

Research Article

## Factors Influencing Treatment of Nitrate Contaminated Water using Batch Electrocoagulation Process

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### Abstract

*Electrocoagulation process involves applying an electrical current to sacrificial electrodes inside a reactor where the current generates coagulant agent "in situ" and gas. Electrocoagulation is currently experiencing both increasing popularity and considerable technical improvements. Nitrate contamination of water resources has become an increasing problem globally; nitrate levels in water are limited due to health concern. A laboratory batch electrocoagulation reactor was designed to investigate the influence of different operating parameters on the removal efficiency of nitrate from wastewater. The studied parameters was cell current density (6-14 mA/cm<sup>2</sup>), initial pH (7-11), and initial nitrate concentration (100-500 mg/l), also a comparison between aluminum and iron electrodes was investigated. The results showed that electrocoagulation effectively removed nitrate from wastewater, cell current density and initial pH play important roles. The best removal efficiency of nitrate was achieved at pH=9, also the results showed that aluminum electrodes were slightly more efficient than iron electrodes.*

**Keywords:** Nitrate removal, Electrocoagulation, Batch system, Aluminum electrodes.

### 1. Introduction

At the turn of the last century, it was estimated that some 1.1 billion people (one-sixth of the world's population) were without an 'improved' water supply, while in the foreseeable future the demand for water is only expected to grow as human population and industrialization increases (Holt, *et al*, 2005).

The demand for quality drinking water quality is increasing globally and environmental regulations regarding wastewater discharge are becoming increasingly stringent. Therefore, it has become necessary to develop more effective treatment methods for water purification and/or enhance the operation of current methods. This and eco-friendliness have led to increasing global interest in electrocoagulation as a research subject (Kuokkanen, *et al*, 2013).

Nitrate contamination of water resources has become an increasing problem globally due to the extensive use of nitrogen fertilizers and improper treatment of waste water from industrial sites. Within the human body, nitrates may be reduced to nitrites that combine with haemoglobin to form methaemoglobin, which can be fatal to neonates (Li, *et al*, 2010).

The World Health Organization (WHO) and the European Community have set a maximum

contaminant level (MCL) of 50 mg NO<sub>3</sub><sup>-</sup>/l which is equal to 11.3 mg NO<sub>3</sub>-N/l (WHO, 2011), whereas the US Environmental Protection Agency (USEPA) have set an MCL of 10 mg NO<sub>3</sub>-N/l (USEPA, 2009). According to international standards, drinking water must contain no more than 50 ppm of nitrate and a guide level of 25 ppm is highly recommended (Elmidaoui, *et al*, 2001).

There are different technologies to remove nitrates from drinking water such as: ion exchange, reverse osmosis, electrodialysis, catalytic denitrification, biological denitrification, and electrochemical denitrification (Emamjomeh, *et al*, 2009).

Many researches have been carried out on the removal of nitrate by electrocoagulation. Lin and Wu (1996), Paidar *et al* (1999), Koparal and Ogutveren (2002), Ugurlu (2004), Emamjomeh and Sivakumar (2005). However further investigation needed to be done. Electrocoagulation (EC) is an emerging technology that combines the functions and advantages of conventional coagulation, flotation, and electrochemistry in water and wastewater treatment. Each of these fundamental technologies has been widely studied separately. However, a quantitative appreciation of the mechanism of interaction between these technologies employed in an electrocoagulation system is absent (Emamjomeh, *et al*, 2009).

The aim of this study was to conduct experiments to investigate the removal of nitrate from wastewater using batch electrocoagulation reactor and to study the

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influence of different operating parameters on the removal efficiency.

## 2. Electrocoagulation

### 2.1. Theoretical Background on EC process

Electrocoagulation (EC) is a complicated process involving many chemical and physical phenomena that use consumable electrodes to supply ions into the wastewater stream. In an EC process the coagulating ions are produced 'in situ'. Electrocoagulation process involves applying an electric current to sacrificial electrodes inside a reactor tank where the current generates a coagulating agent and gas bubbles. This process has three stages: 1) coagulants formation due to anode electrical oxidation, 2) destabilizing pollutants and suspended substances and emulsion breaking and 3) combining instable particles to form floc (Malakootian, *et al*, 2011).

