Nitrate Removal from Synthetic Industrial Wastewater by Using Electrocoagulation with monopolar Fe-Fe Electrodes

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Abstract

This work investigates the possibility of removing nitrate electro coagulation ions from synthetic industrial wastewater by using monopolar Fe-Fe electrodes in a batch electrochemical unit. Variables studied were current density, initial nitrate concentration, electrolysis time and supporting electrolyte. The results show that for initial concentration of 630 mg/L complete removal (100 %) of nitrate was noted at the current densities of 45 mA/cm^2 with a total energy consumption of 115 kWh/m^3, after 60 min of EC treatment, whereas when supporting electrolyte concentrations of 29 mg/L (NaCl) was used, complete removal of nitrate was noted at the current densities of 10 mA/cm2 with a total energy consumption of 1.88 kWh/m^3 after 10 min of EC treatment.

The results suggest that the process is effective for nitrate removal from synthetic industrial wastewater, and the economy of the process is favored by adding very small supporting electrolyte.

Keywords: Electrocoagulation, Nitrate removal, Monopolar electrode configuration, Iron electrodes, Industrial wastewater treatment, Supporting electrolyte.

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إزالة النترات من مياه الصرف الصناعية التركيبية باستخدام التخثير الكهربائي ذو الوصل الفردي لأقطاب الحديد الكهربائية

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الملخص

تم في هذا البحث اختبار إمكانية إزالة أيونات النترات من مياه الصرف الصناعية التركيبية باستخدام أقطاب الحديد الكهربائية ذات الوصل الفردي في وحدة التخثير الكهربائي الدفقي. وقد جرى استخدام عوامل دراسية مختلفة مثل كثافة التيار الكهربائي، وتركيز النترات الابتدائي، وزمن التحلل الكهربائي، والداعم الالكتروليتي. أظهرت نتائج الاختبارات أنّه تم الحصول على إزالة كاملة للنترات 100%، من أجل تركيز أولي للنترات قدره 630 mg/L لدى استخدام كثافة تيار كهربائي قدرها $45\ mA/cm^2$، وقد كان مصروف الطاقة الكلي خلال هذه العملية حوالي $115\ kWh/m^3$ بعد 60min. تم الحصول على إزالة كاملة للتركيز السابق للنترات لدى استخدام داعم الكتروليتي بتركيز 29mg/L (NaCl) وذلك عند استخدام كثافة تيار كهربائي أقل قدرها $10\ mA/cm^2$ ، وقد انخفض مصروف الطاقة الكلي خلال هذه العملية إلى حوالي $1.88\ kWh/m^3$ بعد 10 min . تشير نتائج البحث إلى فعالية عملية التخثير الكهربائي باستخدام أقطاب الحديد ذات الوصل الفردي في إزالة النترات من مياه الصرف الصناعية، وتتحسن كفاءة واقتصادية هذه العملية لدى إضافة كميات قليلة من الداعم الالكتروليتي.

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الكلمات المفتاحية: التخثير الكهربائي، إزالة النترات، الوصل الفردي للأقطاب الكهربائية، أقطاب الحديد، معالجة مياه الصرف الصناعية، الداعم الالكتروليتي.

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1. INTRODUCTION:

In some Middle East countries, including some places in Syria (e.g. east and south Ghota around Damascus), high nitrate concentrations in groundwater have been detected. Nitrate originated from agriculture is increasingly growing all over the world due to the extreme use of fertilizers. Nitrate salts reach the groundwater as they percolate through the soil. Other sources of nitrate in ground and surface water are from uncontrolled land discharges of unfinished treated or raw wastewater from domestic and industrial wastes, landfills and animal wastes particularly from animal farms. Some sources of nitrate in ground and surface water are from mineralization of nitrogen organic compounds in drainage water because of abundance of oxygen inside drainage pipes.

The use of nitrate (NO_3^-) in several industrial processes (as an oxidizing agent and in the production of explosives and as purified potassium nitrate in glass making and in inorganic fertilizers production etc.) leads to contamination of the environment and threaten natural life and public health for their hazardous and carcinogenic properties. Nitrate is one of nutrient elements, which can cause eutrophication problems of rivers and lakes. Nitrate is identified as one of the hazardous contaminants in potable water that may reduce to nitrosamines in the stomach which is suspected to cause gastric cancer [1].

