*Indonesian Journal of Science & Technology 6 (1) (2021) 17-30*

Indonesian Journal of Science & Technology

Journal homepage: http://ejournal.upi.edu/index.php/ijost/

# The Comparison of Electrodialysis and Nanofiltration in Nitrate Removal from Groundwater

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## ABSTRACT

Nitrate groundwater contamination is of major interest all over the world. This problem arises in agricultural regions across Morocco. An excess amount of nitrate causes a serious problem in urban water networks and human health. Because of these health risks, considerable attention has been paid to find effective treatment processes to reduce nitrate concentrations to safe levels. The World Health Organization has set an acceptable level for nitrate in drinking water at 50 mg/L. The aim of this study is to reduce the nitrate concentration from groundwater using two membrane processes: Electrodialysis (ED) and Nanofiltration (NF). Efficiencies of these two technologies are compared in respect to nitrate ions removal, cost process and final quality of water. The results of technologies show that, for electrodialysis standards level can be achieved for a demineralization rate of 15% and the physico-chemical quality of the produced water is satisfactory. For nanofiltration we obtain a nitrate removal of 90% but the produced water is very de-mineralized and must be remineralized.

## ARTICLE INFO

***Article History:***
*Submitted/Received 07 Aug 2020*
*First revised 28 Sep 2020*
*Accepted 03 Dec 2020*
*First available online 20 Jan 2021*
*Publication date 01 Apr 2021*

***Keywords:***
*Electrodialysis,*
*Groundwater treatment,*
*Nanofiltration,*
*Nitrate removal.*

## 1. INTRODUCTION

The increase of nitrate concentrations in ground water around the world mostly results from intensive application of fertilizers. Nitrate is a main source of nitrogen for plants and it is the form of dissolved nitrogen that occurs naturally in soil and water. In Morocco, nitrate concentrations exceeding the 70 mg/L standard is the main reason for closure of wells in the coastal aquifers. An excess amount of nitrate causes human health problems (Kikhavani *et al.,* 2014).

Among the harmful effects of nitrates on human health are methemoglobinemia and other diseases (Cheikh et al., 2013), and on the ecosystem we find eutrophication phenomena; In addition to infant mortality, several reports indicate that the use of drinking water with a high level of nitrate could lead to some types of cancer, including stomach cancer, central nervous system, congenital anomalies and hypertension (Nicolas *et al.,* 2010; Banasiak *et al.,* 2009).

The nitrate concentration limit of drinking water in Morocco and in some European countries is set at 50 mg/L. The World Health Organization has set a maximum limit of 50 mg/L in drinking water (Djouadi *et al.,* 2018). Unfortunately, the policy of countermeasures, especially these concerning agriculture and environment to limit pollution by nitrates, is efficient only in the long term plan (Ousmana *et al.,* 2018). So, technical solutions become obligatory. To overcome this problem, considerable attention has been paid to find effective treatment processes to reduce nitrate concentrations to safe levels.

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Several methods for nitrate removal from drinking water resources have been applied. These methods available for the removal of nitrate are ion exchange (Boumediene & Achour, 2004; Samatya *et al.,* 2006), biological denitrification (Samatya *et al.,* 2006; Wasik *et al.*, 2006), catalytic reduction (Samatya *et al.,* 2006; Lüdtke *et al.,* 1998), reverse osmosis, nanofiltration (Schoeman & Steyn, 2003; Touir *et al.,* 2021 ; El-Ghzizel *et al.,* 2020) and electrodialysis (Elimidaoui *et al.,* 2002; Tahaikt *et al.,* 2008). However, these techniques have many limitations. Conventional adsorption techniques require an estimation of the adsorption efficiency, disposal of the adsorbents with nitrate, reusability and proper selection of the adsorbent such that it is robust and can work under variable environmental conditions (Bhatnagar *et al.,* 2011). Ion exchange techniques are sensitive to various contaminants present in the water (Kapoor & Viraraghayan, 1997) and require post-treatment. Biological denitrification method using degradation of microorganism offers the possibility of a very specific and selective reduction of nitrate to nitrogen. However, there are some limitations due to contamination of drinking water with germs and metabolic substances. Because of this, an extensive reconditioning of the drinking water by filtration and germicidal treatment is necessary (Samatya *et al.,* 2006). Chemical methods cause toxicity in the water, the effects of pH and temperature are important and the post-treatment is required due to the production of y-products (Bhatnagar *et al.,* 2011).

Thus, the use of membrane processes is becoming more popular for the removal of nitrates. In fact, the advantage of membrane processes over conventional separation methods is its high removal capacity, operational flexibility and cost-effectiveness (Elmidaoui *et al.,* 2002). There are only three membrane processes available for ion separation solutions, reverse osmosis, nanofiltration and electrodialysis that have reached the practical application stage for the removal of nitrates in drinking water and wastewater (Tahaikt *et al.,* 2008).

In Morocco, the nitrate elimination from central Morocco groundwater (Boujaad) by electrodialysis and nanofiltration was examined with the collaboration of many organizations: ONEE (National Office of Electricity and Potable Water), Eurodia Corp., France (subsidiary of Tokuyama Corp., Japan) and Ibn Tofail University.

