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Ultrasonic-Electrocoagulation method for nitrate removal from water

Saeed Al-Marri^{1,*}, Saif Salah AlQuzweeni², Khalid S. Hashim^{2,3}, Rafid AlKhaddar³, Patryk Kot³, Rasha Salah AlKizwini², Salah L. Zubaidi⁴, Zainab S. Al-Khafaji⁴

¹BSc student, Civil Engineering Department, Liverpool John Moores University, UK.

²Faculty of Engineering, University of Babylon, POBOX 4, Iraq.

³Department of Civil Engineering, Liverpool John Moores University, UK.

⁴Faculty of Engineering, University of Wasit, Wasit, Iraq.

⁵Department of Civil Engineering\Al-Mustaqbal University College\Babylon\Iraq.

*Email: S.H.AIMarri@2016.ljmu.ac.uk

Abstract. Water contamination with nitrates is a serious problem due to the detrimental effects of nitrates on both human life and the global ecosystem; therefore, it is essential to remove nitrates using efficient methods. Accordingly, various methods have been used to treat nitrate-containing solutions, but recent studies focused on electrocoagulation (ELE) as it produces high quality water at low cost and it is environmentally friendly. However, passive layers are growing on the aluminum anodes after short time of treatment, which substantially affects the efficiency of ELE. In this investigation therefore, ultrasonic filed was used to remove these passive layers, and consequently improves the efficiency of ELE. This new method, ultrasonic-assisted ELE (U-ELE), was used to remove nitrates from water under various operational conditions. In particular, the impacts of water initial pH (WIP) (4.0-8.0), applied current densities (ACD) (6.0-9.0 mA/cm²), flow rates (FR) (60-100 ml/min), and initial nitrate concentrations (INC) (100-200 mg/L), which were optimized using the Central Composite Design (CCD). The ultrasonic irradiation time (UT) has been kept at 10 minutes for all experiments. The best possible removal of nitrate using only ELE method was about 77% at WIP of 6, UT of 10 minutes, FR of 40 ml/min, INC of 150 mg/l and ACD of 7.5mA/cm². However, it was found that exerting ultrasonic for 10 minutes, U-ELE method, has increased nitrates removal to 87.80% under the same conditions of ELE treatment.

1. Introduction

In the Earth planet ecosphere that consists of ground and water, water is the most important part due to its importance in maintaining all forms of life, such as humans, animals and plants. This element of environment is the most abundant item in the Planet of Earth; water covers 71 percent of the Earth 's surface [1, 2]. Nevertheless, millions of people have no clean potable water due to the lack of the availability of freshwater, which represents not more than 2.5 percent of the total quantity of water [3-7]. In addition, other reasons, namely the rapid upsurge in the world population, the intensification of agricultural activities (to achieve the rapid production of foodstuffs) and the expansion of industries, were led to a dramatic rise in wastewaters generation and demands for freshwater, which limited the availability of freshwater [8-13]. This led to massive over-exploitation and massive depletion of the accessible water sources. Increasing contamination of groundwater and surface water, of drastic estimates, has prompted the concentration of toxic contaminants, like heavy metals and sulfates; in the freshwater supplies that human is using to satisfy their daily requirements [8, 14-16]. One of the major



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issues of water pollution is the accumulations of nitrates to unsafe level that threaten the sustainability of freshwaters sources [17, 18]. The natural presence of nitrates, along with phosphates, in freshwaters bodies (surface or ground waters) is highly expected since these elements are commonly available in the geological sphere of the Earth (soils and rocks) [17]. Nitrates still occur in the natural waters since it is a fundamental component of the nitrogen cycle [15, 17]. However, the most serious problem is the natural concentration of nitrates in water sources has been substantially altered due to the persistent and considerable rejection of urban effluents, especially agricultural discharges that contain dangerous amount of nitrate-based fertilizers. As a result, the accumulated nitrate concentrations in freshwater sources have accumulated to unsafe levels (near to 100 mg/L) [3, 17].

