

Nitrate Removal from Water Using Electrocoagulation with Activated Carbon

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Abstract: The present work studies the clarification of drinking water by removing Nitrate and Nitrite using electrocoagulation (EC) and the addition of activated carbon which produced from the almond peel (AP). The efficiency of coupling process studied by using different amounts of adsorbent material to identify the most appropriate dosage, for enhancing drinking water purification, The efficiency of the current density used in the removal of Nitrate and Nitrite was studied, energy consumption of the process was also determined. The coupling between electrocoagulation and the addition of the activated carbon showed that the addition of an appropriate dosage resulted in the enhancement of the removal rate of Nitrate and Nitrite especially at lower current densities, and resulted in a considerable reduction in the needed time compared to the conventional process that does not use activated carbon. The electrocoagulation with the addition of activated carbon achieved efficiency removal of approximately 58.5% for Nitrate and 98.9% for Nitrite. Based on the high removal efficiency, short contact time, and low energy consumption, the electrocoagulation with the addition of activated carbon method could be recommended instead of the conventional process. Furthermore, the results suggest that the productivity of the process is enhanced by adding a very small amount of NaCl as a supporting electrolyte.

Keywords: Electrocoagulation, Nitrate removal, Nitrite removal, adsorption, monopolar electrode configuration, activated carbon, tap water purification, supporting electrolyte.

إزالة النترات من المياه بالتخثير الكهربائي مع الكربون المنشط

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المستخلص: تتناول هذه الدراسة تنقية مياه الشرب من خلال إزالة النترات والنيتريت باستخدام تقنية تجمع بين التخثير الكهربائي (ت.ك) والكربون المنشط المصنع من قشور اللوز (ق.ل). تمت دراسة خواص هذه التقنية المزدوجة باستخدام كميات مختلفة من المادة المازة لتحديد الجرعة الأمثل، وذلك لتحسين كفاءة تنقية مياه الشرب الملوثة. كذلك تمت دراسة تأثير شدة التيار الكهربائي على كفاءة الإزالة واستهلاك الطاقة للمعالجة المزدوجة ت.ك/ق.ل. أظهرت نتائج المعالجة المزدوجة أنّ إضافة كمية مناسبة من الكربون المنشط ق.ل أدت إلى تحسين معدل الإزالة للنترات والنترات وخاصة عند استخدام تيار منخفض، وإلى انخفاض واضح في زمن المعالجة بالمقارنة مع عملية التخثير الكهربائي التقليدي وعملية التخثير البسيط التي لا تستخدم الكربون المنشط، حيث وصل معدل الإزالة إلى ما يقارب 58.5% للنترات، و98.9% للنيتريت. طريقة التنقية المزدوجة ت.ك/ق.ل قد تكون مفضلة على طريقة التخثير الكهربائي التقليدي، وذلك بالنظر إلى كفاءة الإزالة العالية، سرعة المعالجة، والاستهلاك المنخفض للطاقة. تشير النتائج إلى تحسن في كفاءة الإزالة عند إضافة كمية محددة من الداعم الإلكتروني.

الكلمات المفتاحية: التخثير الكهربائي، إزالة النترات، إزالة النتريت، امتزاز، مجموعة أحادية القطب، فحم منشط، تنقية مياه الصنبور، داعم إلكتروليتي.

1- Introduction

The main objective of the study is to investigate the influence of electrocoagulation (EC) process parameters using Iron (Fe) electrodes to remove Nitrates and Nitrites from tap water, and then studies the efficiency of the coupling of electrocoagulation and the adsorption on active carbon (AC).

1.1- Theoretical framework

Nitrate (NO_3^-) uses in many industrial technologies (such as industries of explosives, glasses, and inorganic fertilizers, etc.) leads to contamination of the water and the environment and threatens public health because of the hazardous properties of Nitrate. Nitrate can cause eutrophication problems and toxicity in water.

Nitrates might result in mutagenicity, teratogenicity, and birth defects, contribute to the risks of non-Hodgkin's lymphoma, bladder, ovarian cancers, which play a role in the etiology of insulin-dependent diabetes mellitus and the development of thyroid hypertrophy, or cause spontaneous abortions and respiratory tract infections.

Indirect health hazards can occur as a consequence of algal toxins, causing nausea, vomiting, diarrhea, pneumonia, gastroenteritis, hepatoenteritis, muscular cramps, and several shellfish poisoning syndromes. Other indirect health hazards can also come from the potential relationship between Nitrate and human infectious diseases such as malaria and cholera (Camargo and Alonso, 2006, Olawuyi, O. J. et al., 2020, Semitsoglou-Tsiapou, S. et al., 2018).

Nitrate formed by the biochemical activities of microorganisms or added in chemically synthesized forms to the lithosphere and biosphere enters hydrosphere with relative ease, as all these environmental components are dynamically interconnected. The high solubility of Nitrate in water and its low retention by soil particles make it a major component of groundwater in areas of high Nitrate formation. Nitrate enters the human body through the use of groundwater for drinking and causes several health disorders, namely, methemoglobinaemia, gastric cancer, goiter, birth malformations, hypertension, etc., when present in high concentration in drinking water (Majumdar et al., 2000, Olawuyi, O. J. et al., 2020, Semitsoglou-Tsiapou, S. et al., 2018).

The World Health Organization (WHO) has set the maximum allowable concentration of Nitrate in drinking water at 50 mg/l and 10 mg/l of Nitrite (measured as NO_3^- , NO_2^- respectively (Organization, 2004, Uzun, H. I. et al., 2019).

Nitrate is a stable and highly soluble ion with low potential for co-precipitation or adsorption. These properties make it complicated and expensive to be removed from the water.

Different technologies are used to remove Nitrate and Nitrite from water including ion-exchange (IX) (Nujić, M et al., 2017, Mendow, G., Grosso, C. I., Sánchez, A., & Querini, C. A., 2017, Primo et al., 2009), adsorption onto various materials (Ghadiri, S. et al, 2017, Hui, C. et al, 2018, Öztürk and Bektaş, 2004), electrodialysis (ED) (Belkada, F. et al., 2018, Albornoz, L. L. et al., 2019, Sahli et al., 2008), nanofiltration (NF) / reverse osmosis (RO) (Jadhav, S. V. et al., 2016, Yousefi, N. et al., 2016, Richards et al., 2010), biological denitrification (Zhang, Z. et al., 2018, Cao, S., & Zhou, Y., 2019, Wang et al., 2009), chemical reduction (Guo, J. et al., 2018, Seraj, S. et al., 2017, Suzuki et al., 2012) and electrochemical reduction (Zhang, Y. et al., 2018, Dreyse et al., 2011, P., Li et al., 2009).

All membrane-based technologies and ion exchange cannot decompose Nitrate. However, it creates secondary wastes. Ion exchange requires high maintenance of the media. Biological denitrification is opposed because it also needs intensive maintenance, constant supply of organic substrates, and it requires a biomass sludge disposal and and it keeps biological traces in treated water. Microbial processes are time-consuming process and sometimes it is practically difficult to incomplete compared with chemical reduction. (Ait-Ahmed, N. et al., 2017, Zhang et al., 2005).