Nanofiltration (NF) is an intermediate process of ultrafiltration and reverse osmosis which carries advantages such as the efficient removal of dissolved solutes, including multivalent ions and organic compounds of high molar mass (El Harrak *et al.,* 2015); however, this is done with lower pressure requirements and higher flows then reverse osmosis (López *et al.,* 2019). The evaluation of the optimum operating conditions for each specific NF system allows improvement of the overall performance of the process, both in terms of quality of the permeate and fouling (Andrade *et al.,* 2014).

Electrodialysis (ED) is a membrane process driven by the electric force in which ions are removed via ion exchange membranes under the influence of an electromotive force. With this technique one can decrease/increase ion concentration in solutions. The ED's influencing parameters are the type of the membrane (material, selectivity), feed's chemical composition, applied voltage, temperature, pH of liquid, and flow rate. Moreover, Electrodialysis is an emerging technology for drinking water production, seawater desalination or recovery of certain elements (Torma & Eséfalvay, 2018).

According to several studies and in the range of salinity being studied, electrodialysis remains the most appropriate method compared to other technologies such as reverse osmosis and nanofiltration (Geluwe *et al.,* 2011).

Research has been conducted for nitrate removal by nanofiltration and electrodialysis. A study by Paugam et al. focused on nitrate removal by nanofiltration using the Nanomax 50 type polyamide composite spiral membrane (Millipore, USA) with a filter area of 0.37 m2 and a cut-off threshold of 350 daltons and which made it possible to obtain an elimination rate of 73.7% of nitrate with 50 mg/L in raw water (Paugam *et al.,* 2001). Santafe´-Moros et al. tested three nanofiltration membranes NF90, NF270 and ESNA1-LF composite thin-film polyamide in the removal of nitrate and they showed that the NF90 membrane allowed having a higher rejection rate at 80% (Santafe´-Moros *et al.,* 2005). Elmidaoui et al. worked on the use of electrodialysis in the elimination of nitrate, and they tested different anionic membranes (AFN, ACS, AMX, ADP and ADS) coupled by a cationic CMX membrane and they found that the couple membrane AFN/CMX gave an elimination rate of 80.5% of nitrate (Elmidaoui *et al.,* 2001). In another study, Elmidaoui et al. treated the nitrate with the membrane pair ACS / CMX, the authors obtained levels of removal of 80.5% and 88.8% for a desalination rate of 30% and 50% respectively (Elmidaoui *et al.,* 2002). Laura et al. 2009 used the AMX / CMX membrane pair, a nitrate reduction rate of 94.1% was achieved (Laura *et al.,* 2009).

It is difficult to analyze and compare the costs that are based on the operating conditions and the quality of the produced water. Moreover, there are not enough experiments in brackish water desalination by NF or in specific treatments, especially nitrates by ED and NF. In the literature, F. Alazhar et al. have estimated the cost of desalination and defluoridation of brackish water by nanofiltration followed by remineralization by line saturator as post-treatment. The total cost has been estimated at 0.212 €/m3 (Elazhar *et al.,* 2009). For the same water and the same design conditions, S. Lahnid et al. estimated the total cost of defluoridation by electrodialysis and they obtained 0.154 €/m3 (Lahnid *et al.,* 2008). For an existing NF desalination plant in Florida with a capacity of 53,000 m3/d of groundwater, Bergman gave a total treatment cost of 0.23 €/m3 (Bergman, 1995). For several plant capacities, Wiesner et al. have estimated to 0.24 €/m3, 0.32 €/m3 the cost of the treatment of surface water for drinking water production by NF (Wiesner *et al.,* 1994).

The aim of this study is to evaluate the performance of nanofiltration as compared to electrodialysis to reduce nitrate ions from groundwater in the Boujaad region. Efficiencies of two technologies were compared in terms of nitrate ions removal, quality of the produced water and operation cost.

## 2. METHODS AND MATERIALS

The experiments are performed on an NF and ED pilots plant. The effluent of the current study is supplied from on underground water in the Boujaad region. During the period of the study a very slight variation in the physico-chemical parameters of the water was observed which is due to the drought which has hit the region in recent years. **Table 1** gives the characteristics of the untreated water, which is brackish and contains many excessive ions, especially nitrate.

The electrodialysis operation is carried out on a commercial pilot plant of one m3/h supplied by Eurodia Corp (**Figure 1**). The plant is equipped with two stacks arranged in series; each stack is packed with 30 pairs of anion and cation exchange membranes providing for each an available membrane area of 500 cm2.The membranes used are ACS as an anion exchange membrane and CMX-Sb as a cation exchange membrane; both were made by Tokuyama Corp.

The water treated circulates through the rinse, the dilute and the concentrate compartment using centrifugal pumps. To prevent scaling and fouling of membranes, the polarity is reversed automatically each20 min and the stack is flushed periodically with hydrochloric acid (0.1 M) to remove eventual precipitation of salts.

