding of nitrite to haemoglobin and form methaemoglobin in the red blood cells (Eq. 1) [11].

$$\text{Haemoglobin (Fe}^{2+}) \rightarrow \text{Methaemoglobin} \tag{1}$$
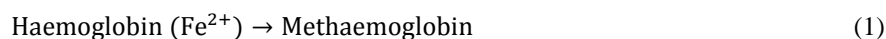
In studies reported via the World Health Organization (WHO), 97% for the cases where symptoms of cyanosis were observed occurred in infants that were mostly under 3 months old, and concentration of nitrate in water was over 50 mg/L. Beside the Methaemoglobinaemia, nitrates in drinking water can cause various types of cancer in people exposed to high concentrations quantities [12]. Thus, drinking water quality agencies and many governments are paying close attention to limiting nitrates in drinking water. Table 1 lists several international standards for the concentration of nitrates in drinking water in accordance with several international water quality standards. To meet these requirements, several treatment methods have been applied to remove nitrate from water and wastewater, such as adsorption, separation, electrocoagulation, and bio-degradation methods [13-22]. Electrocoagulation is one of the widely used method because no chemical are required in this method and the volume of the produced sludge is very small, which decreases the operational cost because the sludge (as a solid waste) required special handling and proper landfills to dispose in [23-30], or recycle it other industries such as concrete [31-33]. For example, Hashim et al. [7] used aluminium-based electrocoagulation cell to remove nitrate from synthetic drinking water. The authors stated that as high as 85% of nitrate could be removed after about an hour of treatment at current density of 2 mA/cm^2 and initial pH of 7. Another study about the removal of nitrate from water was carried out by Abdel-Aziz et al. [34]. The authors used electrocoagulation cell supplied with aluminium tubes (as electrodes) that horizontally oriented. The authors stated that complete removal efficiency could be achieved after 100 min of treatment at current density of 10.3 mA/cm^3.

In this context, the current study has been carried out to investigate the removal of nitrate from synthetic water samples under continuous flow conditions considering the effects of key operational parameters. It is noteworthy to mention that the novelty of this work is linking between the effects of HDT and each one of the studied parameters.

Table 1. International standard for nitrate concentrations in drinking water.

| Countries/ Organizations	Concentration of Nitrate	Reference
WHO	50 ppm (mg/L as NO_3^- /L)	[12]
The USEPA	10 mg NO_3^- N/L	[35]
Canada	45 mg/L as NO_3^- /L	[36]
Australia	50 ppm (mg- NO_3^- N/L)	[37]
Malaysia	50 ppm (mg -NO_3^-/L)	[38]

2. Mechanism of Electrocoagulation (EC)

The mechanisms of *EC* for water and wastewater treatment are very complex. It is believed that there are three other possible mechanisms involved besides *EC*, namely electro flotation, electrochemical oxidation and adsorption [39]. The main electrochemical reactions at the electrodes during *EC* process are [40]:
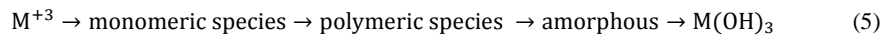
At the cathode, H_2 gas is liberated:

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_2 + 3OH^- \tag{2}$$

The metal cathode may be chemically attacked by OH^- especially at high pH values [41]:

$$2M + 6H_2O + OH^- \rightarrow 2M(OH)_4 + 3H_2 \tag{3}$$
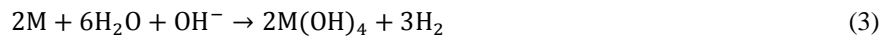
The sacrificial metal anode is dissolved [42]:

$$M \rightarrow M^{+3} + 3e \tag{4}$$

In conclusion, the formation of metal hydroxide flocs proceeds according to a complex mechanism, which may be simplified as follows:

$$M^{+3} \rightarrow \text{monomeric species} \rightarrow \text{polymeric species} \rightarrow \text{amorphous} \rightarrow M(OH)_3 \tag{5}$$

In the case of Al electrode monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^{2+}$, $Al_2(OH)_2^{4+}$, and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$ are formed during the EC process.

