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Study the Performances of Electrodialysis using Anionic Membrane Exchange AXE for Removing Nitrate Ions from Ground Water

Sakina Belhamidi^{1,2*}, Omar Elrhaouat^{1,3}, Hajar Zeggar^{1,3}, Fatima Elhannouni^{1,3}, Mohamed Taky^{1,3}, Azzedine Elmidaoui^{1,3}

¹Laboratory of advanced materials and process engineering, Department of Chemistry, Morocco ² Superior School of Technology, Ibn Tofail University, Kenitra, BP.1246, Morocco ³Faculty of Sciences, Ibn Tofail University, Kenitra, BP.1246, Morocco

Abstract

Pollution of groundwater and surface water by nitrates is becoming a common problem for both industrial and developing countries. The concentration of nitrate in many regions in Morocco greatly exceeds the standards for drinking water. The harmful effects of nitrates on infants in particular, are well known. Methemoglobinemia and the carcinogenicity of nitrosamines constitute the main risks of drinking water polluted with nitrate. The objective of this study is to evaluate the efficiency and the feasibility of the anion exchange membrane: AXE using an electro-dialysis pilot plant for the reduction of nitrate ions of groundwater of Sidi Taibi (Region of Kenitra), containing a concentration of nitrate in order of 60-120mg/l. The study of the optimization of the parameters influencing the efficiency of the anionic membrane exchange, namely: Flow rate, Voltage, Specific energy consumption, Optimization of the recovery rate, Demineralization rate, confirmed the feasibility and the efficiency of this membrane. The results of this study show that only 33% demineralization rate, the standard concentration of nitrate (50ppm) was obtained.

Keywords: Electro dialysis, Nitrate removal, Optimization, Anionic membrane exchange (AXE)

1 Introduction

Nitrate focus on surface and ground water has expanded in practically all regions of the world so much that the conceded world wellbeing association standard of 50 ppm, has to a great extent been surpassed in numerous areas. This pollution has caused closure of wells and delivered numerous springs unusable as drinking water sources. In Morocco, the convergence of nitrate in ground water in certain districts surpasses 250 ppm [1]. This ascent identifies with an expansion in modern and agrarian nitrates squanders and particularly to the weighty usage of counterfeit composts. The NO₃ is non-toxic for humans and animals; it can be reduced to NO2 and is readily absorbed into the bloodstream where it combines with the ferrous ion of hemoglobin to form met-hemoglobin [2]. This process can lead to oxygen deficiency in the body's tissue and a dangerous condition called Methemoglobinemia, which is fatal to infants [3]. In addition, the formation of nitrosamines by nitrite can cause cancer in the digestive tract because nitrosamines are the most effective carcinogens in mammals [4]. Therefore, the removal of nitrate from water is a very critical and essential topic and has attracted considerable attention. For this reason, several treatment processes including Adsorption [5, 6, 7] ion exchange, biological denitrification, chemical denitrification, reverse osmosis, Nano filtration, electro dialysis, and catalytic denitrification can remove nitrates from water with varying degrees of efficiency, cost, and ease of operation [8, 9, 10, 11, 12, 13, 14], also chemical methods are some of the conventional techniques implemented. Electro dialysis can be considered promising technology in water treatment to treat groundwater with such nitrate due to its high efficiency, high effluent water quality, modularity, and flexibility [15, 16, 17, 18, 19].