Nitrite reacts with the hemoglobin in blood and converts the hemoglobin into methemoglobin, which does not carry oxygen to cell tissues. This phenomenon results in a bluish color of infant’s skin so called methemoglobinemia or the blue baby syndrome [2]. Therefore, it is very important to control the nitrate level in natural water. Nitrate is a stable and highly soluble ion with low potential for co-precipitation or adsorption. These properties make it difficult to be removed from water. Therefore, treatment for nitrate is typically very complicated and expensive.

New technologies are used to remove nitrate from water including ion-exchange (IX) [3-4], electro dialysis (ED) [5], nanofiltration (NF)/ reverse osmosis (RO) [6], biological denitrification [7], chemical reduction [8-9] and electrochemical reduction [10].

Reverse osmosis, electro dialysis and ion exchange are not selective for nitrate and require frequent regeneration of the media. All these processes do not destroy nitrate and generate secondary brine wastes. Biological denitrification is unfavorable because it requires intensive maintenance and constant supply of organic substrates. Additional drawbacks include biomass sludge disposal and treatment (aeration and disinfection) of denitrified water. Moreover, these microbial processes are generally slow and sometimes incomplete compared with chemical reduction [11]. When ferrous iron, hydrogen and formic acid are used as reducing agent in chemical reduction, catalysts must be used. Using Fe^{2+} and zero-valent iron as reducing agents also produces voluminous amounts of sludge. Using hydrogen and formic acid as reducing agents not only need control the composition of the catalysts, but also the pH of the treated solution, and these might be difficult in the treatment process of drinking water.

Compared with chemical reduction of nitrate, the electrochemical method might be the most favorable one. Among the electrochemical techniques, electrocoagulation - EC is found to be quite interesting both for economical and environmental reasons.

EC has the potential to be the distinct economical and environmental choice for treatment of industrial wastewater and other related water management issues. EC is an efficient technique since adsorption of hydroxide on mineral surfaces are a 100 times greater on in situ rather than on precipitated hydroxides when metal hydroxides are used as coagulant. EC needs simple equipments, designable for virtually any size. It is cost-effective, and easily operable. The start-up and operating costs are relatively low. It requires low maintenance cost with no moving parts. Since practically no chemical addition is required in this process, it brings minimum chance of secondary pollution. It needs low current, and it can be run even by green processes, such as, solar, wind-mills and fuel cells. EC process can effectively destabilize small colloidal particles. It produces minimal sludge. Gas bubbles produced during electrolysis can enhance flotation. It removes the smallest colloidal particles efficiently compared to conventional chemical and biological techniques, because the smallest charged particles have greater probability of being coagulated by the electric field that sets them in motion. In addition to these, the following advantages of EC process can be added: electrochemical-based systems allow controlled and rapid reactions, smaller systems become viable and, instead of using chemicals and microorganisms, the systems employ only electrons to facilitate water treatment [12].

The advantages of EC have encouraged many studies on the use of this technology for treatment of several industrial effluents such as paper wastes [13], oil wastes [14-15], textile dyes wastes [16], polymeric wastes [17], phenolic waste [18]. EC has been found a promising technique in treating urban wastewater [19], restaurant wastewater [20], potato chips wastewater [21], laundry wastewater [22], alcohol distillery wastewater [23], house wastewaters [24], dairy wastewater [25] and biodiesel wastewaters [26].

In recent years, there is much interest in using EC for treatment of water and wastewater containing phosphate [27], fluorine [28], phenol [18], sulfide, sulphate and sulfite ions [29], suspended particles [30], humic acids [31] and heavy metals [32- 35].

Recently, EC has been used in removing of nitrate from water and wastewater. EC was investigated by various workers [36-38] for nitrate removal. These studies show the successful removal of nitrate from water and wastewaters. It is therefore important and necessary to find out technical and economic feasibility of EC for removal of nitrate from water and wastewater.