2.1. EC using aluminium electrodes

It has been well documented that in aluminium-based *EC* process the main reactions are [43, 44]:

$$Al^{+3} \rightarrow Al^{+3} + 3e^- (\text{Anode}) \tag{6}$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- (\text{Cathode}) \tag{7}$$

When the anode potential is sufficiently high, secondary reactions may occur, such as oxygen evolution [44]:

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \tag{8}$$

Aluminium ions (Al^{3+}) produced by electrolytic dissolution of the anode immediately undergoes spontaneous hydrolysis reactions, which generate various monomeric species according to the following sequence (omitting coordinated water molecules for convenience) [43]:

$$Al^{+3} + H_2O \rightarrow Al(OH)^{+2} + H^+ \tag{9}$$

$$Al(OH)^{+2} + H_2O \rightarrow Al(OH)_2^+ + H^+ \tag{10}$$

$$Al(OH)_2^+ + H_2O \rightarrow Al(OH)_3 + H^+ \tag{11}$$

Hydrolysis reactions make the anode vicinity acidic. Conversely, hydrogen evolution at the cathode makes the electrode vicinity alkali.

3. Material and Methods

3.1. Experimental set up

A continuous reactor is made of glass container with net dimensions of 0.21 m × 0.24 m × 0.3 m. All tests were carried out at initial pH of 7±0.2 and at room temperature (25±3 °C). Electrodes materials used in the *EC* process were brass, copper, aluminium, and stainless steel. In each experimental run, two electrodes (one cathode and one anode), with dimensions of 20 cm × 20 cm × 0.06 cm and inundation depth of 20 cm, were used. The effective area of the anode is 400 cm². Before each experiment, the pH of the solution was adjusted to 7 using a suitable volume of HCl or NaOH solution, while the conductivity was adjusted using NaCl. The hydraulic detention time (HDT) is defined by the following equation:

$$\text{HDT} = \frac{v_r}{Q} \tag{12}$$

where v_r is the volume of reactor (m^3) and Q is the flow rate (m^3 /s). Figure 1 shows schematic for the electrocoagulation system used in this work.
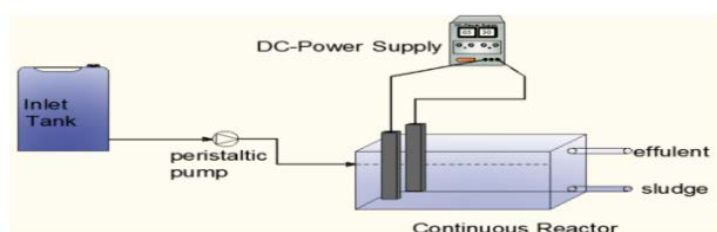

Fig. 1. Schematic diagram of the EC continuous flow.

3.2. Apparatus

The following apparatuses was used in the experiments

(1) DC power supply (type: UNIROI, 0-30 V, 0-5 A), (2) Aluminium, stainless steel, (3) Brass and copper electrodes, (4) Water pump machine, (5) Stop watch, (6) Spectrophotometer (SP-3000nano, OPTIMA), (7) pH meter, (8) HCl and NaOH solutions for pH correction, (9) Electrical coagulation cell: a continuous flow reactor with dimensions (length 0.3 m × 0.21 m × 0.24 m) made from glass, and (10) Flow meter.

4. Analytical Methods

4.1. Nitrate determination method (4500-NO₃⁻ B. ultraviolet spectrophotometric screening method)

Nitrate concentration has been calculated using the ultraviolet spectrophotometric screening method. In this method, the measurement was carried out at two different wavelengths, 275 nm and 220 nm [45, 46]. Wavelength of 275 nm can be eliminated in the nitrate detection; however, it was useful to detect nitrite. According to this standard method, common interfering material, such as bacterial cells, resulted in scattering at 275 nm and produced absorption at 218 nm two times as much as that at 275 nm. Table 2 shows that the blank absorbance (for deionised water).

Table 2. Blank (deionized water) absorbance.

Method	Wavelength
A275	Absorption (A) at 275 nm
A220	Absorption (A) at 220 nm
A_1 = A220 - 2*A275	Corrected absorption at 220 nm

4.2. Evaluation of removal efficiency

The efficiency of nitrate removal from synthetic water, treated with electrocoagulation method, has been calculated as follows [7]:

$$R\% = \frac{C_0 - C}{C_0} \times 100 \tag{13}$$

where R is the nitrate removal efficiency, C_o is the first nitrate concentration (mg/L as NO₃), and C is the nitrate concentration in any time (mg/Las NO₃).