A full-scale electro dialysis treatment plant has shown that it is possible to pass a nitrate concentration in raw water from 155 mg NO₃⁻ /L to 40 mg NO₃-/L thanks to 3 stacks of membranes in parallel, each with a hydraulic capacity of 48 m³/h. The concentrate was shipped to the civil wastewater treatment plant [20]. Pilot trials of an electro dialysis process reducing the nitrate concentration by 73 mg NO₃-/L to 8.2 mg NO₃-/L, with a recovery rate selective nitrate membrane indicated that approximately 80% of the nitrate could be removed to a concentration of 13 mg NO₃-/L in the treated water. The system operated at 1 m³/h with 2 stacks of membranes; its recovery rate was 95% [21,17] published results by using a 24 m³/day electro dialysis pilot plant capable of 96.8% for 50% of desalting. To get these outcomes, a normal qualification of 23.8 V was applied, and the power consumption was 0.43 kWh/m³ [16, 22]. Published data on a full-scale electro dialysis treatment system capable of removing more than 93% of nitrate, reaching a concentration of 4.3 mg NO₃-/L in the treated

Corresponding author: Sakina Belhamidi, (a) Laboratory of advanced materials and process engineering, Department of Chemistry, Morocco and (b) Superior School of Technology, Ibn Tofail University, Kenitra, BP.1246, Morocco. E-mail: sakinabelhamidi@hotmail.fr

water. The system consisted of 3 steps, and its water recovery rate was 90% [22]. In addition, other studies have been conducted on electro dialysis treatment systems for optimization, evaluation of the efficiency of new membranes, and examination of the effect of the source of water variability [23, 18, 24, 19]. Moreover, the main considerations when using electro dialysis systems to remove nitrate are the complexity of the operation of the system, the elimination of water discharges and the need to adjust the pH of the treated water [25].

Anion exchange membranes selectively removing nitrate have been developed [21], (ACS) membrane has been shown to be the best membrane for nitrate removal [18]. The objective of this work was to evaluate the performances and the efficiency of an anionic membrane exchange (AXE) in the nitrate ions removal of groundwater. For this purpose, experiments were conducted of groundwater from the region of Sidi Taybi (Kenitra region) containing 60-120 mg/L of nitrate ions by using an electro dialysis pilot plant. In addition, an optimization operation was conducted for various experimental conditions including the determination of the demineralization - nitrate reduction correlation, the influence of voltage on the performances of their reduction, the influence of flow rat and finally the optimization of the recovery rate.

2 Experiment

2.1 Pilot Plant

The electro dialysis type TS 2-10 used in our study is provided by the company Eurodia Corp (Figure 1). A subsidiary of the Japanese company Tokuya Masoda. The electro dialysis stack was equipped with 22 compartments through which the fluids circulate. The stack design characteristics are given in table 1.

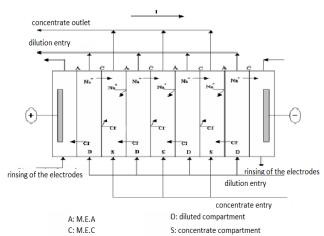


Figure 1: Synoptic scheme of the electrolysis system

Table 1: Characteristics of pilot equipment's

Equipment	Number
Dilution compartments (diluât)	10
Concentration compartments (concentrated)	10
Electrode rinse compartments (rinse)	2

The raw water circulates in the dilute and concentrates stream. In the wash, there was a solution of sulfonic acid was utilized to evade among others the accompanying burdens: precipitation of salts, rapid consumption of electrodes. The qualities of the parameters of the samples taken occasionally were resolved logically observing standard methods [26]. Table 2 gives the characteristics of the used membranes.

Table 2: Characteristics of the used membranes

Membrane	AXE	CMX
Thickness, (mm)	0.1	0.18
Electrical resistance, (Ohm cm ²)	1.4	3.0
Exchange capacity, (meq.g-1)	2.0	1.65
Burst strength, (Kg.cm ²)	2.75	5.5
Active surface, (cm ²)	200	200

2.2 Raw Water

The electro dialysis operations were conducted in Sidi Taibi underground water (Province of Kenitra, Morroco). The analytical results of the untreated water are shown in Table 3. The quality of Sidi Taibi underground water is not in conformity with sanitary standards because of the high concentrations of nitrate which exceeds the standards required by WHO (50 mg/L). These results could be explained by the prevailing sandy nature of the soil in the area, the frequency of fertilizer usage and the closeness of the water table [27].