Compared with the chemical reduction of Nitrate, the electrochemical method might be the most favorable one. Among the electrochemical methods, electrocoagulation (EC) is suggested to be a practical choice for both economic and environmental aspects. EC process is known to be simple, reliable, and cost-effective method for industrial wastewater and other related water management issues without the need for additional chemicals, and thus, without the production of secondary pollution (Chen, 2004, Darban, A. K. et al., 2020).

EC is an efficient technique since adsorption of hydroxide on mineral surfaces is 100 times better than on precipitated hydroxides when metal hydroxides are used as a coagulant. It requires low maintenance with no moving parts. EC needs low current densities, and it can be run even by green technologies. EC produces less sludge. Gas bubbles produced during the process can enhance flotation (which is good for the sludge removal. Furthermore, its electrochemical-based systems suits the automation, allows using smaller systems, and instead of using chemicals, the systems employ only electrons for water treatment (Darban, A. K. et al., 2020, Zaroual et al., 2009, Zaied and Bellakhal, 2009).

In recent years, there is much interest in using EC for the treatment of industrial wastewater containing, phosphate (Hashim, K. S. et al., 2019, Vasudevan et al., 2008), fluoride (Drouiche et al., 2012, López-Guzmán, M. et al., 2019), boron (Chen, M. et al., 2020, Isa et al., 2014), sulfide, sulfate and sulfite ions (Murugananthan et al., 2004, Soltanian, M. et al., 2016), humic acids (Barhoumi, A. et al., 2019, Ghernaout et al., 2014), and heavy metals (Al Aji et al., 2012, El-Ashtoukhy, E. et al., 2020).

EC was investigated by various workers for Nitrate removal (Kumar and Goel, 2010, El-Shazly et al., 2011, Vasudevan et al., 2010, Ghanim and Ajjam, 2013, Hossini and Rezaee, 2014, Sharma and Sobti,

2012), and Nitrite removal (Nazlabadi, E. et al., 2019, Amarine, M. et al., 2020). These studies show the successful removal of Nitrate and Nitrite from polluted water.

Currently, there are many studies on the development of low-cost adsorbents, namely by using waste materials for that purpose. Also, several reviews report a great deal of work done on their application for the removal of specific pollutants from the aqueous phases, such as Nitrate (Bhatnagar and Sillanpää, 2011, Demiral and Gündüzoğlu, 2010, Sulyman, M. et al., 2017).

Several suitable by-products agricultural including olive cakes, olive stone, date stone, cherry stones, tobacco stems, rice husk, rice straw, rice bran, wheat straw, grape seeds, cotton stalks, corn cob, waste tea, waste apricot, sawdust, sugar canes, mango pit, durian shell, herb residues, coffee husks, almond shell, and almond peel have been investigated in the last years as AC precursors and are still receiving good attention (Danish, M. et al., 2018, Dias et al., 2007).

The addition of activated carbon as an adsorbent to the process resulted in a fast increase in the removal efficiency especially at lower current densities, and resulted in minimizing the operating time compared with the conventional electrocoagulation process (de Carvalho, H. P. et al., 2015, Secula, M. S. et al., 2012).

There are two procedures to use activated carbon, as a coupling process (simultaneously with the electrocoagulation) or as a separated process (after the electrocoagulation). When activated carbon adsorption is applied as a separated step to complete the treatment, the acute toxicity decreased (GilPavas, E. 2019), another paper (Barhoumi, A. et al., 2019) showed that due to large concentrations of Humic acid (HA), the cellulose-based industries produce large volumes of wastewater which are difficult to treat. In subsurface water, HA reacted with active chlorine to produce carcinogenic compounds. Carcinogenic compounds causing different ecological problems and danger to human health.

It has been observed from the experimental results (Barhoumi, A. et al., 2019) that combination of electrocoagulation with adsorption on GAC notably reduces the treatment duration without inducing a strong increase in cost. The current study extends the investigation of these promising processes. The results suggest that combining adsorption onto AC with EC could greatly improve the economics of treatment of wastewaters.

In this present study, the adsorption process using low-cost adsorbents (almond peel - AP) has been coupled with electrocoagulation for Nitrate removal enhancing to avoid the operational difficulties and high cost of electrocoagulation process for large-scale treatment of tap water rich in Nitrate or Nitrite. The effect of initial Nitrate or Nitrite concentration, current density, treatment time, initial pH, and adsorbent concentration was explored and discussed on the removal efficiency to determine the optimum operational conditions.

2.0 - Materials and Methods

2.1 - Activation of Almond Peel (AP)

Almond peel has been converted to an effective adsorption substance of alternative activated carbon by physical and chemical activations. After washing and drying the almond peel was undergone thermal activation by choosing the optimum temperature ($800\text{ }^{\circ}\text{C}$) and the optimum activation time (2hrs) to obtain a high BET surface area. The optimum normalities for chemical activation of the previous sample were chosen ($n=0.5$) for HCl and ($n=0.3$) for H_2SO_4 to obtain the highest BET surface area which became $517.97\text{ m}^2/\text{g}$.

2.2 - Electrocoagulation Experiments

In this step, the removal of Nitrate and Nitrite ions from aqueous solutions by EC was investigated using AP as a supporting electrolyte. For this purpose, potassium Nitrate (HIMEDIA Extra pure) was used to prepare solutions with the concentration of (800 mg/l NO_3^-), and sodium Nitrite was used to prepare solutions with the concentration of (200 mg/l NO_2^-). (0.0 to 2.0) g of AP was added to the different initial concentrations of Nitrate and Nitrite ($800\text{-}200\text{ mg/l}$ respectively) and were shaken for 5 minutes at 150 rpm.

To demonstrate the effect of varying operating parameters, a stock solution of 1.0 litre containing a determined concentration of Nitrate or Nitrite was prepared with potassium Nitrate (HIMEDIA Extra pure) and sodium Nitrite. Stock solutions of Nitrate were made by adding the specific values of potassium Nitrate (KNO_3) in tap water, also stock solutions of Nitrite were made by adding the specific values of sodium Nitrite (NaNO_2) in tap water to get (800 mg/l NO_3^- and 200 mg/l NO_2^-). The model tap water concentration was prepared from the stock solution by suitable dilution. After taking a sufficient number of samples from clear parts of solutions, the equilibrium concentrations of Nitrate and Nitrite were determined. The removed amount was calculated from the difference between initial and equilibrium concentrations. The residual concentrations of Nitrate were determined using a DR Spectrophotometer (DR-890 Colorimeter Hach, USA) see Figure (1), according to standard methods. Nitrate and Nitrite experiments were carried out at laboratory temperature and an original pH of Approximately 7.0.

Experiments were made by using a cylindrical glass cell of 1000 mL on a magnetic stirrer (Nuova II stir plate, USA), see Figure (3). 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 electrodes with serial connections mode in the experiments. The inter-electrode distance was 0.4 cm. A DC-power supply (RXN-605D, China), see Figure (2), characterized by the ranges of 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 Figure (4).