The coagulation / destabilization mechanisms and removal efficiencies closely relate to the coagulant species present in the system. Thus, it has been reported in the literature that nitrates can be removed from wastewater via their adsorption onto the surfaces of hydroxide precipitates, which are generated from metals and released by the electrodes [36-37]. From this point of view the target of the present study is to examine the performance of an electrocoagulation unit with vertical bipolar Fe-Fe electrodes operating in batch regime for the removal of nitrate from synthetic wastewater and to explore the effects of varying operating parameters such as the applied current density, initial concentration and contact time on nitrate removal.

2.MATERIALS AND METHODS:

To demonstrate the effect of varying operating parameters, a stock solution of 1 L containing different concentrations of nitrate was prepared with potasium nitrate (HIMEDIA Extra pure). Stock solutions of nitrate were made by adding the specific values of potassium nitrate (KNO_3) in deionized water. The model wastewater concentration was prepared from the stock solution by suitable dilution.

Experiments were made by using a cylindrical glass cell of 450 mL on a magnetic stirrer (Nuova II stir plate, USA). Six plates of iron electrodes were installed vertically with a spacer to ensure fixed distance and immersed to a 4 cm depth with an effective area of 100 cm^2, were used as electrodes in monopolar electrods with serial connections mode in the experiments. The inter-electrode distance was 0.3 cm. A DC-power supply (RXN-605D, China) characterized by the ranges 0–6 A for current and 0–80 V for voltage was used to apply different current densities. The Bench-scale EC reactor with monopolar electrodes in serial connection is shown in Fig. 1.

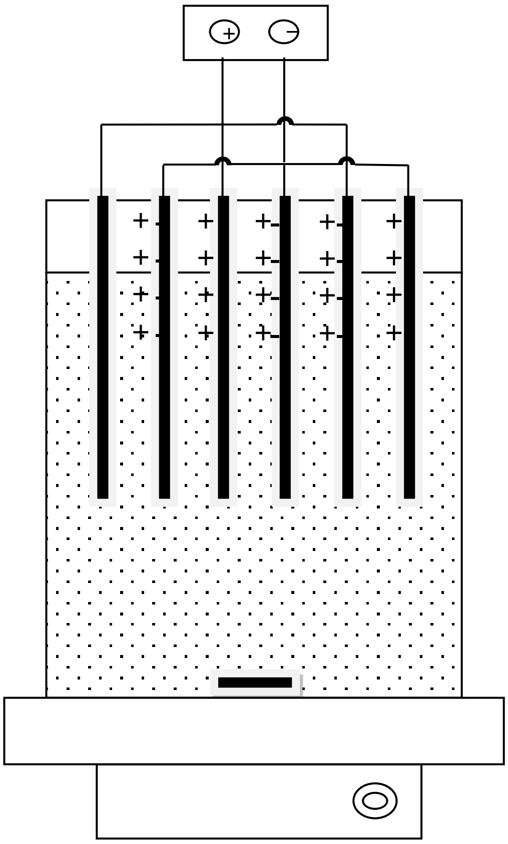

Fig. 1. Bench-scale EC reactor with monopolar electrodes in serial connection.

The effect of initial nitrate concentrations with different current densities on the system performance was investigated. A set of experiments were conducted with three different solutions containing nitrate concentrations of 630, 432 and 255 mg/L. The electrode surfaces were rinsed with diluted HNO_3 and distilled water and then dried to eliminate the oxides and passivation layers between the experiments. The polarity of the cell was reversed after each experiment to limit the formation of the passivation layers on the electrodes. To determine the system performance, samples of 2 mL were taken periodically from the reactor at predetermined time intervals (20 min) and diluted to 50 ml with distilled water. The residual nitrate concentrations were determined each run by using a DR Spectrophotometer (DR-890 Colorimeter Hach, USA). The pH of solutions at the begining of the EC process was 5.5. The experimental setup is shown in Fig. 2 .

To investigate the effect of current density on the removal yield, a series of experiments were carried out at different current densities i.e. 10, 25, 35 and 45 mA/cm^2. A series of experiments were carried out at different supporting electrolytes of 0, 1, 10 and 29 mg/L NaCl, to investigate the effect of supporting electrolyte on the removal yield. Using supporting electrolyte was for overcoming a high ohmic resistance between the anode and the cathode when electrical conductivity of the solution is low. All the runs were performed at laboratory temperature and at the stirring speed of 400 rpm.