The NF pilot plant (E 3039) used, is supplied by TIA company (**Figure 2**). The pressure applied over the membrane varied from 5 to 70 bars with manual valves. The pilot plant is equipped with two identical spiral wound modules operating in series. Each module contains one element. The pressure loss is about two bars corresponding to one bar of each module

**Table 1.** Characteristics of the feed water

| Temperature, °C | 23 |
|---|---|
| **Conductivity, µS/cm** | 1400 ± 90 |
| **pH** | 7.34 |
| **Hardness, °F** | 55.5 |
| **Alkalinity, °F** | 25 |
| **$NO_3^-$, mg/L** | 85 ± 7 |
| **Langelier index** | 0.02 |

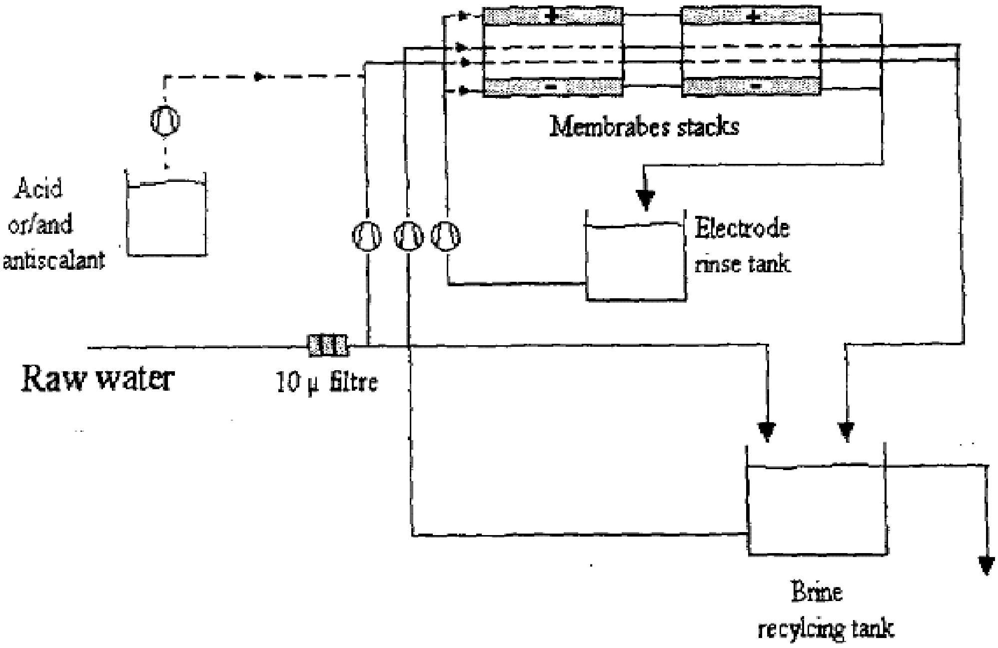

**Figure 1.** Schematic diagram of the Electrodialysis (ED) system.

**Table 2.** Characteristics of ED membranes used.

| Membrane type | ACS | CMX-Sb |
|---|---|---|
| **Thickness (cm)** | 0.17 | 0.18 |
| **Electrical resistance (Ω.cm$^2$)** | 2.0 | 3.0 |
| **Exchange capacity (méq.g$^{-1}$)** | 1.8 | 1.65 |
| **Burst strength (Kg.cm$^2$)** | 4.0 | 5.5 |

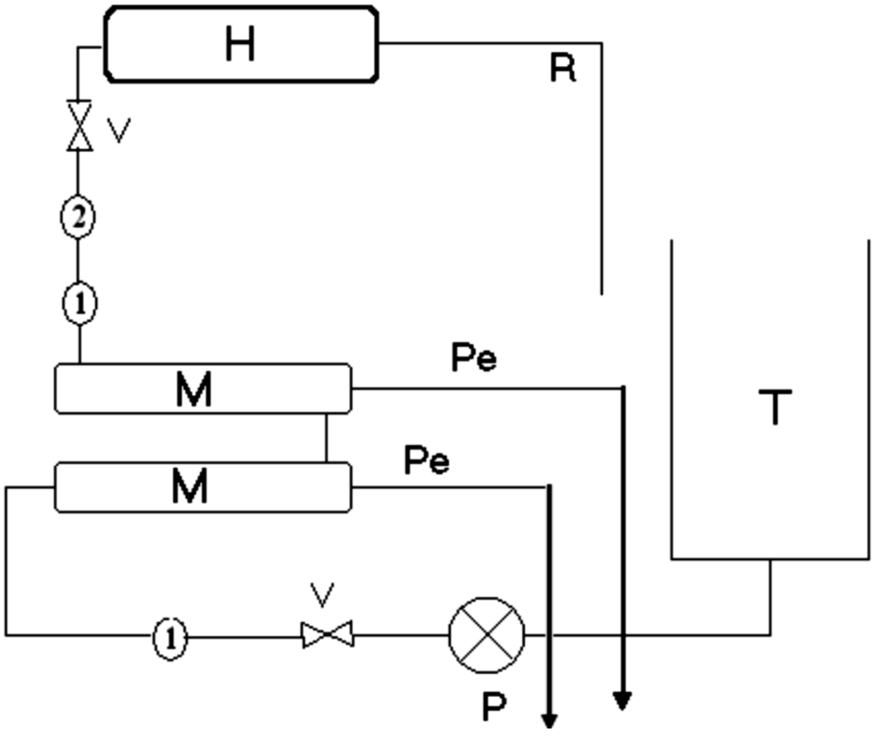

**Figure 2.** Schematic diagram of the Nanofiltration pilot plant

**Table 3.** Characteristic of NF membrane used.

| Membrane | NF90 |
|---|---|
| **Cut-off (Da)** | 90 |
| **Surface (m$^2$)** | 7.6 |
| **Material** | polyamide |

**Table 3** gives the characteristics of the commercial membranes used. After experiments, membranes are cleaned using alkaline and acidic solutions according to the manufacturer's recommendations. Samples of permeate are collected and the water parameters are determined analytically following standard methods previously described (Elmidaoui *et al.,* 2002; Tahaikt *et al.,* 2008). However, other parameters arefollowed such as the ion rejection (R) and energy consumptions (W) by nanofiltration and electrodialysis, which are defined as $R\ (\%) = \left(\frac{1-Co}{Cp}\right)100$ , where $C_P$ and $C_0$ are respectively permeate and initial concentrations.