Fig.1 shows the complex, interdependent nature of the electrocoagulation process. A sacrificial metal anode (usually aluminum, but sometimes iron) is used to dose polluted water with a coagulating agent. Simultaneously, electrolytic gases (mainly hydrogen at the cathode) are generated. It is possible to identify three separate categories of mechanistic processes/ electrochemistry, coagulation, and hydrodynamics that form the basis of electrocoagulation. The fact that these processes are difficult to investigate separately in an operational reactor goes some way towards explaining the lack of a detailed technical literature on electrocoagulation (Holt, *et al*, 2002).

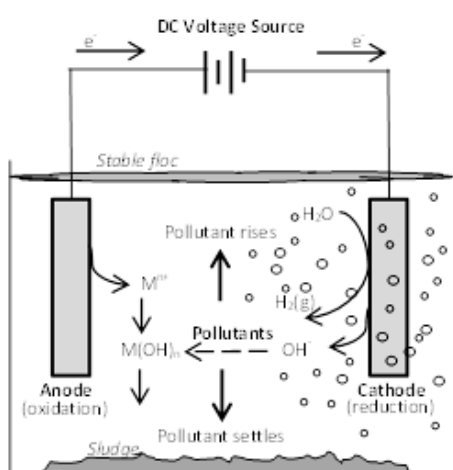


Fig.1 Schematic diagram of two-electrodes EC cell

The measured potential of the EC cell is the sum of three components:

$$\eta_{AP} = \eta_k + \eta_{Mt} + \eta_{IR} \quad (1)$$

where  $\eta_{AP}$  is the applied overpotential (V),  $\eta_k$  the kinetic overpotential (V),  $\eta_{Mt}$  the concentration overpotential (V),  $\eta_{IR}$  the overpotential caused by

solution resistance or IR-drop (V). The IR-drop is related to the distance ( $d$  in cm) between the electrodes, surface area ( $A$  in  $m^2$ ) of the cathode and specific conductivity of the solution ( $k$  in  $mS\ m^{-1}$ ) and current ( $I$  in A) by the equation shown below

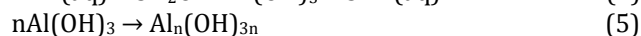
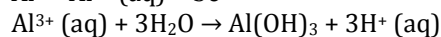
$$\eta_{IR} = I \frac{d}{A \cdot k} \quad (2)$$

The IR-drop can be easily minimized by decreasing the distance between the electrodes and increasing the area of cross-section of the electrodes and the specific conductivity of the solution. Concentration overpotential, also known as mass transfer or diffusion overpotential, is caused by the change in analyte concentration occurring in the proximity of the electrode surface due to electrode reaction. The concentration overpotential is negligibly small when reaction rate constant is much smaller than the mass transfer coefficient. The mass transport overpotential can be reduced by increasing the masses of the metal ions transported from the anode surface to the bulk of the solution and can be achieved by enhancing the turbulence of the solution (Mollaha, *et al*, 2004).

### 2.2. Mechanism of EC

#### 2.2.1. Aluminum electrode

Electrocoagulation of wastewater using an aluminum electrode has been reported by a number of authors. The electrolytic dissolution of the aluminum anode produces the cationic monomeric species such as  $Al^{3+}$  and  $Al(OH)_2^+$  at low pH, which at appropriate pH values are transformed initially into  $Al(OH)_3$  and finally polymerized to  $Al_n(OH)_{3n}$  according to the following reactions (Yousuf, *et al*, 2001):

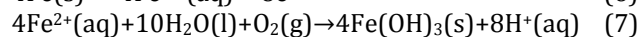


#### 2.2.2. Iron electrode

Iron upon oxidation in an electrolytic system produces iron hydroxide,  $Fe(OH)_n$ , where  $n = 2$  or  $3$ . Two mechanisms have been proposed for the production of  $Fe(OH)_n$ .

##### • Mechanism 1

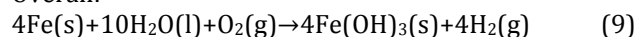
Anode:



Cathode:

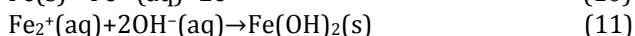
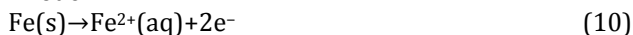


Overall:

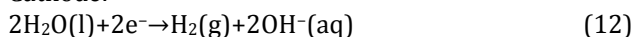


### • Mechanism 2

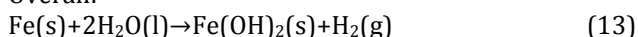
Anode:



Cathode:



Overall:



### 2.3. Advantages and disadvantages of

#### 2.3.1 Advantages of EC

Some of the advantages of using EC are EC (Yousuf, et al, 2001):

1. EC requires simple equipment and is easy to operate with sufficient operational latitude to handle most problems encountered on running.
2. Flocs formed by EC are similar to chemical floc, except that EC floc tends to be much larger, contains less bound water, is acid-resistant and more stable, and therefore, can be separated faster by filtration.
3. The gas bubbles produced during electrolysis can carry the pollutant to the top of the solution where it can be more easily concentrated, collected and removed.
4. The EC technique can be conveniently used in rural areas where electricity is not available, since a solar panel attached to the unit may be sufficient to carry out the process.

#### 2.3.1. Disadvantages of EC

Some of the disadvantages of using EC are:

1. An impermeable oxide film may be formed on the cathode leading to loss of efficiency of the EC unit.
2. High conductivity of the wastewater suspension is required.

## 3. Materials and Methods

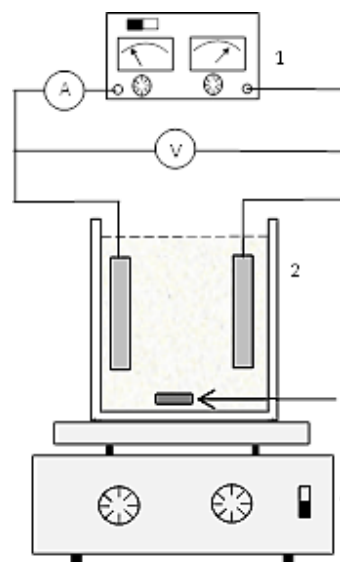
### 3.1. Wastewater sample

The wastewater used in this study was a synthetic nitrate solution prepared by adding potassium nitrate ( $\text{KNO}_3$ ) (purity  $\geq 95\%$ ) to distilled water. 1 g/l of sodium chloride ( $\text{NaCl}$ ) was added to the nitrate solutions as a supporting electrolyte.

### 3.2. Experimental Setup

The experimental setup used for the electrocoagulation studies is shown in Fig. 2. The volume of the wastewater was one liter. To achieve good mass transfer, a magnetic stirrer (LMS HTS-1003, Japan) was used. A DC power source (Dazheng Model PS-303D, 0-30V, 0-3A) was used to supply constant cell currents.

Aluminum and iron electrodes plates of size 8 cm  $\times$  3 cm  $\times$  0.3 cm were used, and the distance between the electrodes was set at 3 cm.



**Fig.2** Schematic diagram of electrocoagulation (EC) process unit. (1) DC power supply; (2) EC cell; (3) Teflon bar (4) Magnetic stirrer

### 3.3. Experimental Procedure

All experiments were conducted at ambient temperature  $25 \pm 1$  °C. One liter wastewater was placed into the reactor and all the runs were performed at constant stirring speed.

Many variables were investigated for its effect on the removal efficiency of the  $\text{NO}_3^-$  from wastewater such as, electrolysis time that ranged from 10 to 90 minutes, initial  $\text{NO}_3^-$  concentration ( $C_0$ ) from 100 to 500 mg/l, current density ( $i$ ) ranged from 6 to 14  $\text{mA/cm}^2$ , solution pH in the range from 7 to 11. Aluminum and iron electrode were used in this study.

After the EC process, the power was switched off and the electrodes were dismantled.

### 3.4. Analytical Procedure

$\text{NO}_3^-$  concentrations were measured by using a UV/Visible Spectrophotometer (Jenway Model 6800 Double Beam). A calibration curve was prepared for the  $\text{NO}_3^-$  concentration range used by adjusting the wave length at 220 nm.

5 ml samples were drawn at different time intervals, diluted to 50 ml with distilled water, filtered to remove any possible interference from suspended particles, and then acidified with HCl to prevent interference from hydroxides.