4.3. Electro-coagulation model for removal of nitrates

The following formula has been developed, using Data Fit software (version 9.1. 32), basing on the collected data from the experimental work using aluminium electrodes at monopolar connection method:

$$y = 7.1703\,Ln(x) + 28.773 \quad (R^2 = 0.957) \tag{14}$$

where y is the overall coefficient of adsorption (or enmeshment) of the anionic pollutants (NO₃) on aluminium (Al) coagulants, which indicates the amount of anionic pollutants (AP) that can be removed by this coagulation mechanism for a given concentration of coagulant [47]. While $\times$ (net charge, expressed in mg L⁻¹) is the magnitude of the charges in the solution.

4.4. Energy and electrode consumption

Energy consumption can be calculated as follows [48, 49]:

$$E = \frac{C \times A \times Mw}{Nd \times K} \tag{15}$$

where E, C, A, M_w, N_d and K are the quantity of molten electrode material (g-M/cm²), current density (A/cm²), electrolysis time (s), the molecular weight of the metal electrode (g/mole), the number of electrons in the oxidation / reduction reaction and constant of Faraday (96,500 coulomb/mole), respectively.

4.5. Power consumption

Power consumption could be calculated as follows [6]:

$$En = \frac{I \times U \times T}{Vol.} \tag{16}$$

where En, I, U, T and Vol. represent power consumption (kW.h/m³), current (A), voltage (V), electrolysis time (h) and volume of water to be treated (m³), respectively. The operational conditions that will be tested in the current study are the flow rate, current density, conductivity, distance between electrodes, initial nitrate concentration and type of connection (bipolar, mono-polar in parallel and mono-polar in series).

5. Results and Discussion

5.1. The effect of HDT on nitrate removal

The effect of HDT on nitrate removal has been examined by treating nitrate-containing solutions at different HDTs (from 2-6 hours). To investigate the effect of HDT, the flow rate was changed from 0.042, to 0.1, and 0.15 L/min that gives three different HDTs (2, 4 and 6 hours). Figure 2 shows the influence of HDT on nitrate removal for different electrodes materials. It is noted from this figure that the removal efficiency decreased upon increasing of the flow rate (i.e., decreasing the HDT). This reduction in efficiency could be explained by the fact that the decrease of residence time in the electrocoagulation reduces the quantity of liberated ions from anode, which decreases the removal efficiency [50].
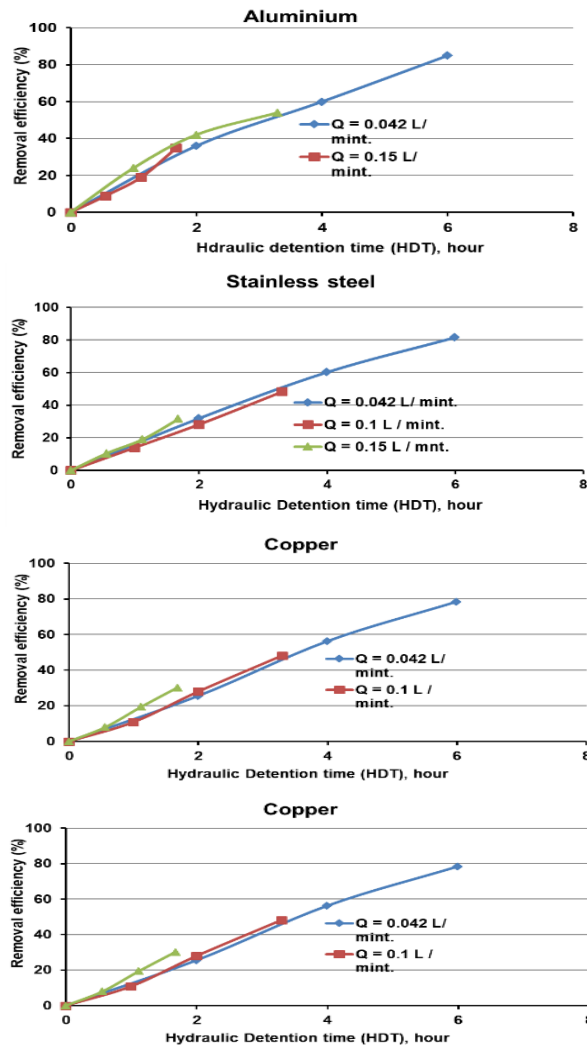