Specific energy consumption for pumping is defined as $W = \frac{\Delta P * 100}{\eta * r * 36}$ , where $\Delta P$ is the trans-membrane pressure in bar, $\eta$ is the global pumping system efficiency and $r$ is the conversion rate.

Specific energy consumption for electro-dialysis is defined as $W = \int I * V * dt$ , where $I$ is the current intensity, $V$ is the applied voltage and $dt$ is the time interval

## 3. RESULTS AND DISCUSSION

### 3.1 Electrodialysis

**Figure 3** represents the variation of *U*/*I* as a function of 1/*I* in order to determine the fundamental parameter in electro-dialysis, the limiting current (Ilim). to reduce the phenomenon of concentration polarization and avoid diffusion, the current density applied through the ED cell is set at a value lower than the limit current (Ilim). The results show that the limiting current is equal to 6.2 A. For more safety, the currents applied in all the experiments are less than I = 0.8 Ilim.

**Figures 4 (a, b, c and d)** represent respectively, the pH, the Langelier index, the conductivity and the rejection rate of nitrates, sulphate, hardness and alkalinity as function of the demineralization rate (DR).

The results show that the pH decreases slightly with the rate of demineralization following the decrease in salinity and bicarbonate content according to the following relationships: $\mathrm{pH} = pK_1 - \mathcal{E} + \log[\mathrm{HCO_3^-}] - \log[\mathrm{CO_2}]$ , with $\mathcal{E}$ is expressed as a function of the ionic strength, $\mu$ of the solution $\mathcal{E} = \frac{\sqrt{\mu}}{1+\sqrt{\mu}}$ , and $pK_1$ is the acidity constants of carbonic acid.

The Langelier index decreases and Beyond 20% demineralization, the treated water becomes negative and therefore aggressive. Waters with a positive Langelier index are scaling and if it is zero, the water is balanced. Hardness and alkalinity decrease almost linearly as the rates of demineralization.

A nitrate rejection rate of 90% is achieved for only a 60% of demineralization rate, in addition to, the 70% rejection rate for alkalinity and 68% for hardness but only 20% for sulphate are obtained. These results can be explained by the nature of the ACS membrane which is preferably selective to monovalent anion.

According to these results, two treatments are possible; the choice depends on the local conditions and the desired final quality of water. The first optionis the demineralization rate of 35% allows us to operate under optimal conditions and to obtain a balanced water with a nitrate content of 23 mg/L. The second option is the reduction of nitrate ions to the maximum allowable concentration (50 mg/L) with a demineralization rate of 15% increase with an acceptable quality of water produced.

The optimization of the conversion rate at the demineralization rate of 15% shows that the adjustment of the concentrate at pH 7 is sufficient to treat without showing any sign of precipitation and for this, we proceed to the addition of the HCl acid in brine compartment that allowed us to achieve a conversion rate of 98%. The same results are registered by Kikhavani *et al.* (2014 ) and Menkouchi Sahli *et al.* (2004).

### 3.2 Nanofiltration

**Figure 5** shows the variation of flux in raw water and pure water as function of pressure. The permeate flow increases almost linearly with the pressure applied according to Darcy's law for the two waters studied. The increase in pressure improves driving forces and overcomes membrane resistance. The permeate flux with pure water rate is higher than that with raw water which is due to the phenomenon of concentration polarization which increases with increasing salinity.

**Figure 6** shows the variation of rejection rate of physicochemical parameters of the treated water as a function of pressure. For the different applied pressures, a slight increase in the nitrate, chloride and fluoride rejection rates is observed. The rejection rates that have been achieved are 94% for nitrates, 95% for chlorides, and 97% for fluorides. For sulfate, the rejection rate is 100% and remains practically constant. The anions rejection rate for the NF90 membrane is arranged in the following order:

$\mathbf{SO_4^{2-} > F^- > Cl^- > NO_3^-}$

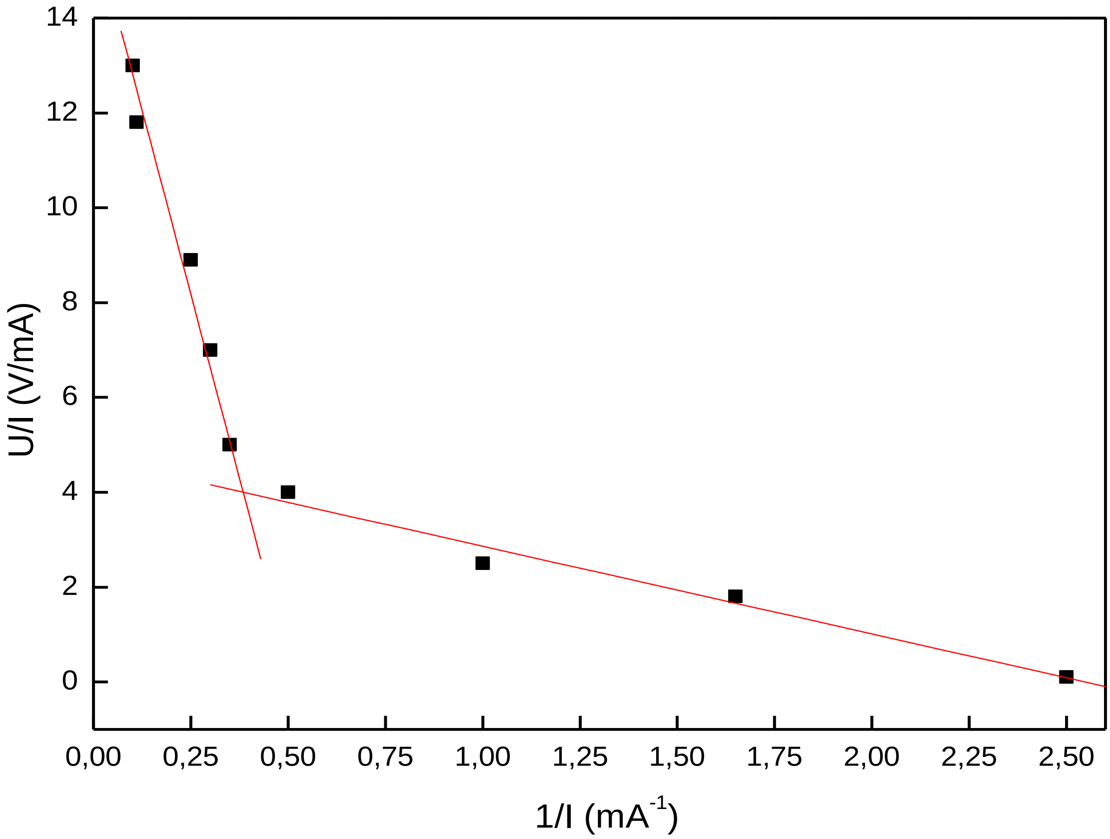

**Figure 3.** variation of U/I as a function of 1/I

The difference in selectivity of the membrane with respect to the anions is attributed to the nature nano-filtration membrane which better retains bivalent ions, to the size of the ions and to the hydration energy (Samatya *et al.,* 2006).

For Conductivity, Alkalinity and Hardness, the obtained rejection rates exceed 93%. According to these results, the water quality obtained by nano-filtration is extremely demineralized and is aggressive; the remineralisation is therefore necessary.