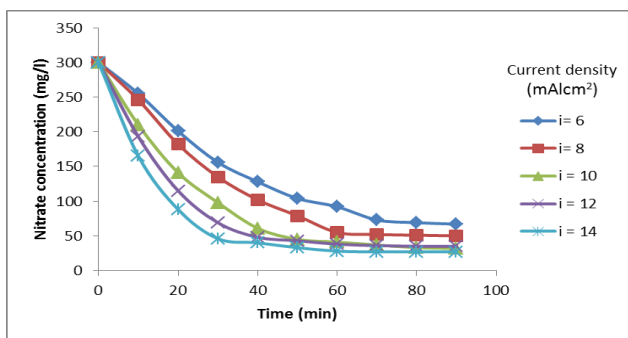
The removal efficiency (R) was calculated using the following equation:

$$\%R = \frac{C_0 - C}{C_0} \times 100\% \quad (14)$$

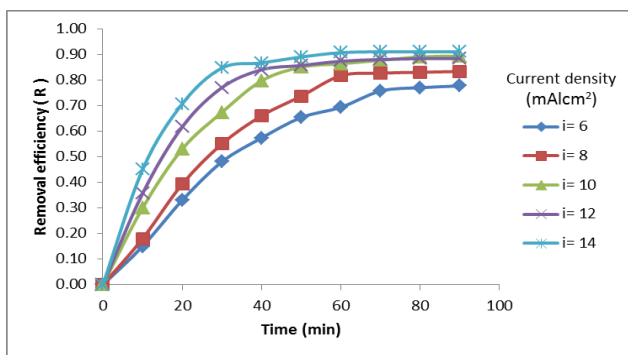
### 4. Results and Discussion

#### 4.1. Effect of current density

The effect of current density on the removal of nitrate from wastewater was studied and the results are shown in Figs. 3-4. From these results it is evident that increasing the current density causes the increase of nitrate removal. At current density 6 mA/cm<sup>2</sup> the removal efficiency of nitrate was 77.67% while at current density 14 mA/cm<sup>2</sup> the removal efficiency of nitrate was 91%. This result can be explained as follow: according to Faraday’s law, when increasing the current density the aluminum released from the anode is increased and hence increases the nitrate removal.



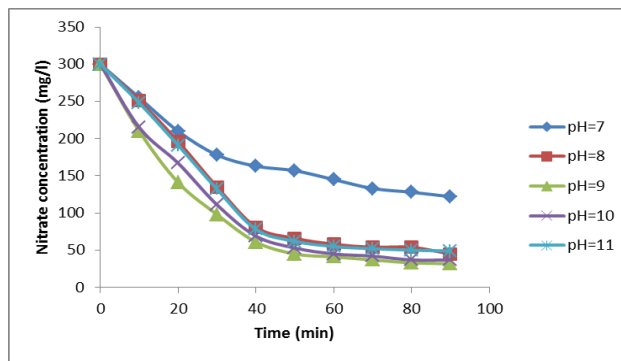
**Fig. 3** Effect of current density on the removal of nitrate (initial pH= 9, C<sub>0</sub>= 300 mg/l, Aluminium electrodes)



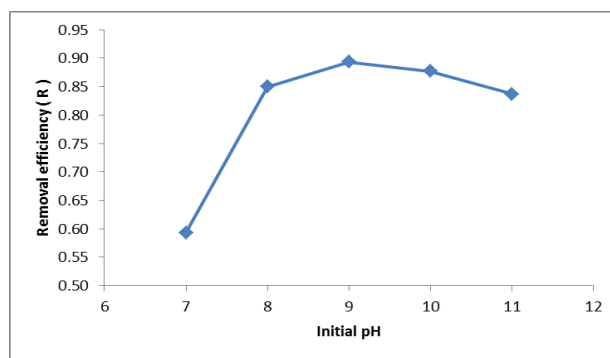
**Fig. 4** Removal efficiency of nitrate at different current densities (initial pH= 9, C<sub>0</sub>= 300 mg/l, Aluminium electrodes)

#### 4.2. Effect of pH

Usually pH is considered as an effective factor on the performance of electrocoagulation. The effect of initial pH on the removal of nitrate from wastewater was studied and the results are show in Figs. 5-6. The removal efficiency values at pH 7, 8, 9, 10, and 11 were 59.33%, 85%, 89.33%, 87.67%, and 83.67% respectively. It is clear that best nitrate removal efficiency was achieved at pH=9. At pH=7 the removal efficiency of nitrate was very low and this may be attributed to the formation of protective oxide layer on the aluminum.