Table 3: Characteristics of raw water

Parameters	Values
Temperature, C°	26.1
pH	7.6
Conductivity, µS/cm	1132
TDS, ppm	808.49
NO3-, ppm	105.6
Cl-, ppm	141.8
HCO3-, ppm	306.3
SO42-, ppm	10
Ca2+, ppm	92.55
Mg2+, ppm	56.42
K+, ppm	7.96
Na+, ppm	34

Determination of the performance of electro dialysis; The parameters taken into account are as follows:

 Recuperation rate (Y%) is determined utilizing the accompanying equation:

$$Y\% = (Qp/Qf) \times 100 \tag{1}$$

where Qf $(L/h/m^2)$ and Qp $(L/h/m^2)$ are the feed and the permeate flow rate, respectively.

• Rate of Rejection (TR%) is defined as follows:

$$TR\% = (C0-CP/C0) \times 100$$
 (2)

where, Cp (g/L) and Cf (g/L) are respectively permeate and initial concentration.

• Demineralization rate (DR %) is defined as follows:

$$DR\% = (Condf-Condp / Condf) \times 100$$
 (3)

where, Condp and Condf; are respectively; the conductivity ($\mu S/cm$) of permeate and the feed water.

3 Results and Discussions

The approach consists in determining the conditions allowing a good denitrification for that, the ED operation was carded out by following the evolution of these parameters: Voltage, rejection of ions, demineralization rate, flow rate and recovery rate on both ends of the stack. The performances of ion exchange membranes were studied. Under these optimal conditions, the nitrate in water treated should be less than 50 ppm.

3.1 Current limit

The limit current is an inseparable important parameter in the operation of electro dialysis, it is essential to calculate it prior to the experimental runs to avoid polarization in the ED system, the limit current reflects the maximum amount of current that can carry the counter-ions through the membrane. Beyond this value, the surplus current will contribute exclusively to the formation of polarization phenomena [28]. As shown in figure 2, the limit current is calculated by the Cowan and Brown method, the curve is divided into three zones. In the first zone, the resistance decreased with the inverse of current. In the later region, the detour point, at a current of about 0.75 A, was determined due to the depletion of ions in the attenuator current. This point has been referred to as the bound current of the ED system or even the point of polarization. In the last zone, polarization occurred when ED system was operating at a current above the limiting current, which led to grow in the system resistance. To operate in full security, the operating current must be less than 80%

of the limit current. Therefore, the operating current of this study must be lower than 0.6 A.

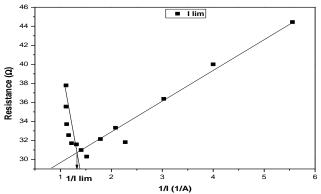


Figure 2: Variation of current limit

3.2 Impact of stream rate

Flow rate of concentrate and dilute compartment solution is one of the parameters affecting the removal of electro dialysis. To study the effect of flow rate, the experiment was performed with various flow rate conditions and the voltage was fixed at 10 V, which the flow rates of concentrate and dilute compartment solution were simultaneously set at 100, 180 and 260 L/h.