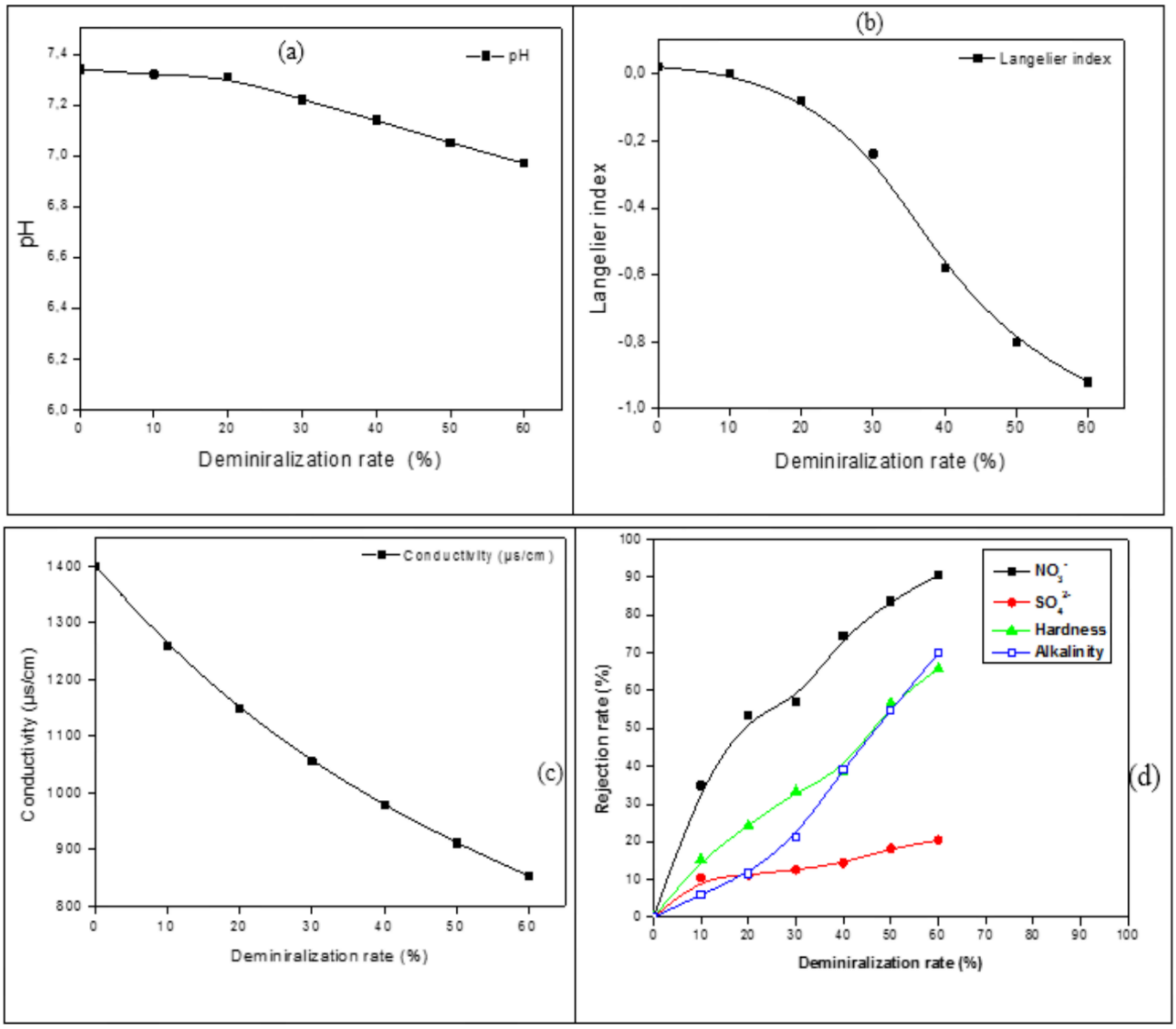

**Figure 4.** (a) pH, (b) Langelier index, (c) Conductivity, (d)Rejection rate of Nitrates, Sulphate, Hardness and Alkalinity as a function of DR.

**Figure 7** shows the variation of conductivity, pH, Cl$^-$ and $NO_3^-$ rate for treated water a function of recovery rate and for a pressure of 10 bars. To study the effects of the recovery rate on the water quality produced by nano-filtration and to avoid the precipitation phenomenon, 0.2 g/m3 of an anti-scaling agent (AF200) is added. The results show a slight increase in all the other physico-chemical parameters in the permeate but they remain largely lower than the Moroccan standards and those recommended by the WHO. This comportment is due to the increase of the salinity of the raw water by recycling of the concentrate from the membranes in the feed tank. The precipitation in the concentrate occurred at conversion rate of 89%. The nitrate concentration in the permeate for the recovery rates reached does not exceed 25 ppm.

**Table 4** gives the physicochemical quality of the treated water by electrodialysis for a demineralization rate of 15% and recovery rate of 98%, and by nanofiltration for a pressure of 10 bars and a recovery rate of 89%. According to the results obtained, the water quality obtained by electro-dialysis is satisfactory and does not need a remineralization. On the other hand, the produced water by nanofiltration is much demineralized and the remineralization for the nanofiltrated water is mandatory.

**Table 5** gives the operating cost of the treatment, which includes pumping energy, treatment energy and consumption of reagents without taking into account the replacement of the membranes.

For electrodialysis operation the cost is 0,061 €/m$^3$, and for the nanofiltration operation, the cost of the treatment is 0,046 €/m$^3$, but if we take the reminirelization step into consideration for nanofiltration, the cost of treatment is similar which is in agreement with the costs found by Elazhar and coworkers and Lahnidi her team for the defluoridation of water whose salinity of the raw water is close to the salinity of our water.

Technically, the strong points of the electro-dialysis are its flexibility with regard to the seasonal variations of the nitrate content and the robustness of the membranes compared to the membrane of NF, whereas the strong points of NF are its costs that appear lower and its simplicity use for small installations.