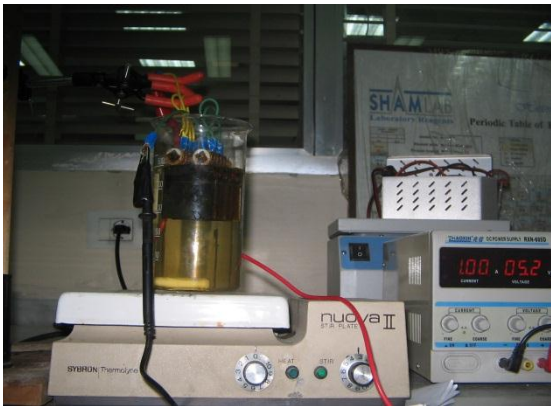

Fig. 2. Experimental setup used in the studies

3. RESULTS AND DISCUSSION:

3.1. Effect of current density

It is well known that the current density determines the production rate of coagulant, adjusts also bubble production, its size and distribution, and hence affects the growth of flocs coagulate particles in the EC reactor.

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.

For the initial concentration of 630 mg/L of nitrate, the removal efficiency values were 28.1, 38.6, 66.7 and 82.5% at the current densities of 10, 25, 35 and 45 mA/cm^2, respectively after 40 min EC treatment. Complete removal (100%) of nitrate was noted at the current densities of 45 mA/cm^2 after 60 min of EC treatment.

These results can be explained by the fact that, increasing current density will increase the Fe releasing from the anode according to Faraday's law with the formation of $Fe(OH)_3$ coagulant. Figs 3 and 4 also show that increasing the electrolysis time has increased the % NO^{3-} removal which can be attributed to fact that increasing the electrolysis time will certainly increase the available Fe^{+3} according to Faraday's law, and consequently increase the amount of $Fe(OH)_3$ which is responsible for NO^{3-} removal.

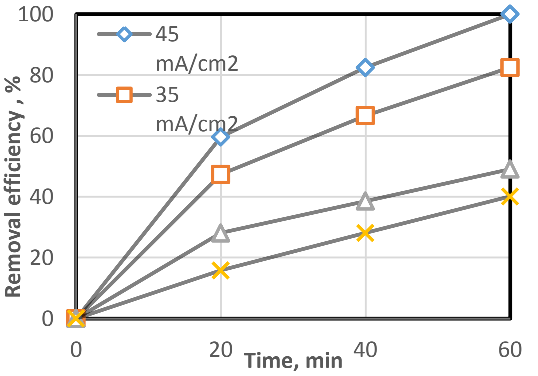

Fig. 3. Effect of current density on the removal efficiency of nitrate (Co= 630 mg/L)

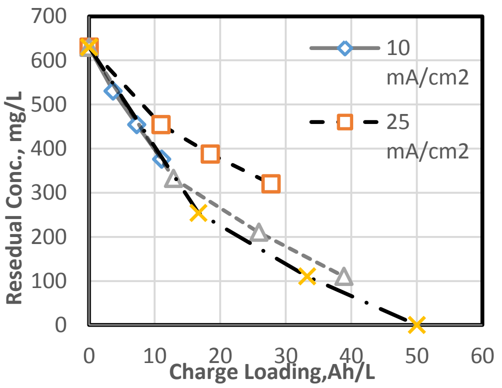

Fig. 4. Effect of current density on the residual concentrations of nitrate (Co= 630 mg/L)

First order kinetic model was applied to the results obtained. Nitrate removal kinetic fitted well as seen in Table 1. According to the Table 1, correlation coefficients (R^2) for nitrate increase with increasing current densities.

Therefore, it can be said in general that the removal of nitrate fits well with the first-order kinetic model at high current densities.

For instance, correlation coefficients of nitrate were calculated as 0.9723, 0.9856 and 0.9899 for current densities of 25, 35 and 45 mA/cm^2, respectively.