The incidence of nitrate in potable waters at greater levels than the allowable concentrations (15 mg/L according to the World Health Organization (WHO)) induces negative consequences on both in human health (like the production of extreme blue baby syndrome and stomach cancer) and on the natural ecological environment (like the creation of eutrophication issues) [17]. These facts highlight the importance of utilizing different techniques for effective removal of nitrates [17]. Chemical oxidation and biological treatment are the most commonly used methods for de-nitrification of water and urban effluents [17, 19, 20]. The expensive treatment, high maintenance costs, sensitivity for external parameters and probability of creating toxic pollutants limit the use of these methods [17, 21-23]. Moreover, the majority of the conventional methods generate large volumes of sludge that also limits their application due to the high cost of sludge handling and the need for and specific equipment and strategies for the removal and transport of the sludge to the landfills (as solid wastes) [24-26]. However, some recent studies indicated that the sludge of treatment facilities could be recycled in different applications [27-30]. Recently, the electrocoagulation technology (ELE) has been used as a promising alternative for traditional treatment methods to remove many contaminants from urban effluents and water, including nitrates [17], fluoride [22], iron [23] and bacteria [21, 31, 32]. Other advantages of the ELE are: it does not produce secondary toxic pollutants, it has low operating and maintenance cost and it does not produce large volumes of sludge [33-35]. However, one of the annoying disadvantages of the ELE is the growing of passive layers on the aluminum anode after short time of operation, which substantially affects the efficiency of ELE.

Therefore, this research attaches the ELE to an ultrasonic bath to efficiently remove nitrates from drinking waters. The aim of utilizing the ultrasonic bath is to get rid of the anodic passive layers in the ELE system [32].

2. Methodology

2.1. Preparing of artificial wastewater

In the current study, the experiments were conducted utilizing artificial nitrates solution that was prepared by mixing the necessary weight of potassium nitrates (KNO_3) with suitable volume of deionized water (to produce solutions with different initial nitrates concentrations (INC) (100 to 200 mg/L)). All used glassware (containers and flasks), in this work, were rinsed three times with deionized water before using them to avoid any external contamination. Diluted acidic and basic solutions, 1M of NaOH and 1M of HCl, were used to control the WIP of the fresh nitrates solutions; WIP was set to 4, 6 and 8. Additionally, NaCl was used to keep the conductivity of the fresh nitrates solutions at 0.32 mS/cm.

2.2. Measurement cuvettes and tools

Nitrates concentration, before and during the filtration process, was accurately measured using LCK-standard cuvettes (manufactured by Hach Lang) and a Hach Lang spectrophotometer (DR-2800). A pocket meter (Hannah-98130) was utilizing to determine the pH, conductivity and temperature of water. Before conducting these tests, samples were filtered at 0.45 μm filter (Whatman brand) to remove flocs that could cause light scattering in the spectrophotometer, which alters the measurements.

2.3. Nitrates removal using ELE

The experiments were conducted in two phases; the first one has been allocated to explore nitrates removal using only ELE, whereas the second phase has been allocated to apply the U-ELE to remove nitrates.

In ELE phase, nitrates removal have been conducted utilizing a lab-scale continuous ELE system, consisting of a Perspex container having a 1.5 L capacity (1 cm rim, and has three 0.8 cm openings to manage water) and four electrodes of aluminum having total area of 404 cm². The polluted water was pumped via the ELE system utilizing a peristaltic (Model: 504-U), and the necessary current was supplied utilizing a DC rectifier (PS3010). The distance between electrodes had been held constant at 0.5 cm in the experiments of ELE, the temperature of water and conductivity have been also kept constant at 20°C and 0.32 mS/cm, respectively, these factors are compatible to the previous investigations [17, 23]. The WIP, FR, INC and ACD values were varied in different ranges to attain the best removal of nitrates, see table 1.

Table 1. Studied values of WIP, FR, INC and ACD.

Factors	Range
INC (mg/l)	100 to 200
FR (ml/min)	60 to 100
ACD (mA/cm ²)	6 to 9
WIP	4 to 8

To attain the best performance of the ELE, central composite design (CCD) has been applied to plan the experiments of nitrates removal. The CCD approach has been chosen in this research as it could provide reliable estimation of the overall impact of several variables on the removal of pollutants, and is commonly applied in the previous researches to optimize water and/or effluents treatment process [36]. Here, the CCD was utilized to design the tests for the impact of the mentioned four factors, which are WIP, FR, INC and ACD. For these four factors, the CCD suggests thirty experiments were needed with five levels at $\pm \alpha$ distances from the center. Table 2 presents a summary for the studied five levels and the $\pm \alpha$ values for each factor.

Table 2. The results of the investigated factors with $\pm \alpha$.

Factors	Five levels				
	$-\alpha$	-1	0	+1	$+\alpha$
INC (mg/l)	50	100	150	200	250
FR (ml/min)	40	60	80	100	120
ACD (mA/cm ²)	4.5	6	7.5	9	10.5
WIP	2	4	6	8	10

The design of nitrates experiments is presented in table 3, which was produced using the CCD technique.