Figure (1) - DR-890 Colorimeter Hach, USA



Figure (2) - RXN-605D, China



Figure (3) - Nuova II stir plate USA

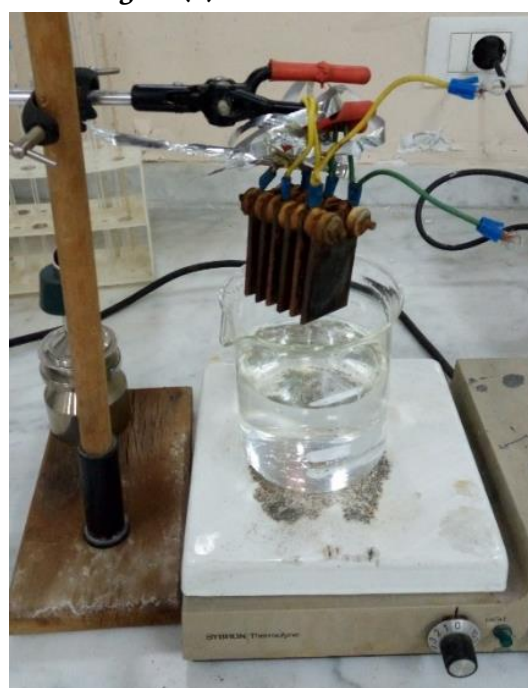


Figure (4) - Bench-scale EC reactor with monopolar electrodes in serial connection

Table (1) - Nitrate Experiments

Nitrate (800 mg/l NO ₃ ⁻)			
1	1.0 Ampere	0.0 (g/l) AC	0.0 (g/l) NaCl
2	3.0 Ampere	0.0 (g/l) AC	0.0 (g/l) NaCl
3	5.0 Ampere	0.0 (g/l) AC	0.0 (g/l) NaCl
4	1.0 Ampere	0.5 (g/l) AC	0.0 (g/l) NaCl
5	1.0 Ampere	1.0 (g/l) AC	0.0 (g/l) NaCl
6	1.0 Ampere	2.0 (g/l) AC	0.0 (g/l) NaCl
7	3.0 Ampere	0.5 (g/l) AC	0.0 (g/l) NaCl
8	3.0 Ampere	1.0 (g/l) AC	0.0 (g/l) NaCl

Nitrate (800 mg/l NO ₃ ⁻)			
9	3.0 Ampere	2.0 (g/l) AC	0.0 (g/l) NaCl
10	5.0 Ampere	0.5 (g/l) AC	0.0 (g/l) NaCl
11	5.0 Ampere	1.0 (g/l) AC	0.0 (g/l) NaCl
12	5.0 Ampere	2.0 (g/l) AC	0.0 (g/l) NaCl
13	5.0 Ampere	2.0 (g/l) AC	0.5 (g/l) NaCl
14	5.0 Ampere	2.0 (g/l) AC	2.0 (g/l) NaCl

Table (2) - Nitrite Experiments

Nitrite (200 mg/l NO ₂ ⁻)			
1	0.5 Ampere	0.0 (g/l) AC	0.0 (g/l) NaCl
2	1.0 Ampere	0.0 (g/l) AC	0.0 (g/l) NaCl
3	2.0 Ampere	0.0 (g/l) AC	0.0 (g/l) NaCl
4	0.5 Ampere	0.5 (g/l) AC	0.0 (g/l) NaCl
5	0.5 Ampere	1.0 (g/l) AC	0.0 (g/l) NaCl
6	0.5 Ampere	2.0 (g/l) AC	0.0 (g/l) NaCl
7	1.0 Ampere	0.5 (g/l) AC	0.0 (g/l) NaCl
8	1.0 Ampere	1.0 (g/l) AC	0.0 (g/l) NaCl
9	1.0 Ampere	2.0 (g/l) AC	0.0 (g/l) NaCl
10	2.0 Ampere	0.5 (g/l) AC	0.0 (g/l) NaCl
11	2.0 Ampere	1.0 (g/l) AC	0.0 (g/l) NaCl
12	2.0 Ampere	2.0 (g/l) AC	0.0 (g/l) NaCl
13	2.0 Ampere	2.0 (g/l) AC	0.5 (g/l) NaCl
14	2.0 Ampere	2.0 (g/l) AC	1.0 (g/l) NaCl

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, 20, 30, and 50 mA/cm². Another series of experiments were carried out at different dosage of AC 0.5, 1.0, and 2.0 g/l and supporting electrolytes of 0, 0.5, 1.0, and 2.0 g/l NaCl to investigate the effect of supporting electrolyte on the removal yield, see Table (1) and Table (2).

All the runs were performed at laboratory temperature and the stirring speed of 100 rpm.

The charge loading (Q_e) and energy consumption (E_{con}) per gram of molecule were calculated using Eq. (1) and Eq. (2).

$$Q_e = I * t_{EC} C_o \quad \text{Eq. (1)}$$

$$E_{con} = U * I * t_{EC} / (C_o * \mu) \quad \text{Eq. (2)}$$

Where:

Q_e	charge loading per gram of dye (A.h/g)
E_{con}	specific electrical energy consumption per gram of dye (W.h/g)
C_o	initial dye concentration (g)
I	current (A)
U	cell voltage (V)
t_{EC}	electrolysis time (h)
μ	purification ratio (%)

The generation of the coagulant during the EC process leads to the consumption of electrode material that can be estimated based on Faraday's law as follows.

$$UEMD = \frac{ItA}{nFVC_oY_t} / 100 \quad Eq. (3)$$

Where:

$UEMD$	unit electrode material demand, (kg/kg)
E_{con}	specific electrical energy consumption per gram of dye (W.h/g)
t	time, (s)
n	number of electrons involved in oxidation/reduction reaction
F	Faraday's constant, (C/mol)
A	the atomic mass of electrode material, (g/mol)
I	current intensity
V	the volume of treated solution, (m ³)
Y_t	Nitrate removal efficiency at time t, (%)

3.0 - Results and Discussion

The present work studies the clarification of drinking water by removing Nitrate and Nitrite using electrocoagulation (EC) and the addition of activated carbon which produced from the almond peel (AP).

Nitrate is the hazardous contaminants in water that convert the hemoglobin into methemoglobin, which does not carry oxygen to body tissues. This phenomenon results in a bluish color of infants' skin that is called methemoglobinemia or the blue baby syndrome. Nitrate may be also cause gastric cancer and tumors (Gupta et al., 2000, Schullehner, J. et al., 2018, Temkin, A. et al., 2019, Yang and Lee, 2005)

Researches showed a high Nitrate concentration in groundwater in some middle-east countries including Syria. Nitrate salts percolate through the soil to finally reach the ground water. Due to the extreme use of fertilizers, Nitrate is increasingly growing all over the world. Other sources of Nitrate in

water are animal farms and incompletely treated wastewater. Another source of Nitrate is the mineralization of nitrogen-organic compounds in agricultural drainage.

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 and Nitrite from tap water as studied and the results are shown in the figures from Figure (7) to Figure (14). According to these results, it is evident that increasing the current density causes an increase in Nitrate and Nitrite removal.

For the initial concentration of 800 mg/l of Nitrate (NO_3^-), the removal efficiency values were 25.0, 28.6, and 47.4% at the current densities of 10, 30, and 50 mA/cm^2 , respectively after 60 min EC treatment with no addition for activated carbon (AC), and were 10.5, 29.2, and approximately 50% at the current densities of 10, 30, and 50 mA/cm^2 , respectively after 60 min EC treatment with the addition of 0.5 g/l of AC, and were 33.3, 38.1 and 53.7% at the current densities of 10, 30, and 50 mA/cm^2 , respectively after 60 min EC treatment with the addition of 1.0 g/l of AC, and were 42.9, 46.7 and 63.4% at the current densities of 10, 30, and 50 mA/cm^2 , respectively after 60 min EC treatment with the addition of 2.0 g/l of AC, see Figure (15).