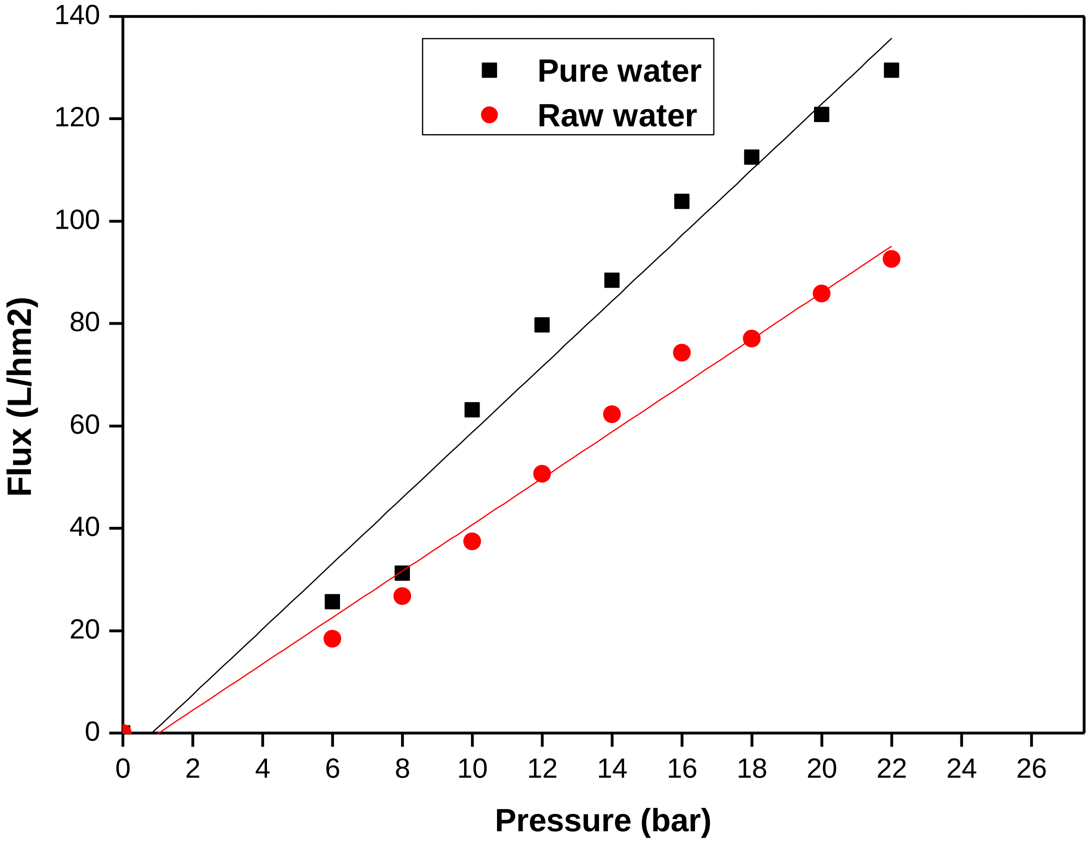

**Figure 5.** Variation of flux in raw water and pure water as function of pressure

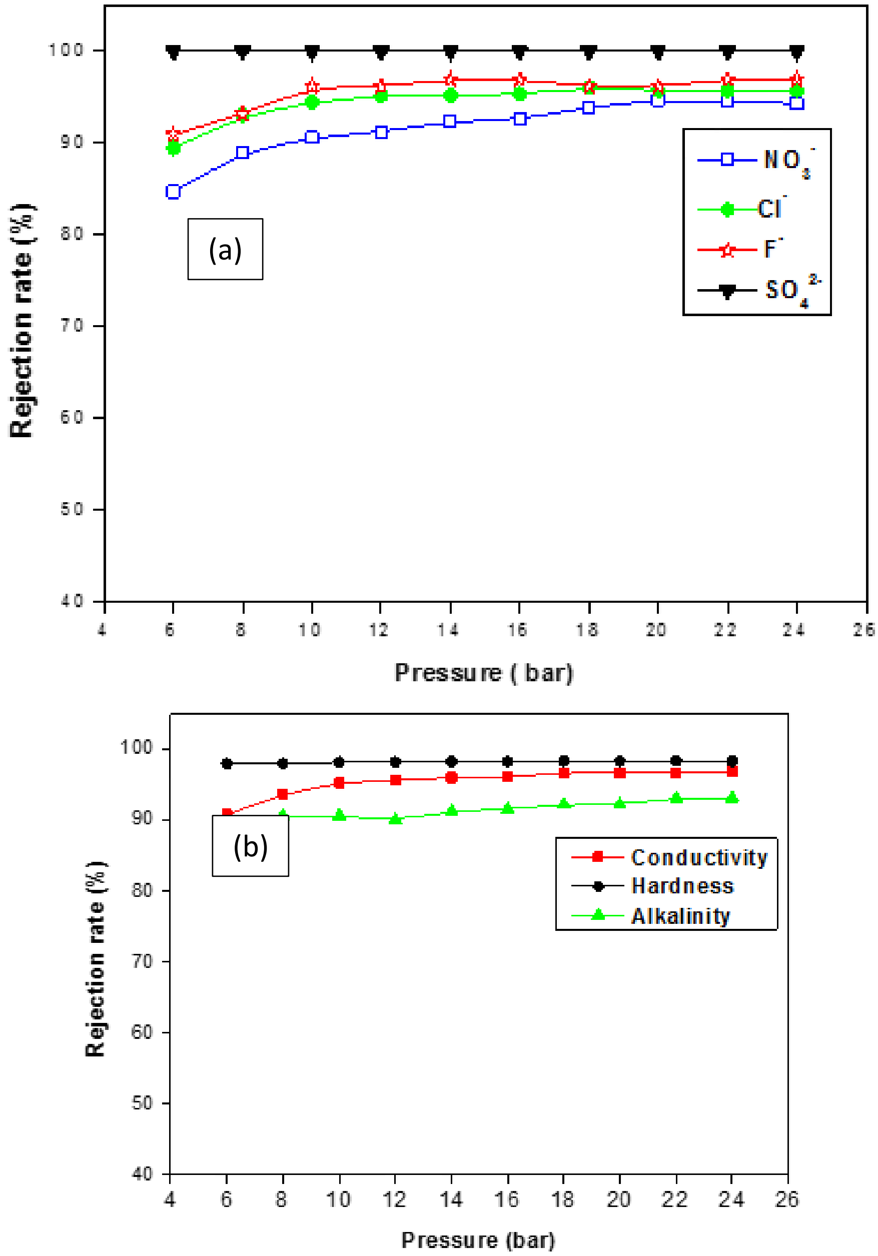

**Figure 6.** Rejection rate of physicochemical parameters of the treated water as a function of pressure: (a) effect of ion, and (b) conductivity, hardness, and alkalinity

**Table 4.** Characteristic of the treated water by Electrodialysis and Nanofiltration

| Parameters | Nanofiltration Permeate | Electrodialysis Permeate |
|---|---|---|
| **T, °C** | 31.3 | 31.7 |
| **Conductuvuty, µS/cm** | 101 | 902.7 |
| **pH** | 6.34 | 7.32 |
| **Hardness, °F** | 1.58 | 43.95 |
| **Alkalinity, °F** | 3.7 | 22.7 |
| **$SO_4^{2-}$, mg/L** | 1.1 | 204 |
| **$NO_3^-$, mg/L** | 24.3 | 50 |
| **Langelier index** | - | -0.06 |