These experiments were conducted to attain the best removal of nitrates using the ELE, which then will be used in the experiments of U-ELE.

1.5. Nitrates removal using U-ELE system

In the second phase of this research, the U-ELE system was employed to remove nitrates from water. In this phase, the electrodes were placed inside a laboratory-scale ultrasonic bath (FB-15051), as depicted in figure 1, and separated from the sides of the bath by plastic pieces. The experiments have been conducted by switching on the ultrasonic bath (frequency of 37 kHz) for 10 minutes continuously, and then it was switched off and applies the ELE using the best conditions (from the ELE experiments).

During the experiments of U-ELE, small samples were taken periodically from the bath to assess nitrates concentration according to the mentioned procedures in section 2.2 of this paper.

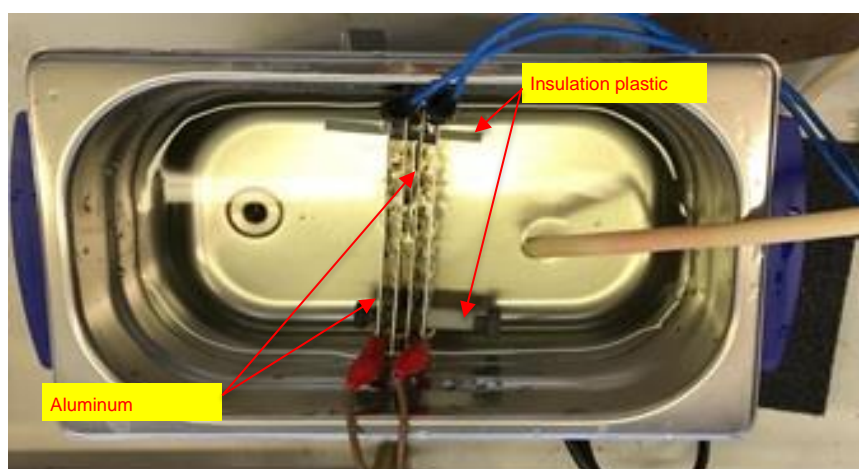


Figure 1. U-ELE system.

Table 3. The design of nitrates removal experiments.

Run	INC (mg/l)	FR (ml/min)	ACD (mA/cm ²)	WIP	Run	INC (mg/l)	FR (ml/min)	ACD (mA/cm ²)	WIP
1	100	60	6	4	17	50	80	7.5	6
2	200	60	6	4	18	250	80	7.5	6
3	100	100	6	4	19	150	40	7.5	6
4	200	100	6	4	20	150	120	7.5	6
5	100	60	9	4	21	150	80	4.5	6
6	200	60	9	4	22	150	80	10.5	6
7	100	100	9	4	23	150	80	7.5	2
8	200	100	9	4	24	150	80	7.5	10
9	100	60	6	8	25	150	80	7.5	6
10	200	60	6	8	26	150	80	7.5	6
11	100	100	6	8	27	150	80	7.5	6
12	200	100	6	8	28	150	80	7.5	6
13	100	60	9	8	29	150	80	7.5	6
14	200	60	9	8	30	150	80	7.5	6
15	100	100	9	8	31	150	80	7.5	6
16	200	100	9	8					

3. Results and discussion

3.1. Nitrates removal using ELE system

As it was stated in section 2.3 of this paper, the first phase was allocated to explore the removability of nitrates from water using the ELE. In this phase, the influences of the WIP, FR, INC and ACD on nitrates removal by ELE were experimentally optimized according to the stated experiments in table 3. The observed removals of nitrates are listed in table 4.

The gained results from the conducted experiments obviously show that the best removal of nitrates using the ELE was 77.7 percent that was attained at WIP, FR, INC and ACD of 6, 40 ml/minutes, 150 mg/L and 7.5 mA/cm², respectively. The results of table 4 show the following:

- 1- The INC has exerted the major influence on nitrate removal by the ELE system. Table 4 shows that raising the INC from 150.0 to 250.0 mg/l reduced nitrates removal from 77.7 to 29.3 percent, respectively. This reduction in nitrate removal is due to the fact that the generated aluminum hydroxides were not adequate to accommodate the high amount of nitrates [3]. However, according

to results, increasing the ACD or minimizing the FR, which allow more production of aluminum hydroxides [17].