For the initial concentration of 200 mg/l of Nitrite (NO_2^-), the removal efficiency values were 82.1, 90.5 and 98.4% at the current densities of 5, 10, and 20 mA/cm^2 , respectively after 60 min EC treatment with no addition for activated carbon (AC), and were 88.0, 91.5, and 94.6% at the current densities of 5, 10, and 20 mA/cm^2 , respectively after 60 min EC treatment with the addition of 0.5 g/l of AC, and were 84.3, 89.4 and 96.8% at the current densities of 5, 10, and 20 mA/cm^2 , respectively after 60 min EC treatment with the addition of 1.0 g/l of AC, and were 85.2, 88.7 and 95.4% at the current densities of 5, 10, and 20 mA/cm^2 , respectively after 60 min EC treatment with the addition of 2.0 g/l of AC, see Figure (16).

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 $\text{Fe}(\text{OH})_3$ coagulant, See Figure (7) to Figure (14). also show that increasing the electrolysis time has increased the NO_3^- and NO_2^- removal which can be attributed to the fact that increasing the electrolysis time will certainly increase the available Fe_3^+ according to Faraday's law, and consequently, increase the amount of $\text{Fe}(\text{OH})_3$ which is responsible for NO_3^- and NO_2^- removal, see Figure (5) and Figure (6).

3.2 - Effect of NaCl as 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 decrease the energy

consumption. Adding a particular concentration of NaCl enhances the result of purification. However, an overdosage of NaCl effects the purification negatively, see Figure (17) and Figure (18). It is obvious that adding 0.5 g/l of NaCl improves the purification more than adding 2.0 g/l of NaCl, this is attributed to the fact that adding more NaCl to the solution increases its contents of Na^+ and Cl^- ions, this increase boosts the electronic permeability of the solution which growths the amount of the coagulants, which causes a blocking layer of coagulant between the cathodes and the anodes, which leads to a current value drop and a purification rate decline, another reason is that addition in general (whether it is activated carbon or NaCl) causes three changes in term of the nature of the sludge produces, in other words, the sludge will be (1) less adsorptive, (2) more prolific, and (3) more settleable; the first change will give a negative effect on the purification, however, the second one will enhance the purification, and the third does not affect the treatment but effects the sludge treatment, and the overall effect will be determined by dominant of first and second changes, in Figure (19), we can see that adding no NaCl gives the best results for the first 30 minutes, and after that, the same curve shows lower values compared with the case of adding NaCl, that can be explained by the fact that, in the first 30 minutes, the amount of flocks produces is similar for the three cases (0, 0.5, and 2.0 g/l of NaCl), so the adsorbability of the flock will be the dominant factor, however, after 30 minutes of treatment, the amount of the flocks dominates.

3.3 - Effect of activated carbon AC:

The addition of AC gives a positive effect on the removal ratio of Nitrate, especially for low current densities, see Figure (15), however, an atypical behavior occurs when adding 2.0 g/l of AC while using a current density of 50.0 mA/cm^2 , that can be explained by the fact that a solution treated with a high value of current density (50.0 mA/cm^2) would have a sufficient amount of flocks, so when adding AC then, a reduction in the adsorption quality of the flocks occurs, making the productivity of the purification drops.

For the Nitrite, an amount of 0.5 g/l of AC, has a great impact on enhancing the productivity of the treatment when using a low current density of 5.0 mA/cm^2 , moreover, the same amount worsens the process for high current densities like 20 mA/cm^2 (which consider high in Nitrite removal), this behavior can be clarified if we noticed that adding AC will enhance the mass of flocks produced (which causes a positive effect on the treatment), and decrease the adsorbability of the flocks (which harms the treatment), and when the current density is low, the mass of flocks will not be sufficient, then adding AC will improve the process, on the other hand, when the current density is high, then adding the AC will harm the process, see Figure (16).

3.4 - Effect of additions and time:

3.4.1 Additions (AC, NaCl) and current density

Additions and current densities make a notable difference in treating Nitrate, however, the difference it makes on the treatment of Nitrite is insignificant, that can be attributed to the fact that Nitrate

removal consumes fewer electrons than Nitrate removal, so a low current density is enough for Nitrite, and any increase besides and/or current density will not help much, see Figure (7) to Figure (14), and Figure (15) to Figure (19).

3.4.2 Time

It is clear that time has no great effect on the treatment of Nitrate after half an hour, while it has a good effect for 60 minutes of treatment for Nitrite, see Figure (5) and Figure (6), accordingly, a long treatment (more than 60 minutes) is not justified for treating Nitrate in normal conditions, however, it is for treating Nitrite.

3.5 Energy Consumption E_{con}

In this study, EC/AP coupling processes were tested to determine the values of rate constant and various experimental conditions. Based on the rate constant values and energy consumption (E_{con}), optimization of the coupling process by adding different amounts of AP and NaCl was evaluated. The influence of current density on the enhancement of removal efficiency and E_{con} of the EC/AP coupling process was established.

Energy consumption can be cut by adding a lower dose of AC (0.5 g/l) for treatment with low current density (10 to 30 mA/cm²) for Nitrate, see Figure (20), simultaneously, an increase in the purification ratio occurs, this enhancement occurs in smaller rates for bigger dosages of AC, see Table (3). However, Nitrate removal ratio shows a drop by increasing the dosage of AC at high current density (50 mA/cm²). On the other hand, Nitrite purification shows a significant drop in energy consumption for high current density (20 mA/cm²) by adding 0.5 g/l of AC, simultaneously, the ratio of the purification drops slightly, see Figure (21). 59.5% of Nitrate removal could be achieved by a process with an energy consumption of 68.54 W.h/g, 98.9% of Nitrite removal could be achieved by a process with an energy consumption of 2.75 W.h/g, see Table (3) and (خطأ! لم يتم العثور على مصدر المرجع). Besides, adding a dose of NaCl to the solution may decrease the consumption, see Figure (22) and Figure (23).

4.0 - Conclusion

Electrocoagulation was investigated for its performance in the removal of Nitrate and Nitrite from tap water using vertical 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 50 mA/cm² for Nitrate and from 10 to 30 mA/cm² for Nitrite, and by increasing AC concentration (as a supporting electrolyte).

In the present work, the effects of Nitrate and Nitrite removal by conventional EC in comparison with the EC/AP coupling process were discussed. It was found that the removal of Nitrate and Nitrite by using simple EC monopolar Fe-Fe electrodes in a batch electrocoagulation unit with the addition of AP

activated carbon are feasible processes. The addition of AP as an adsorbent resulted in a rapid increase in the removal efficiency of Nitrate and Nitrite at lower current densities and operating times compared with the conventional electrocoagulation process. The success of this coupling process is linked to the specific chemical and textural properties of the material. The removal rate and energy consumption were found to increase when the current density was increased from 10 to 50 mA/cm² for Nitrate and from 10 to 30 mA/cm² for Nitrite. Increasing the dosage of AP from 0.0 to 2.0 g/l improved the kinetics of the coupling process rapidly for Nitrate for all current densities examined, and for Nitrite only for lower current densities. Furthermore, in terms of energy consumption and removal efficiency a dosage of 0.5 g/l is optimal (except for Nitrate purification at high current densities like 50 mA/cm²), and further increases resulting in an insignificant purification ratio increase and insignificant reductions in energy consumption, affecting negatively the efficiency of the EC/AP coupling process. Best results can be achieved when using 50 mA/cm², 0.0 g/l AC, and 0.0 g/l NaCl for Nitrate, and 20 mA/cm², 0.0 to 0.5 g/l AC, and 0.0 g/l NaCl for Nitrite; the purification ratio will be 47.4% for Nitrate, and up to 98.8% for Nitrite, with E_{con} of 56.80 W.h/g for Nitrate, and below 10 W.h/g for Nitrite, see Table (3) and (خطأ! لم يتم العثور على مصدر المرجع). It was shown that the EC/AP coupling technique for Nitrate and Nitrite removal is considerably cheap, and is suitable for developed and developing countries. According to the results, EC using monopolar iron electrodes is a convenient route for effective removal of Nitrate and Nitrite from a model tap water 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 for comparison purposes for future studies. Other parameters such as the pH and the effect of other water pollutants on the Nitrate and Nitrite removal efficiency were not taken in this study.

