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Nitrate removal of groundwater by reverse osmosis, nanofiltration and electrodialysis: performances and cost comparison

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A B S T R A C T

The study was conducted to evaluate the comparison of the three technologies, electrodialysis (ED), nanofiltration (NF) and reverse osmosis (RO) in nitrate removal from groundwater on the basis of performances, capital expenditure (Capex) and operating cost (Opex). The comparison was estimated for an industrial plant having a capacity of 2,200 m^3/d (100 m^3/h) corresponding to a water consumption for 50,000 capita following the Moroccan considerations in rural medium (consumption for domestic use of 50 L/d and per capita). The first part of the study was carried out for the comparison of the performances in terms of nitrate removal and recovery rate of the three membrane-based technologies in nitrate removal from Moroccan groundwater. This study confirms the performances of ED, NF and RO in nitrate removal and shows that these performances are comparable. The second-part is dedicated to estimating and calculating Capex, Opex and the specific energy consumption for the three technologies. The cost assessment shows that ED appears to be slightly cost effective for nitrate removal from groundwater than RO and NF.

Keywords: Nitrate removal; Nanofiltration; Electrodialysis; Reverse osmosis; Drinking water; Specific energy consumption; Capital expenditure; Operating cost

1. Introduction

Pollution of groundwater and surface water by nitrate is a planetary preoccupation. It is a common concern of industrial and developing countries. The major causes of nitrate excess in the environment are agricultural runoff, poorly or untreated animal and human wastes, delays in the mastery and generalization of sanitation systems. And the intensive use of pesticides and fertilizers in agriculture. Contamination of drinking water by nitrate can promote the spread of serious diseases such as irritation, allergies, abortion, cancer and chemical poisoning.

To avoid the risks of nitrates and due to the water scarcity and recurrent draught, the main treatments for removing nitrates from water are either biological (denitrification) or physico-chemical (denitratation).

Water in many regions of Morocco usually exceeds the acceptable standards for nitrate, in the southwest of Morocco especially the plateau of Boujaad, where the work was conducted, the nitrate concentration exceeds 85 mg/L and the water is brackish. To prevent the adverse effects of nitrates, the World Health Organization (WHO) has recommended 50 mg/L as threshold not to be exceeded for drinking water. The guide value preconized by WHO is 25 mg/L [1]. The same standards are adopted in Morocco by National Office of Electricity and Drinking Water (ONEE). To reduce

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nitrate to safe level of drinking water, several methods have been applied, mainly ion exchange [2,3], biological denitrification [3,4], catalytic reduction [4].

In comparison with these methods, the membrane-based technologies offer many advantages including no need to use chemicals [5] and their high and specific membrane selectivity, capacity, operational flexibility and cost-effectiveness [6]. Nanofiltration (NF), reverse osmosis (RO) and electrodialysis (ED) process for nitrate removal were largely studied in drinking water [7]. Different technical aspects of the development of NF, RO and ED in nitrate removal were studied and optimized. Schoeman and Steyn [5], used RO process in nitrate removal, they mentioned that 96%–98% of the nitrate was removed at a recovery rate of 50% for a transmembrane pressure (TMP) of 13.75 bar with permeate concentrations below 22 mg NO_3^-/L. Another study, carried out by El-Ghzizel et al. [8], showed that the use of NF process, is capable of reducing the nitrate concentration from 68 to 18 mg NO_3^-/L, is recovery rate of 75% for a TMP of 5 bars. Touir et al. [9], applied ED to remove nitrate from groundwater treatment, they optimized the operating conditions, namely a demineralization rate of 15%, a recovery rate of 98% by maintaining the pH of the brine compartment at 7, at these conditions the final nitrate concentration was at 50 mg/L according to WHO.