- 2- Increasing the ACD led to obvious enhancement in nitrates removal. For instance, table 4 indicates that nitrates removal was 36.1 percent when the ACD was 4.5 mA/cm² (WIP, FR and INC were 4.5, 80 ml/min and 150 mg/L, respectively). However, nitrates removal was improved to reach 62.3 percent when the ACD was increased to 10.5 mA/cm² under the same values of WIP, FR and INC. This enhancement is because the ACD enhances the anodes' melting rate, which enhances the removal of pollutants [37].
- 3- Listed results in table 4 demonstrate that maximizing the FR exerted negative impacts on nitrates by ELE system. For example, maximizing FR from 60.0 to 100.0 ml/min reduced nitrates removal by about 5 percent at INC of 100 mg/L, ACD of 6 mA/cm² and WIP of 4. This adverse consequence of FR is due to the fact that the high flow levels reduce the detention time, which decreases the nitrates removal in consequence [3].
- 4- Table 4 indicates that intensifying the WIP from 2 to 6 improved the nitrates removal by about 20 percent. Reduction in nitrates removal in acidic WIP owing to the producing of aluminum hydroxides with positive charges, which results repulsion forces between coagulants and nitrates as the latter is also negatively charged [3, 17].

In brief overview, the experimental results indicated that the finest nitrates removal (77.7 percent) was accomplished at WIP of 6, ACD of 7.5 mA/cm², FR of 40 ml/min and INC of 150 mg/l. These observations were utilized to perform the U-ELE experiments.

Table 4. Experiment of nitrate removing utilizing the ELT technique.

Run	Removal (%)	Run	Removal (%)
1	59.2	17	58.1
2	56.1	18	29.3
3	54.1	19	77.7
4	46.9	20	45.1
5	58.1	21	36.1
6	61.3	22	62.3
7	62.4	23	31.8
8	52.1	24	59.5
9	70.5	25	53.1
10	58.2	26	52.2
11	60.3	27	54.1
12	57.1	28	51.6
13	73.4	29	52.2
14	66.4	30	52.3
15	68.3	31	53.1
16	63.2		

3.2. Nitrates removal using U-ELE system

In this phase of study, the experiments of U-ELE were performed by positioning the electrodes of aluminum inside the ultrasonic bath, and then the latter was switched on for 10 minutes. After that, the ELE was performed at WIP of 6, ACD of 7.5 mA/cm², FR of 40 ml/min and INC of 150 mg/l. Figure 2 shows that applying of ultrasonic irradiation improved nitrates removal by approximately 10 percent. The reason of improving the removal of nitrates when the ultrasonic was applied is related to the removal of passive layers from the anodes' surface, as well as the ultrasonic increases the electrochemical reactions' rate in the ELE system [38, 39].

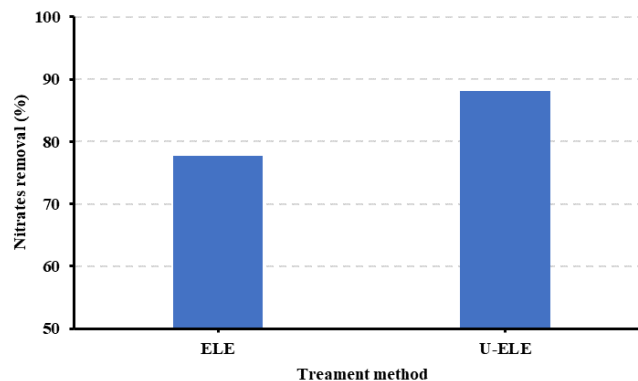


Figure 2. Nitrates removal using U-ELE and ELE.

For future studies, basing on the recent efficacious applications of sensors in different industries [40-44]; the duration of the ultrasonic irradiation could be controlled using a proper sensor to monitor the development of the passive layers on anodes' surfaces.

4. Conclusions

The current investigation combined the ELE and ultrasonic approaches to enhance the ability of ELE system to remove nitrates from solutions. This enhancement is related to ability of the ultrasonic radiation to get rid of the passive layers on the anodes' surfaces. The findings of this investigation showed that combining these two approaches has resulted in about 10 percent increase in nitrates removal from solution. The results of the experiments also demonstrated that the INC, FR, IWP and ACD affect the removal of nitrates; increasing the ACD and IWP positively affect the removal of nitrates, while increasing the rest of factors have negative impact on nitrates removal. In brief overview, the best possible nitrates removal was achieved at ultrasonic time of 10 minutes, INC of 150 mg/l, FR of 40 ml/minutes, WIP of 6 and ACD of 7.5 mA/cm².

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