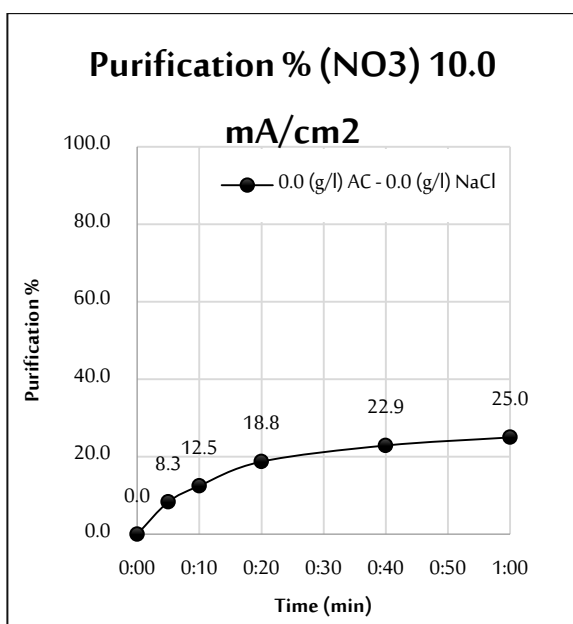


Figure (5) - purification for NO₃ during 60 min of treatment with no addition of NaCl and AC

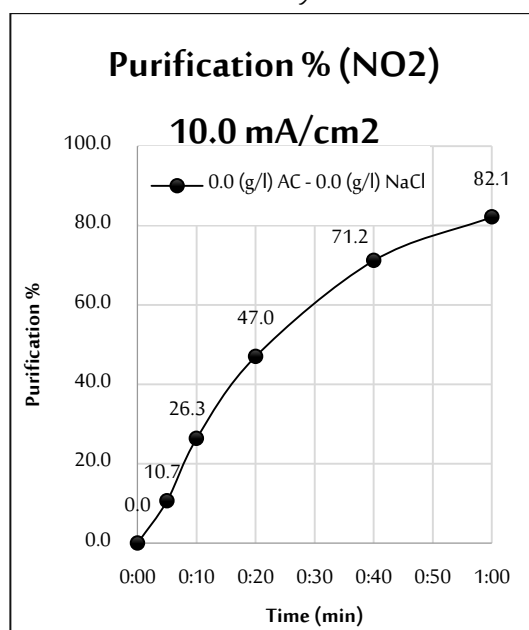


Figure (6) - purification for NO₂ during 60 min of treatment with no addition of NaCl and AC