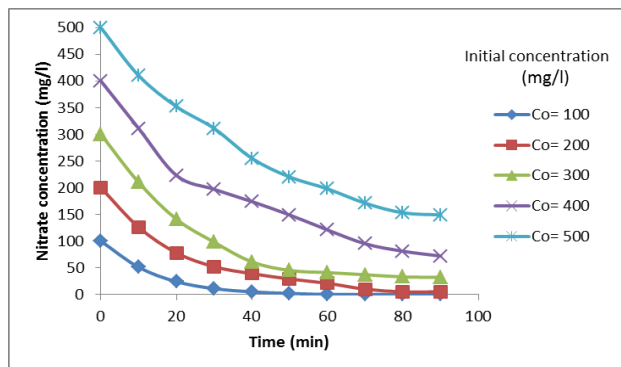
**Fig. 5** Effect of initial pH on the removal of nitrate (i= 10 mA/cm<sup>2</sup>, C<sub>0</sub>= 300 mg/l, Aluminium electrodes)



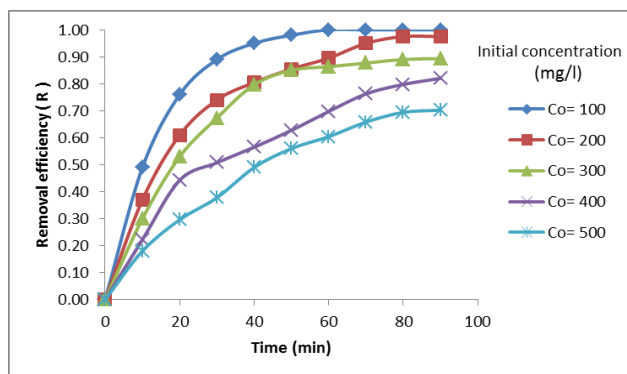
**Fig. 6** Removal efficiency of nitrate at different pHs (i= 10 mA/cm<sup>2</sup>, C<sub>0</sub>= 300 mg/l, Aluminium electrodes)

#### 4.3 Effect of initial concentration

Experiments were carried out at different initial nitrate concentrations to assess the effect of initial concentration on the removal efficiency and the results are shown in Figs. 7-8. These results showed that increasing nitrate concentration, at fixed other parameters, causes decrease in the removal efficiency of nitrate. This can be ascribed to the fact that ascribed to the fact that when increasing nitrate concentration more ions will present in the wastewater and this need more time to remove these ions.



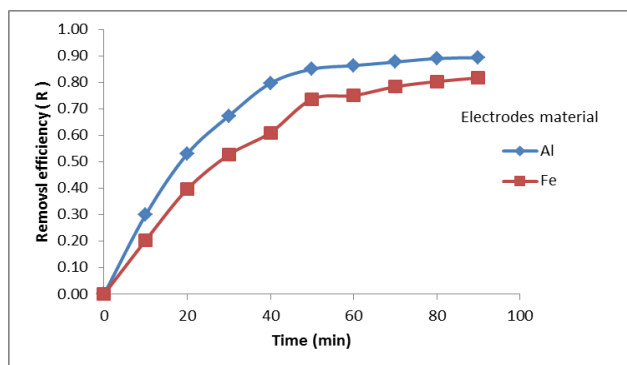
**Fig. 7** Effect of initial concentration on the removal of nitrate (i= 10 mA/cm<sup>2</sup>, pH= 9, Aluminium electrodes)



**Fig. 8** Removal efficiency of nitrate at different initial nitrate concentrations ( $i= 10 \text{ mA/cm}^2$ ,  $\text{pH}= 9$ , Alumium electrodes)

#### 4.4. Effect of electrode materials

Two different materials (aluminum and iron) were used during electrocoagulation. Fig. 9 shows that aluminum electrodes were slightly more efficient than iron electrodes in the removal of nitrate from wastewater. This may be attributed to the fact that aluminum cathode has lower hydrogen overvoltage than iron. Using iron electrodes gives yellow color to the water and therefore its uses are limited in water treatment.



**Fig. 9** Effect of electrode material on the removal efficiency of nitrate ( $i= 10 \text{ mA/cm}^2$ ,  $C_o= 300 \text{ mg/l}$ ,  $\text{pH}= 9$ )

#### Conclusions

Nitrate removal from wastewater was evaluated in a batch electrocoagulation reactor. The effect of different parameters: current density, initial pH, and initial nitrate concentration was studied and the results showed that concentration of nitrate was significantly reduces during the treatment process.

It was observed that pH play an important role in the removal of nitrate by electrocoagulation. In the range of studied parameters, the best removal efficiency of nitrate was observed at  $\text{pH}=9$ .

The results also showed that current density also an important parameter and increasing current density from  $6 \text{ mA/cm}^2$  to  $14 \text{ mA/cm}^2$  increases the removal efficiency of nitrate from 77.67% to 91%.

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