On the other hand, when the first-order rate constants (k) for current densities are investigated , it can be seen that k values are increasing comply with the removal results. As an example, k values are 0.0113, 0.0291 and 0.0387 min^{-1} for 25, 35 and 45 mA/cm^2, respectively for initial nitrate concentration of 630 mg/L. These results, at the same time, mean that at current densitiy of 45 mA/cm^2, nitrate can be removed 3.4 and 1.33 times faster in comparison to 25and 35 mA/cm^2, respectively.

Table1: First order rate constants for different current densities (Co=630 mg/L).

	25 mA/cm^2	35 mA/cm^2	45 mA/cm^2
R^2	0.9723	0.9856	0.9899
K min^{-1}	0.0113	0.0291	0.0387

Besides, energy consumption is directly related with the current density as also seen in Fig.5. For the initial concentration of 630 mg/L of nitrate, total energy consumption was 115 kWh/m^3 to gain 100% removal value at the current density of 45 mA/cm^2. Using the current average electrical energy price of 20 Syrian Pound for 1kWh the cost of energy consumed per m^3 of treated water is 2300 S.P. In the calculation of the operating cost, only energy costs was considered regardless of the output DC-power supply.

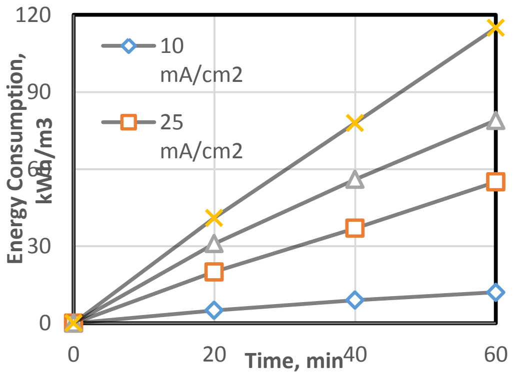

Fig. 5. Total energy consumption values as a function of current density (Co= 630 mg/L).

3.2. Effect of initial NO^{3-} concentration

Experiments were carried out at different initial nitrate concentrations to assess the effect of initial concentration on the removal efficiency.

For the current density of 10 mA/cm^2, the removal efficiency values were 28.1, 92.3 and 100% for the initial NO^{3-} concentrations of 630, 432 and 255 mg/L, respectively after 40 min EC treatment as shown in Fig.6. Complete removal (100%) of nitrate was also noted for the initial NO^{3-} concentrations of 432 mg/L at the current densities of 10 mA/cm2 after 60 min of EC treatment.

Fig. 7 shows that at the current density of 10 mA/cm^2 the NO^{3-} residual concentrations of 530, 199 and 0 mg/L were achieved for the initial NO^{3-} concentrations of 630, 432 and 255 mg/L, respectively after 20 min EC treatment.

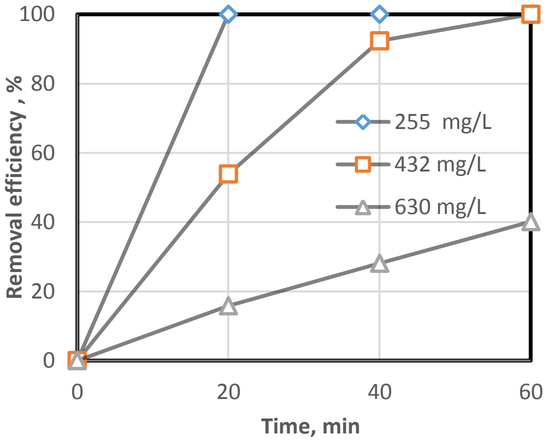

Fig. 6. Effect of initial concentrations of nitrate on the removal efficiency (i = 10 mA/cm^2)

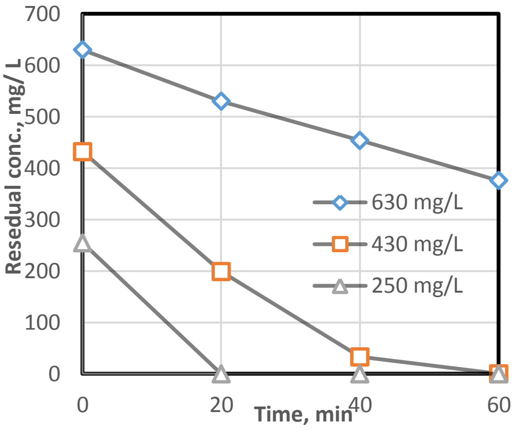

Fig. 7. Effect of initial concentrations of nitrate on the residual concentrations (i = 10 mA/cm^2)