Despite the treatment efficiency of NF, RO and ED technologies in water treatment, some limitations remain in using membranes process. The first and major obstacle for the application of these technologies is the brine produced which causes adverse environmental impacts and limit the water recovery in ED and RO. The second reason is the rapid decline of the permeate flux over time as a result of membrane fouling for NF and RO. However, the cost is the major factor when implementing membrane technologies. Several factors affect the unit product cost for membrane facilities, such as, the operating conditions, the system and module design, the module number and membrane replacement, the operation conditions and the plant capacity [10]. Of all these parameters, the membrane flux is broadly the most influential of the application costs as it is a direct measure of productivity and determines the operating TMP and the membrane area. Pilot studies are critical to the success of a membrane project as they provide a reliable basis for evaluating the effect of the operating parameters on the cost and the performance of a large-scale installation [11]. In the literature, Touir et al. [12], have estimated the cost comparison of denitratation of groundwater using a pilot plant of ED and NF processes, they estimated the cost to be 0.061 and 0.046 €/m^3 for ED and NF respectively and they estimated the cost of the produced potable water to be 0.12 and 0.13 €/m^3 respectively. Moreover, a preliminary study of the cost comparison on the basis of the quality of the treated water was carried out on NF and RO [13]. But as far as we know, no study has included the comparison of these three membrane technologies in the removal of nitrates from groundwater. This is the essential purpose of this work. Comparison will be conducted to evaluate efficiencies of the plant using NF, RO and ED in nitrate removal from a groundwater in the region of Boujaad (Morocco). This comparison will concern performance of the plant, the water quality and finally, the overall cost (capital expenditure (Capex), operating cost (Opex) and the specific energy consumption (SEC)) including membrane module number, membrane replacement, operation conditions and plant capacity.

2. Experiment

2.1. Feed water

The effluent of the current study is taken from a groundwater in the region of Boujaad in the centre of Morocco. The analytical results of the untreated water are shown in Table 1, the water is brackish and the content of nitrate (119 mg/L) exceeds the standard recommended by WHO which is 25 mg/L.

2.2. Description of the electrodialysis pilot plant

ED operations are carried out with the following couple of membranes: NEOSEPTA ACS as anion-exchange membrane and NEOSEPTA CMX-Sb as cation-exchange membrane, all manufactured by Tokuyama Co. Table 2 gives the characteristics of ACS and CMX ion-exchange membranes. The pilot plant was already described [14,15] (Fig. 1). The ED pilot is operated in a continuous mode with inverting polarity of 20 min each, and flushed periodically with hydrochloric acid to prevent scaling and fouling. Previous work conducted in our laboratory [16] which dealt with feasibility experiments have selected the couple ACS/CMX membranes for nitrate removal from groundwater.

2.3. Description of the RO/NF pilot plant

The experiments are performed on an NF/RO pilot plant (E 3039) supplied by TIA Company (Applied Industrial

Table 1
Characteristics of the feed water

Parameters	Feed water
T (°C)	23
pH	7.3
Electric conductivity (μS/cm)	1,330
NO_3^- (mg/L)	119
Cl^- (mg/L)	536
F^- (mg/L)	1.2
Hardness (°F)	55.5
Alkalinity (°F)	30
Langelier Saturation Index (LSI)	0.02

Table 2
Characteristics of the membranes used

Membrane type	ACS	CMX-Sb
Thickness (cm)	0.17	0.18
Electrical resistance (Ω/cm^2)	2.0	3.0
Exchange capacity (meq/g)	1.8	1.65

Technologies, France) and already described [17,18] (Fig. 2). The pilot plant is equipped with two identical spiral wound modules operating in series. Each module contains one element, the TMP applied over the membrane is varied from 5 to 70 bar with manual valves. Table 3 gives the characteristics of the commercial membranes used.

For three operations ED, NF and RO, experiments are performed at 29°C. 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 [19]. Some other parameters are followed such as:

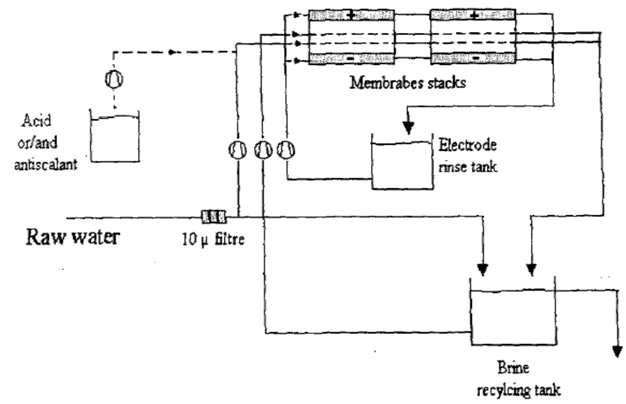

Fig. 1. Schematic diagram of ED pilot plant.