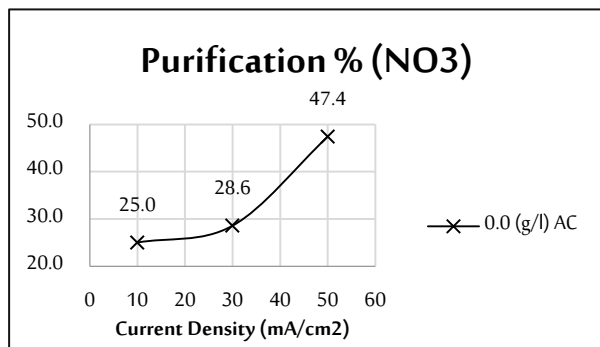


Figure (7) - Adsorption isotherm for NO₃ on 0.0 (g/l) AC

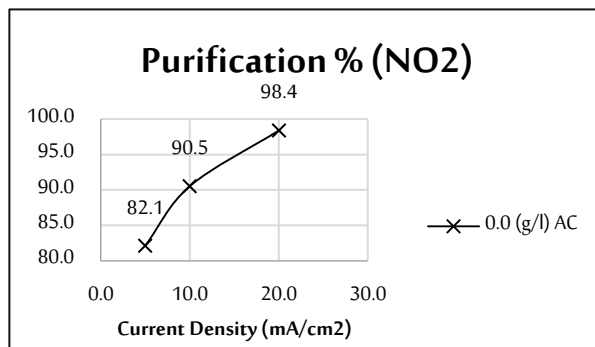


Figure (8) - Adsorption isotherm for NO₂ on 0.0 (g/l) AC

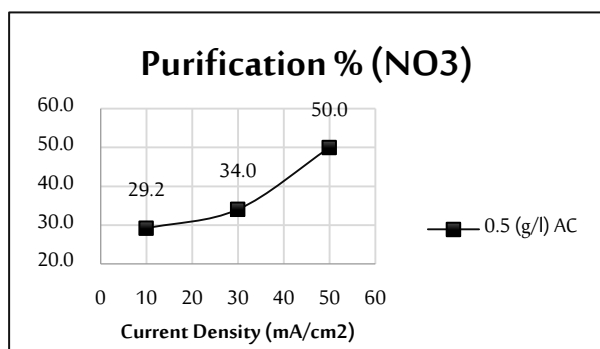


Figure (9) - Adsorption isotherm for NO₃ on 0.5 (g/l) AC

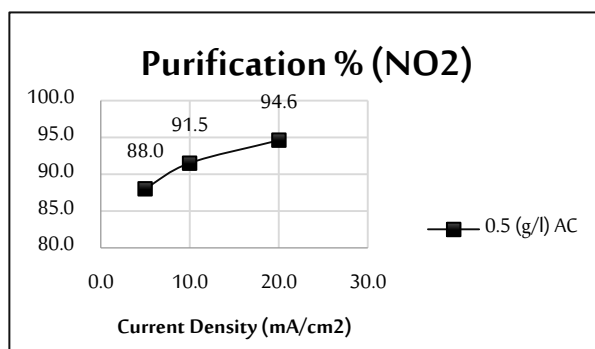


Figure (10) - Adsorption isotherm for NO₂ on 0.5 (g/l) AC

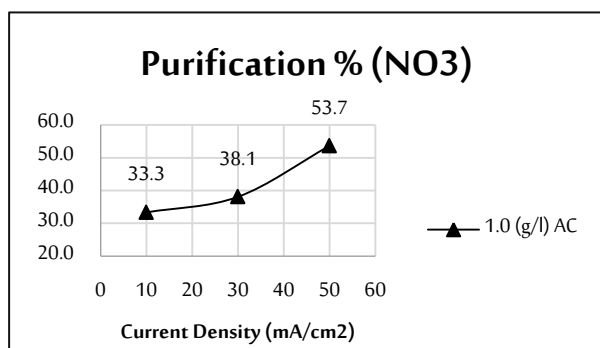


Figure (11) - Adsorption isotherm for NO₃ on 1.0 (g/l) AC

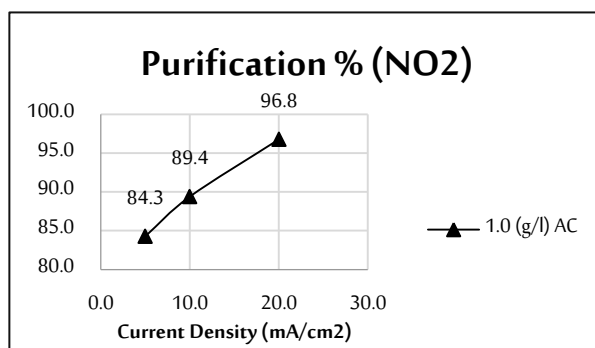


Figure (12) - Adsorption isotherm for NO₂ on 1.0 (g/l) AC

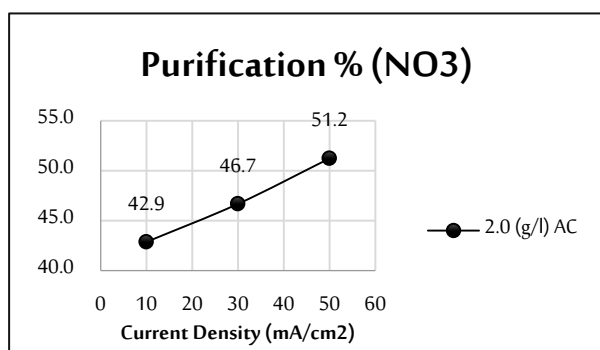


Figure (13) - Adsorption isotherm for NO₃ on 2.0 (g/l) AC

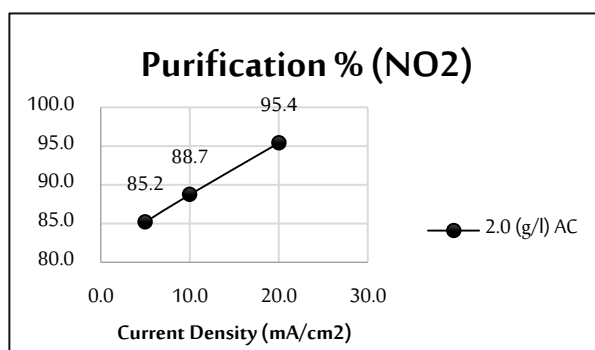


Figure (14) - Adsorption isotherm for NO₂ on 2.0 (g/l) AC

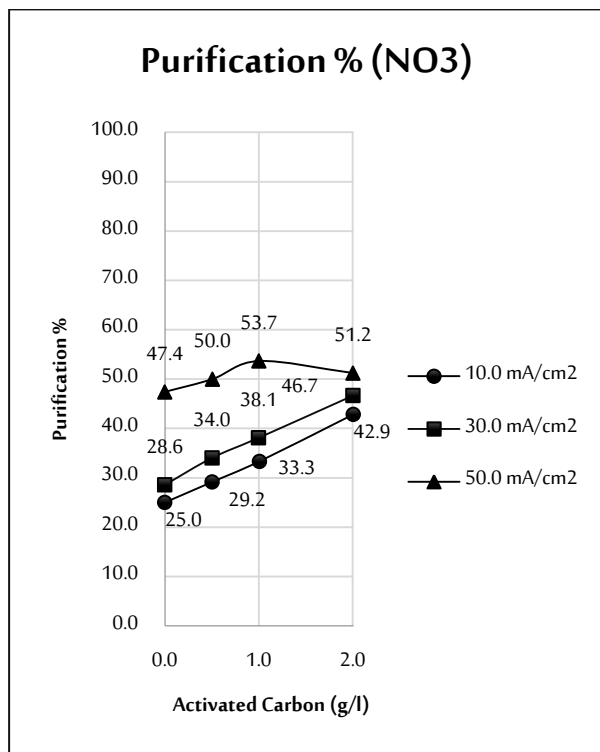


Figure (15) - purification for NO₃ for 60 min of treatment with the addition of AC

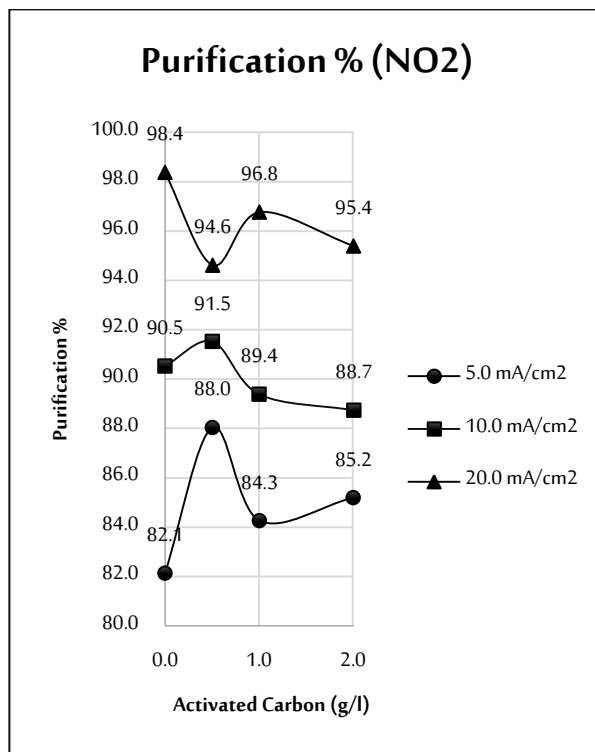


Figure (16) - purification for NO₂ for 60 min of treatment with the addition of AC

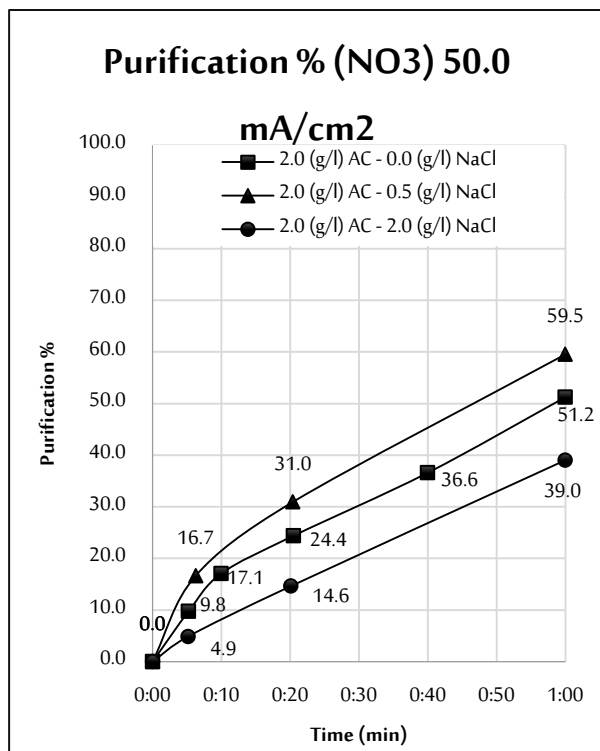


Figure (17) - purification for NO₂ for 60 min of treatment with the addition of NaCl and AC