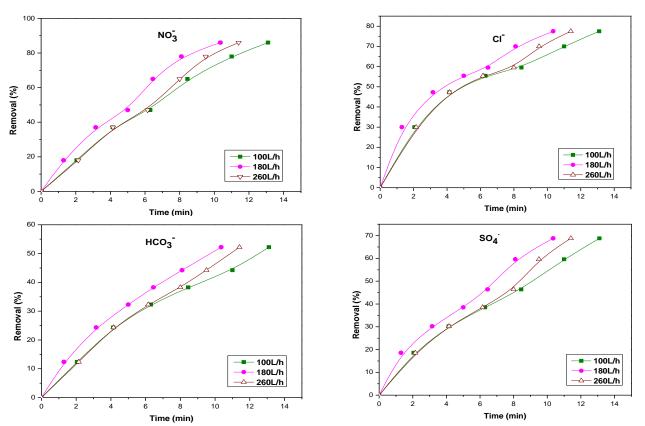
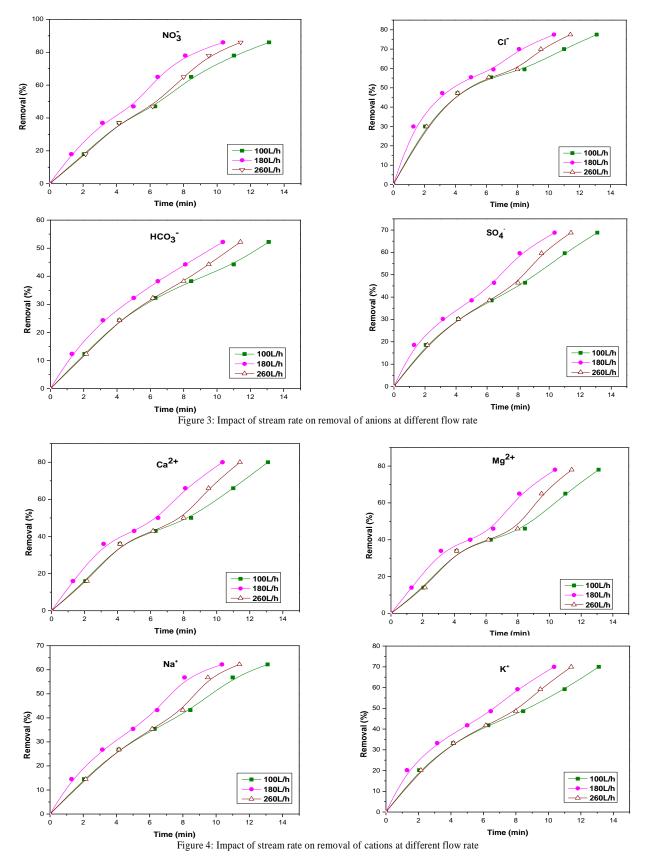


Figure 3: Impact of stream rate on removal of anions at different flow rate



The removal efficiency can be enhanced by the use of high flow rates that permits a better migration of ions through the ion-exchange membrane due to the mitigation of boundary layer or the concentration polarization formed on the membrane. Figure 3 and 4 showed the effect of the flow rate on the removal of anions and cations, the ability to remove various ions increases with the increase in the flow rate of concentrate/dilute compartment solution from 100 to 180 L/h, while for the flow rate of 260L/h the removal efficiency decreases. Similar results are obtained by Min and Kim [29]. This implies that the movement of ions towards the membrane may be disturbed beyond a certain value of flow rate due to higher tangential flow to the membrane against the electric migration flow of normal direction to the membrane, although the resistance of electric migration by boundary layer would be mitigated until the flow rate of solution reaches a certain value [30]. This result suggested that the flow rate of the solution in the system should be also considered for evaluating system optimization.

3.3 Effect of voltage

The effect of voltage on membrane selectivity was investigated for three voltage values (5V, 10V, and 15V), with a flow rate was fixed of 180 L/h for the concentration and dilution solutions. The results of removal anions and cations according to the applied voltage and the operating time shown in figure 5 and figure 6. The time of removal for each ion decreases with an increase in applied voltage. For the disqualified nitrate of 78%, the operation of the electro dialysis takes 29.1 min, 8.1 min, and 5.18 min for the application of the voltage of 5 V, 10 V, and 15 V respectively.