**Fig. 2. Effect of HDT on nitrate removal at
different flow rates for different electrode materials.**

5.2. The effect of HDT on nitrate removal in relation to applied current (I)

The effect of HDT on nitrate removal is examined with solutions of HDTs ranging from 2-6 hours. To investigate the effect of HDT, applied current is fixed at certain value (2, 3, and 5 A) while HDT is changed to 2, 4 and 6 hours. Figure 3 shows that the removal efficiency of nitrate is increased upon increasing the applied current at condition of Dis = 1 cm, EC = 8 ms/cm, monopolar connection, and Ci = 50 mg-NO$_3$/L).
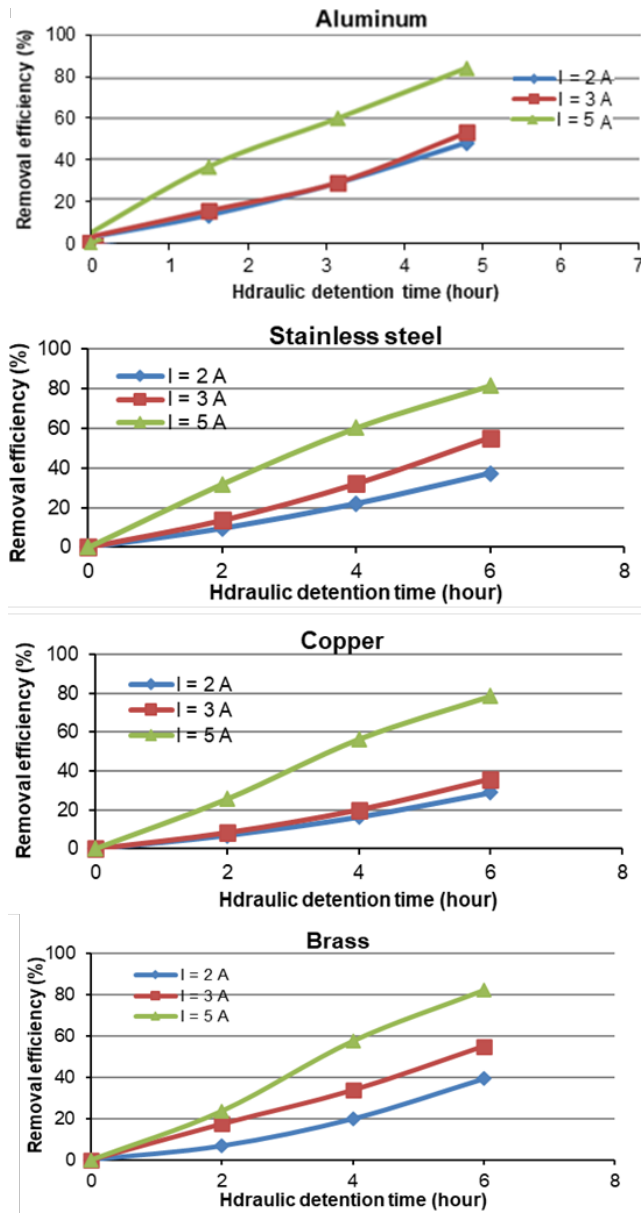

Fig. 3. Effect of HDT on the removal of nitrate at different *I* values.

This enhancement in the removal efficiency with the increase in the applied current could be attributed to the increase in the generation of coagulants that enhances the removal efficiency [7].