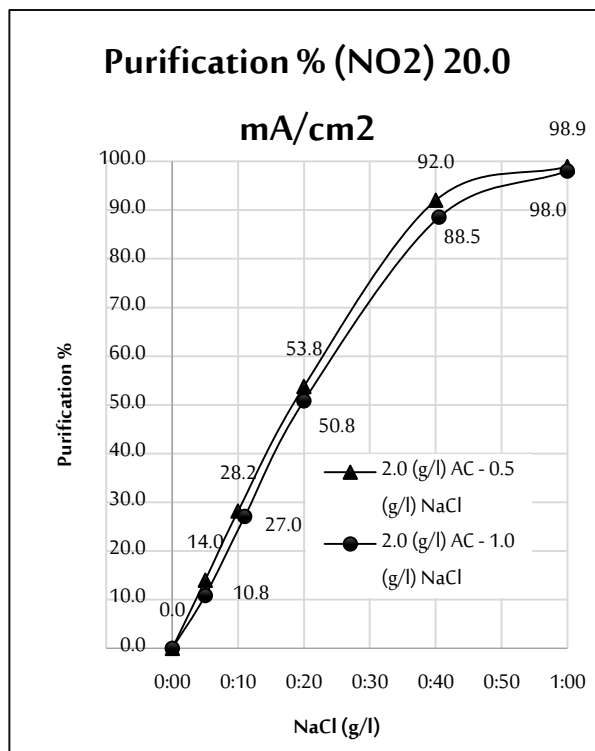


Figure (18) - purification for NO₂ for 60 min of treatment with the addition of NaCl and AC

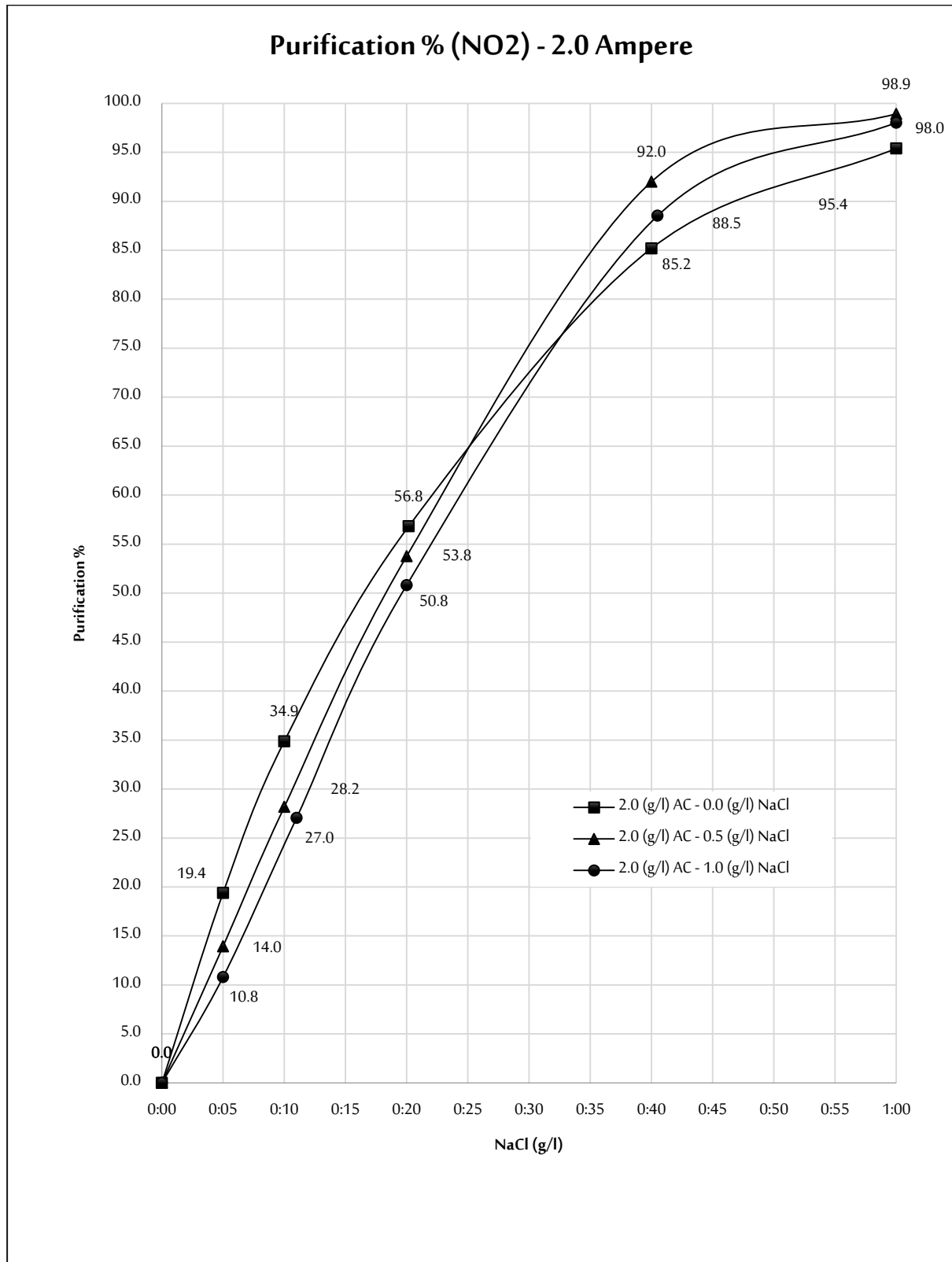


Figure (19) - purification for NO₂ for 60 min of treatment with the addition of NaCl and AC, considering the no-addition case