**Table 5.** Operating cost of the treatment for electro-dialysis and nano-filtration operations.

| | Pumping energy | Treatment energy | Consumption of reagents | Treatment Cost |
|---|---|---|---|---|
| **Electrodialysis** | 0,037 €/m$^3$ | 0,019 €/m$^3$ | 0,0019 €/m$^3$ | 0,061 €/m$^3$ |
| **Nanofiltration** | 0,047 €/m$^3$ | - | 0,0028 €/m$^3$ | 0,046 €/m$^3$ |

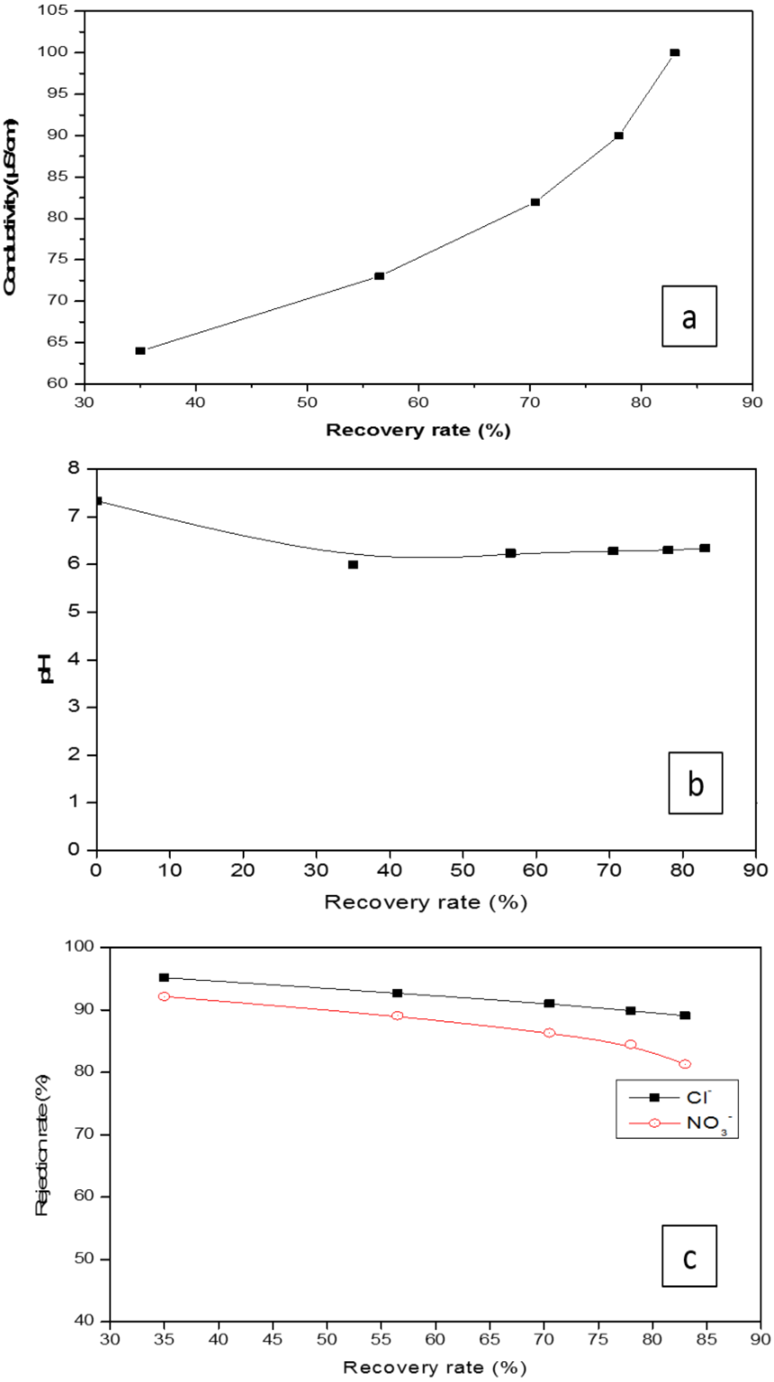

**Figure 7.** Variation of conductivity, pH, Cl$^-$ and NO$_3^-$ rate as a function of recovery rate: (a) conductivity, (b) pH, and (c) rejection rate.

## 4. CONCLUSION

This research work focused on the study of denitration of groundwater from boujaad, morocco. the feasibility of electrodialysis and nanofiltration in the reduction of nitrate ions in groundwater is confirmed. The industrial-scale treatment by an electro-dialysis pilot of using the ACS / CMX membrane couple has made it possible to define the operating conditions, namely a demineralization rate of 15%, a recovery rate of 98% by maintaining the pH of the brine compartment at 7.the physico-chemical quality of the treated water is satisfactory. The operating cost is 0.061 €/m$^3$. The study of denitration by nanofiltration on an industrial pilot and in continuous mode showthe feasibility of this technology and allow the elimination of 90% of the nitrates and reach a conversion rate of 89% by using an anti-scaling (AF200). The calculated operating cost is 0.046 €/m$^3$. On the other hand, the treated water obtained by nanofiltration needs a demineralization before consumption. In terms of comparing the two technologies, it is difficult to choose between these two processes for denitration of groundwater. Their performances are substantially closer, however there is a slight advantage for the nanofiltration in terms of operating cost but electro-dialysis is favored by his flexibility with regard to the variations of the nitrate content and the robustness of the membranes with respect to those of nanofiltration, and there, mineralization is not required before distribution.

## 5. AUTHORS' NOTE

The author(s) declare(s) that there is no conflict of interest regarding the publication of this article. The authors confirm that the data and the paper are free of plagiarism.

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