5.3. The effect of HDT on nitrate removal in relation to conductivity (℧)

In this part of the experimental work, the effect of HDT on nitrate removal has been carried out at different conductivities (4, 6, and 8 ms/cm). Figure 4 indicates that the removal efficiency increased upon increasing the conductivity of solution being treated. The enhancement of nitrate removal with the increase of water conductivity might be explained by the fact that increasing the amount of the added NaCl increases the ionic species that facilitate the passage of the current, which enhances the removal efficiency [51].
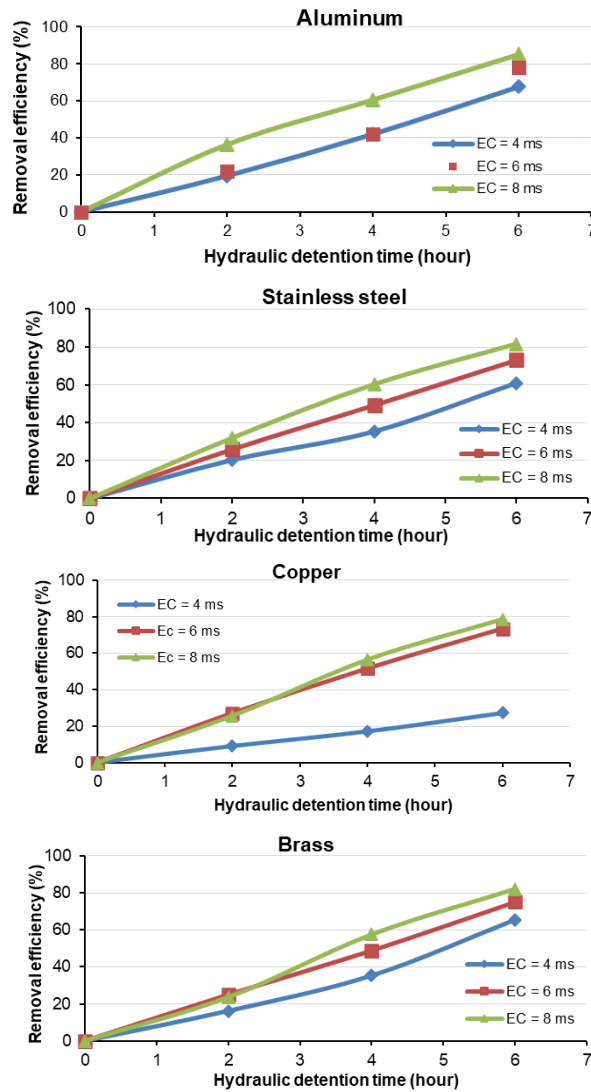

Fig. 4. Effect of HDT on the removal of nitrate at various ℧ values.

5.4. The effect of HDT on nitrate removal in relation to initial nitrate concentration (Ci)

The effect of HDT on nitrate removal has been examined for solutions with three different initial nitrate concentrations (Ci) (50, 100, and 150 mg/L). Figure 5 indicates that the removal of nitrate decreased upon increasing the initial nitrate concentration. The explanation of this decrease in nitrate removal with the increase in the initial concentration is the fact that a constant number of coagulants is generated from the anode for the same applied current and treatment time. Thus, the formed flocs, at high nitrate concentration, will not be enough to absorb all pollution ions [42].
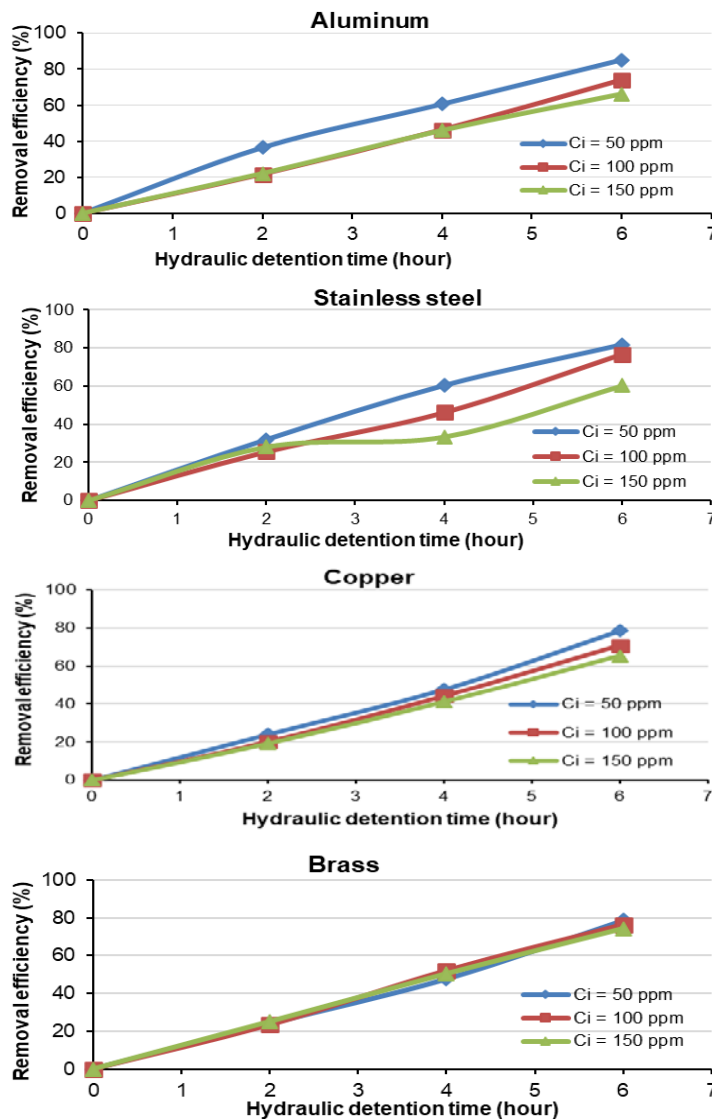