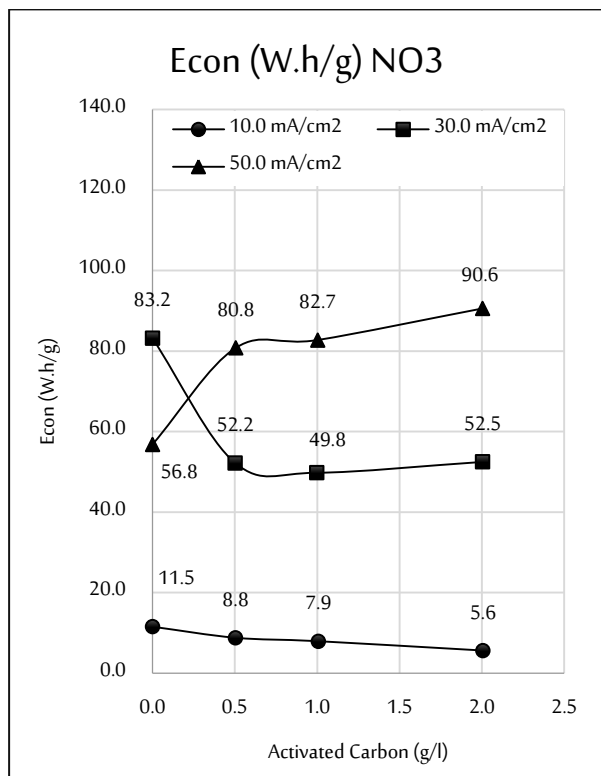


Figure (20) - energy consumption for NO₃ after 60 min of treatment

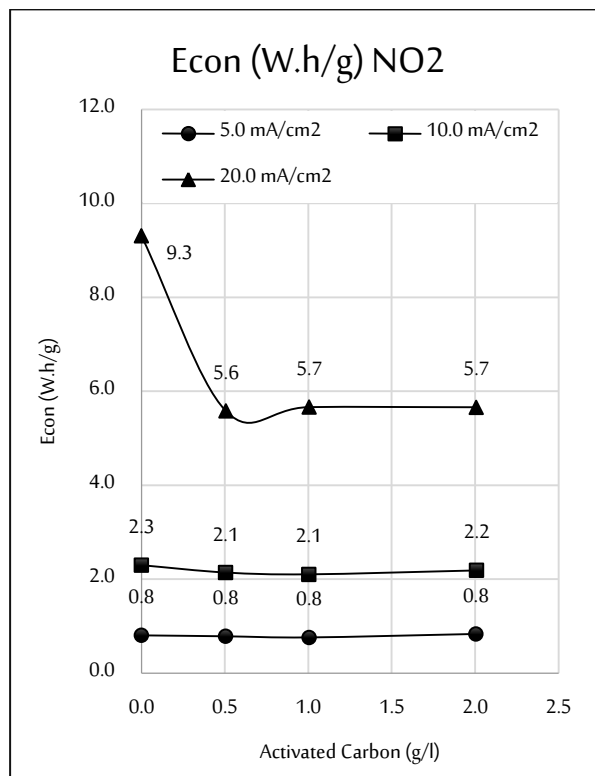


Figure (21) - energy consumption for NO₂ after 60 min of treatment

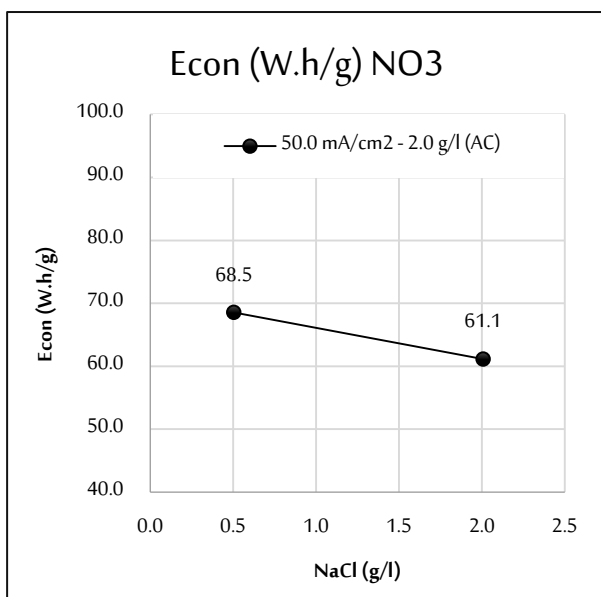


Figure (22) - energy consumption for NO₃ after 60 min of treatment with the addition of AC and NaCl

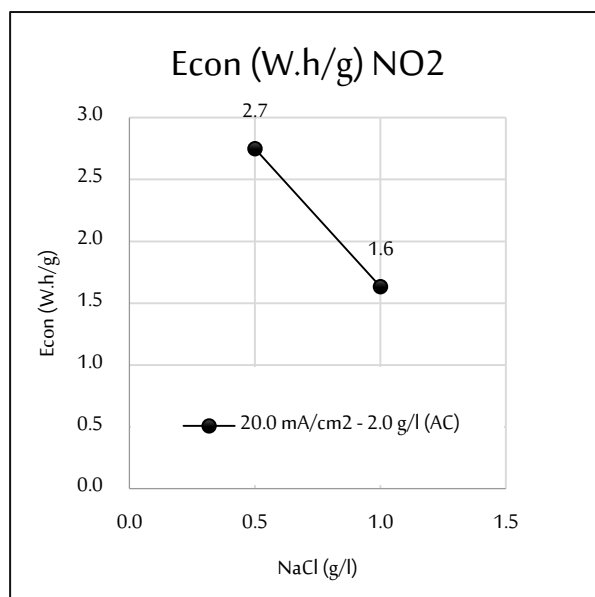


Figure (23) - energy consumption for NO₂ after 60 min of treatment with the addition of AC and NaCl

Table (3) - energy consumption of an 800 mg/l solution of NO_3^-

Current Density (mA/cm^2)	NaCl (g/l)	AC (g/l)	purification %	Econ (W.h/g)
10.0	0.0	0.0	25.0	11.52
10.0	0.0	0.5	29.2	8.78
10.0	0.0	1.0	33.3	7.92
10.0	0.0	2.0	42.9	5.60
30.0	0.0	0.0	28.6	83.16
30.0	0.0	0.5	34.0	52.17
30.0	0.0	1.0	38.1	49.77
30.0	0.0	2.0	46.7	52.46
50.0	0.0	0.0	47.4	56.80
50.0	0.0	0.5	50.0	80.80
50.0	0.0	1.0	53.7	82.74
50.0	0.0	2.0	51.2	90.58
50.0	0.5	2.0	59.5	68.54
50.0	2.0	2.0	39.0	61.14

Table (4) - energy consumption of a 200 mg/l solution of NO_2^-

Current Density (mA/cm^2)	NaCl (g/l)	AC (g/l)	purification %	Econ (W.h/g)
5.0	0.0	0.0	82.1	0.80
5.0	0.0	0.5	88.0	0.78
5.0	0.0	1.0	84.3	0.76
5.0	0.0	2.0	85.2	0.83
10.0	0.0	0.0	90.5	2.30
10.0	0.0	0.5	91.5	2.14
10.0	0.0	1.0	89.4	2.10
10.0	0.0	2.0	88.7	2.19
20.0	0.0	0.0	98.4	9.31
20.0	0.0	0.5	94.6	5.58
20.0	0.0	1.0	96.8	5.66
20.0	0.0	2.0	95.4	5.66
20.0	0.5	2.0	98.9	2.75
20.0	1.0	2.0	98.0	1.63

Multiple Linear regression of E_{con}

Nitrate - Multiple Linear Regression

$$\text{Econ (W.h/g)} = -7.7665 + 0.3215 \cdot \text{Current Density (mA/cm}^2) - 4.6695 \cdot \text{NaCl (g/l)} - 0.5236 \cdot \text{AC (g/l)} + 0.0842 \cdot \text{purification \%}$$

Model: $\text{Econ (W.h/g)} = -7.7665 + 0.3215 \cdot \text{Current Density (mA/cm}^2) - 4.6695 \cdot \text{NaCl (g/l)} - 0.5236 \cdot \text{AC (g/l)} + 0.0842 \cdot \text{purification \%}$

R-Squared: $r^2 = 0.8843$

Nitrite - Multiple Linear Regression

$$\text{Econ (W.h/g)} = 47.1224 + 2.8889 \cdot \text{Current Density (mA/cm}^2) - 32.8494 \cdot \text{NaCl (g/l)} + 17.2585 \cdot \text{AC (g/l)} - 2.497 \cdot \text{purification \%}$$

Model: $\text{Econ (W.h/g)} = 47.1224 + 2.8889 \cdot \text{Current Density (mA/cm}^2) - 32.8494 \cdot \text{NaCl (g/l)} + 17.2585 \cdot \text{AC (g/l)} - 2.497 \cdot \text{purification \%}$

R-Squared: $r^2 = 0.9179$

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