The same inclination was reported that the time of removal of other anions and cations decreased by elevating the applied voltage from 5 to 15 V. The increase in the applied voltage increases the rate of ion transfer through the membranes, and the separation is very fast for 10 and 15 V, and it is slower for 5 V. Similar phenomena were observed for the removal of other ions chloride, sulfate, bicarbonate, sodium, calcium, magnesium, and potassium ions [30]. For demineralization rate of 50%, a removal of (66%, 87%, 56.8%, and 59.2%) was achieved for the cations: Mg2+>Ca2+>K+>Na+ and (78%, 70%, 44.23%, and 59.6%) were achieved for the anions: NO₃->Cl⁻ >SO₄²>HCO₃. The same effect from the size of ions hydrated that was observed with cations was also observed in the removal of anions. They were removed in the following order: NO₃->Cl-> SO₄²->HCO₃-. As it was mentioned, ions with a lesser hydrated ionic span and more fragile hydration shell were removed more efficiently than ions with a bigger hydrated ionic radius and more grounded hydration shell. The hydrated radius of anions from the smaller to the larger as the following order NO³->Cl⁻>SO₄²>HCO₃-, this order corresponds to the removal observed in this study [16,31,32]. The elimination rate of these ions is linked to different focus of many ionic species in the solution, the effect from the size of ions hydrated radii and the ion charge thickness density.

3.4 Specific power consumptions

Specific power consumption (SPC) parameter calculation was also done; this can be described as the energy needed to treat the unit volume of the solution without taking the energy of the pumps into consideration. SPC was determined utilizing the following equation [33]

$$SPC = \frac{I}{Vd} \int_0^t U \, dt \tag{4}$$

where I (Ampere) is the enforced current intensity, U (V) the Potential, Vs (L) the weaken stream volume and t is the time. The SPC is calculated at different voltage values 5, 10, and 15 V for a stream rate of 180 L /h. The results of the SPC calculation show that the increase of the voltage for a constant flow rate affects significantly the power consumption. As shown clearly in figure 7, the SPC values increase from 0.023 Wh/L to 0.062 Wh/L with the increased voltage from 5 V to 15 V. According to these results the energy consumed by 5 V is the lowest. In the rest of the study, the voltage will be fixed at 10 V which has been set by the manufacturer at the rate of $1 \, \text{V}$ / cell of 80% of the total volume.

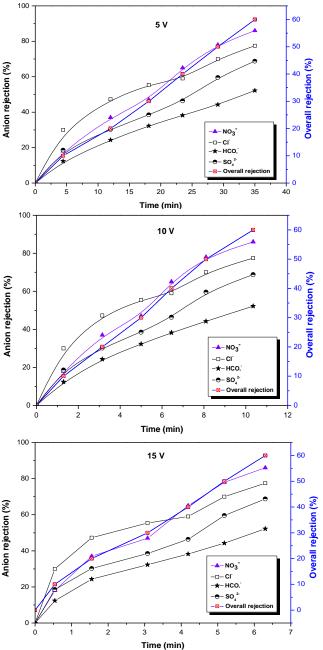


Figure 5: Effect of voltage on removal of anions at different voltage

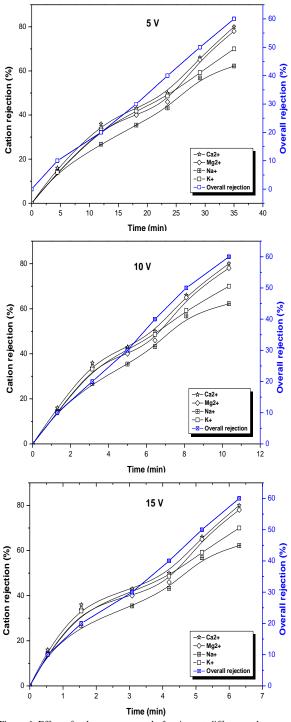


Figure 6: Effect of voltage on removal of cations at different voltage

3.5 Demineralization - nitrate reduction correlation

The concentration of nitrate is calculated for each demineralization rate, figure 8 shows the evolution of the demineralization rate with retention of nitrates. The concentration of nitrates equal to 50 ppm is obtained at a demineralization rate of 33 %. In the rest of the work, the demineralization rate will be set at 40% to obtain an optimal nitrate concentration of 36 mg/L.