Fig. 5. Effect of HDT on the removal of nitrate for various Ci values.

5.5. The effect of HDT on Nitrate removal in relation to types of connection

The effect of HDT on nitrate removal is examined at different two different connection methods, namely monopolar and bipolar methods at Q = 0.042 L/min, Dis = 1 cm, I = 5 A, EC = 8 ms/cm, and Ci = 50 mg-NO$_3$/L). Figure 6 indicates that the best type of connection for nitrate removal is monopolar.
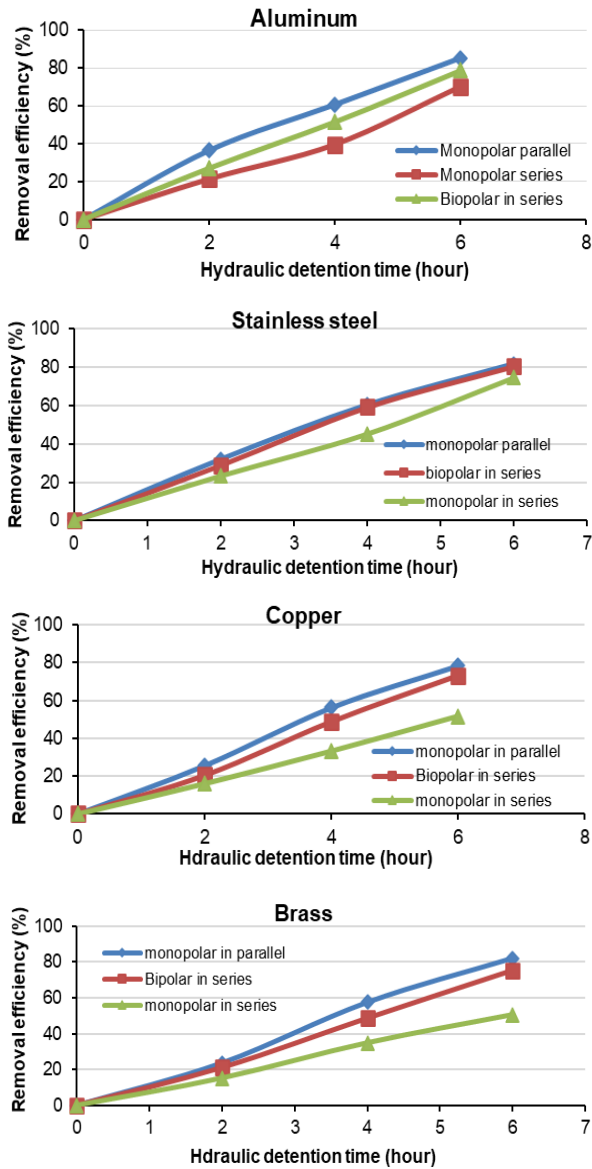