As shown in figs 6-7, the nitrate removal efficiency decreased by increasing initial NO^{3-} concentration in the range from 255 to 630 mg/L. This can be ascribed to the fact that, increasing NO^{3-} concentration would probably increase activation polarization via adsorption on the anode and cathode with a consequent decrease in the rate of Fe dissolution at the anode and hydrogen evolution at the cathode. These results can be also explained by the fact that, increasing initial nitrate concentration will increase the needed time to remove nitrate ions, and the formed Fe will not be enough. It can be also noted when the nitrate removal efficiency has been increased by increasing the current density or adding supporting electrolyte.

In addition the figs 6-7 show that at higher concentrations of NO^{3-} (630 mg/L) the removal rate approximately reached maximum value at certain period approximately 60 minutes, which can be ascribed to the fact that increasing the NO^{3-} concentration will block the adsorption sites of $Fe(OH)_3$ rapidly and decreases its ability to adsorb more NO^{3-} ion.

3.3. Effect of supporting electrolyte

Using supporting electrolyte was also for decreasing the passivity of electrodes by removing the oxide layer on the electrode surface and for increasing the solution conductivity and energy consumption.

Fig. 8 shows that at the current density of 10 mA/cm^2, the NO^{3-} residual concentrations of 586, 220, 155 and 11 mg/L were achieved for supporting electrolyte concentrations of 0, 1, 10 and 29 mg/L (NaCl) respectively after 5 min EC treatment for the initial concentration of 630 mg/L of nitrate.

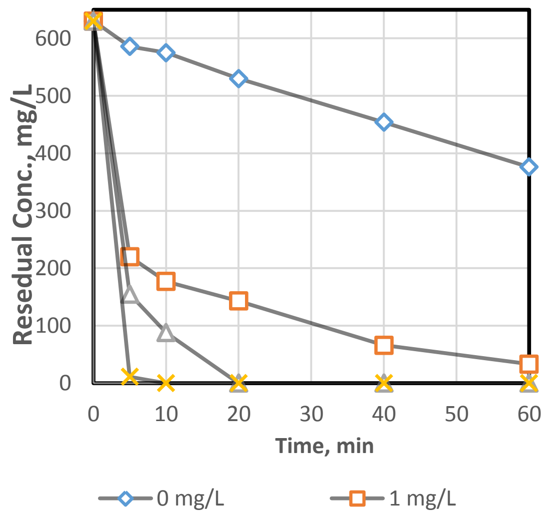

Fig. 8. Effect of supporting electrolyte (different concentrations of NaCl) on resedual concentrations of nitrate (Co= 630 mg/L, i = 10 mA/cm^2)

Complete removal (100 %) of nitrate was noted for the initial NO^{3-} concentrations of 630 mg/L, at the current densities of 10 mA/cm2 for supporting electrolyte concentrations of 29 and 10 mg/L (NaCl) after 10 and 20 min of EC treatment respectively. For the previous case total energy consumptions were 1.88 and 3.7 kWh/m^3 respectively.

4. CONCLUSIONS

Electrocoagulation was investigated for its performance in the removal of nitrate from wastewater using vertical cylindrical monopolar Fe-Fe electrodes in a batch electrocoagulation unit. The results show that, the nitrate removal efficiency has been increased by increasing the current density from 10 to 45 mA/cm^2, decreasing initial concentration, and by increasing supporting electrolyte concentration.

The removal of nitrate fits well with the first-order kinetic model at high current densities.

According to the results, EC using monopolar iron electrodes is a convenient route for effective and removal of nitrate from a model wastewater and achieves faster removal in comparison to chemical coagulation, where several hours are needed for adsorption on activated carbon. Using both EC and adsorption on activated carbon can be employed to comparison purposes for future studies.

Other parameters such as pH and effect of other water pollutants on the nitrate removal efficiency were not taken in this study.

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