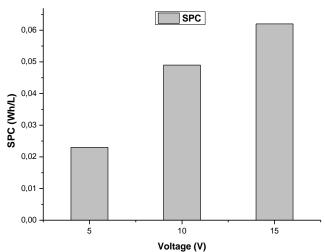


Figure 7: Effect of the applied voltage on the specific power consumption for the flow rate of 180 L/h

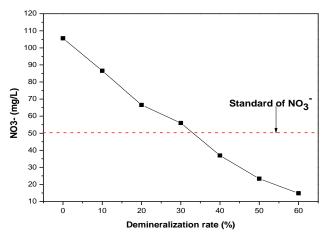


Figure 8: Evolution of the demineralization rate Vs retention of nitrates

3.6 Optimization of recovery rate

The experiments were carried out for a demineralization rate of 40 %, the voltage was 10 V and the flow rate was 180 L/h. The conductivity was 940 us/cm, the nitrate inlet was 105 ppm and the outlet were 33 ppm. The recovery rate is limited by the precipitation of divalent salts, especially sulfate or carbonate scale. The precipitation was controlled with the naked eye and by following the variation with time or with conductivity of pH, and Ca2+/Mg2+concentration ratio. To optimize the water recovery, the impact of the addition of acid to the brine stream was calculated. Optimization consists of creating the conditions allowing the obtaining of a high recovery rate with a low consumption of reagents [26, 34]. The improvement was carried out for the following running conditions: without addition of acid to water to the brine stream, with modification of the brine pH to 6.5.

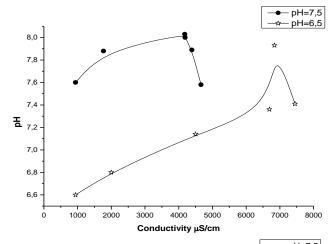
• Recovery rate without acid (pH = 7.5)

The start of scaling was observed after 85 min of electro dialysis operation and for a pH of about 8.03 and conductivity of about 4180 μ S/cm. Figure 9 shows the variation with time of the pH, and Ca2+/Mg2+ in the brine stream. These parameters increase with time

and after 85 min, a decrease is observed indicating the start of scaling. The recovery rate in this case was 91.6%.

• Recovery rate with acid HCI (pH = 6.5)

The brine stream pH was adjusted to 6.5 using a concentrated HCI solution. In this case the scaling was not observed with the naked eye. The analysis shows that the scale starts after 229 min, pH = 7.41 and conductivity 7450 μ S/cm (Figure 8.), the calculated recovery rate was 94.8%.



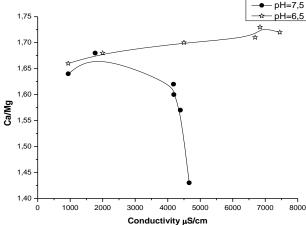


Figure 9: Conductivity's pH, and Ca2+/Mg2+ ratio for (pH = 7.5 and 6.5)

3 Conclusion

This paper highlights the feasibility and the effectiveness study of the anionic membrane exchange: AXE to remove nitrate ions from groundwater, the most prominent conclusions are the following:

- The study was carried out for the first time on the AXE membrane to remove nitrate ions, which was designed only for sugar in many previous works [35].
- Nitrate elimination capacity ions increases with the increase in the flow rate to 180 L/h,
- At only 33% demineralization rate, the standard concentration of nitrate was obtained (50ppm).
- The specific power consumptions have been set at 0.046Wh/L for the applied voltage of 10V which is the optimum conditions.

 The AXE anionic exchange membrane confirms the satisfactory and the effectiveness performances from removing nitrate ions.

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Ethical issue

The authors declare that this manuscript is original from our laboratory (Advanced Materials and Process Engineering) and respects the publication standards with regard to: Double publication, references and results illustrated in this manuscript.

Competing interests

The authors appear their names in this scientific work, certify that they have no conflict of interest and that there is no funding for this work.

Author's contribution

All the authors contributed of the production of this manuscript including data collection, writing and statistical analysis.

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