**Fig. 6. Effect of HDT on the removal
of nitrate at different types of connection.**

5.6. The effect of HDT on nitrate removal in relation to distance between electrodes (Dis)

The effect of HDT on nitrate removal is examined at different distances between electrodes (1, 2, and 3 cm). Figure 7 shows that the removal efficiency decreased with the increase of Dis.
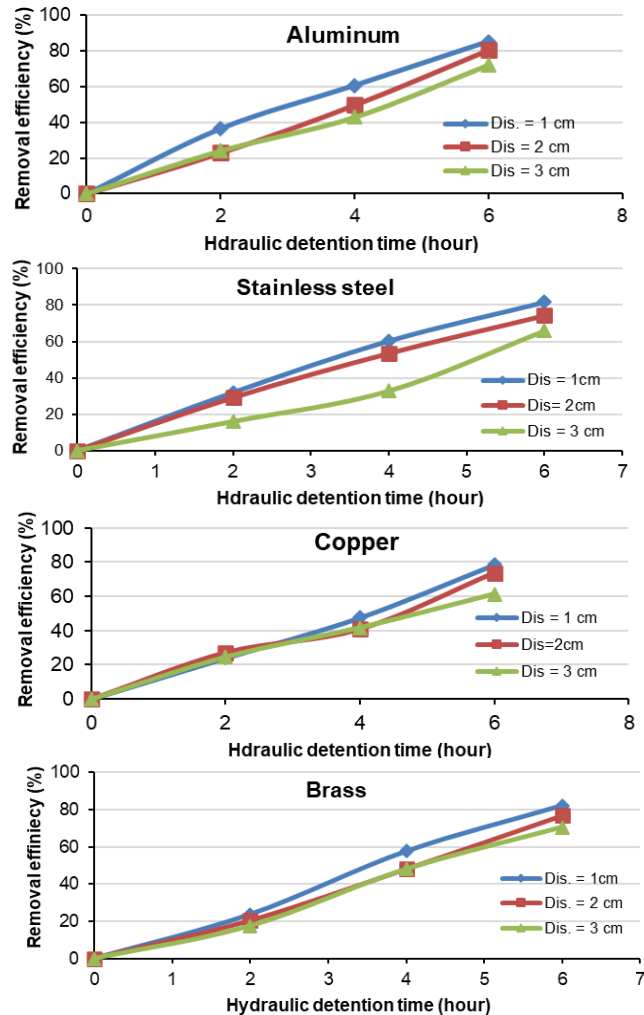

Fig. 7. Effect of HDT on the removal
of nitrate at different distances between electrodes.

This decrease in the removal efficiency could be attributed to the increase in the resistance to the flow of current and the acceleration of growth of the passive layer on the surfaces of anode [7]. The best is distance between electrodes for nitrate removal, in this study, is 1 cm. By the end of this work, the authors recommends, for future works, to use a proper type of sensors to monitor and control the key

operating parameters in the electrocoagulation units because the sensing technology has recently been applied in different industries, such as water quality and health of structures [52-55].

The outcomes of the current study are very comparable to those in the literature, for example a similar removal efficiency (about 85%) was obtained by [7].

6. Conclusions

Continuous flow experiments are designed to investigate the effects of the different parameters including flow rate (Q), applied current (I), hydraulic detention time (HDT), conductivity ($\mho$), initial concentration of nitrate in solution (Ci), electrode spacing (Dis), method of connection, and electrode materials on nitrate removal from water samples using electrocoagulation method. The main conclusions obtained from this work are:

- Electrocoagulation method is successfully alternative for the remove nitrate from aqueous solution. The performance of this method has been found to be dependent on the initial nitrate concentration, distance between be influenced by several operating parameters, such as electrodes, flow rate (Q), applied current (I), hydraulic detention time (HDT), conductivity ($\mho$), initial concentration of nitrate in solution (Ci), electrode spacing (Dis), connection method, and electrode materials.

- The performance of the electrocoagulation method, in terms of nitrate removal, can be enhanced by increasing the applied current (I), hydraulic detention time (HDT) and conductivity ($\mho$).

- The increase in initial nitrate concentration, distance between electrodes, and/or flow rate negatively influenced the ability of the electrocoagulation method to remove nitrate from water.

Nomenclatures	
A_{eff}	Surface area of electrodes, m^2
C_i	Initially nitrate concentration, mg/L
Dis	Distance between electrodes, mm
I	Applied current, mA
pH	Initial pH
Q	Flow rate, L/min
Ti	Initial temperature, °C
Greek Symbols	
$\mho$	Conductivity, ms/cm
Abbreviations	
HDT	Hydraulic detention time, hours

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