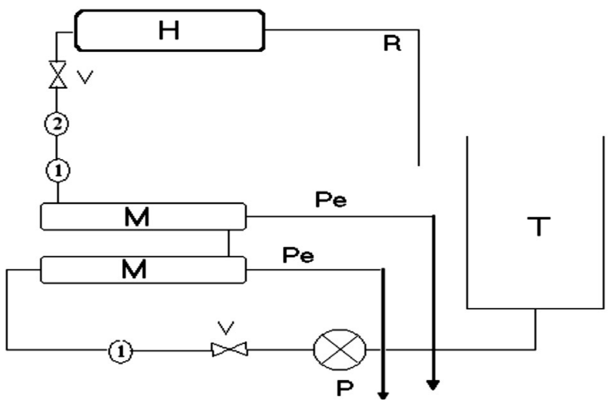

Fig. 2. Schematic diagram of RO/NF pilot plant. P: Feed pump; V: Pressure regulation valves; M: Reverse osmosis/nanofiltration module; Pe: Permeate recirculation; R: Retentate recirculation; H: Heat exchanger; 1: Pressure sensor; 2: Temperature sensor.

Table 3
Characteristic of the membranes used

Membranes	NF90	BW40-40
Cut-off (Da)	90	40
Surface (m^2)	7.6	7.6
Material	Polyamide	Polyamide

For ED:

Removal rate R (%) of each ion: $$R(\%) = \frac{(C_0 - C_p)}{C_0} \times 100 \quad (1)$$

where C_p and C_f are, respectively, permeate and initial concentration of the ion considered.

Demineralization rate DR (%): $$\mathrm{DR}(\%) = \frac{(E_f - E_p)}{E_f} \times 100 \quad (2)$$

where E_p and E_f (μS/cm^2) are the electric conductivity of permeate and feed water respectively.

Specific energy consumption (SEC): $$E(\mathrm{kWh/m^3}) = \frac{U \cdot Q_i}{1{,}000} \quad (3)$$

where U is the potential difference applied to each unit cell (V) and Q_i the amount of electricity needed per cubic meter of water (Ah/m^3).

For RO and NF:

Permeate flux, J_p (L/h m^2): $$J_p = \frac{Q_p}{S} \quad (4)$$

where S (m^2) and Q_p (L/h) are respectively the surface area of the membrane and permeate flow.

Recovery rate Y (%): $$Y(\%) = \frac{Q_p}{Q_f} \times 100 \quad (5)$$

where Q_f and Q_p are the feed and the permeate flow rate respectively.

Specific energy consumption (SEC): $$\mathrm{SEC}(\mathrm{kWh/m^3}) = \frac{(P \cdot 100)}{(\eta \cdot Y \cdot 36)} \quad (6)$$

where P is the applied TMP (bar), η is the global pumping system efficiency and Y is the recovery rate.

2.4. Capex

Capex includes the cost of the treatment process itself and on-site construction. The different costs that contribute in the Capex are:

- Cost of construction and building.
- Cost of pretreatment.
- Cost of NF, RO and ED group.
- Cost of auxiliary equipment.
- Cost of various services.

These costs are based on real purchase prices and the assumptions given above, and may change as these assumptions change. Moreover, all Capex components are annualized considering an amortization factor calculated as a function of the interest rate for capital investments (6.7%) and for a design life of the plant of 20 y. Total Capex of the various materials have been evaluated according to the reported international price and of the local market price, which is the price that has been evaluated net of tax and except expenses of the customs.

2.5. Annual Opex

The Annual Opex covers all expenditures incurred after plant commissioning and during actual operation. These include:

2.5.1. Amortization or fixed charges

When the required funds of the project have to be borrowed, there will be an interest charges for the use of the required funds. This item accounts for annual interest payments for capital cost. It is obtained by multiplying the capital cost by an amortization factor a, which is given by the study of Tsiourtis [20]:

$$a = \frac{i(1+i)^n}{(1+i)^n - 1} \tag{7}$$

where 'i' is the annual interest rate (%) and 'n' is the life time of the facility.

2.5.2. Operating and maintenance (O&M) costs

This includes the operation and maintenance staff cost, the spare costs etc. This cost shall be expressed on a yearly base for each item for all the commercial operation period [21]. The annual O&M costs are estimated at 20% of the plant annual payment.

2.5.3. Membrane replacement

For RO and NF processes, membrane replacement rate depends largely on raw water quality. Replacement rate may vary between 5% and 20%/y. The lower bound applies to low salinity brackish water and the upper would reflect the high salinity seawater [22]. Membrane replacement cost is estimated for a 1-y period and divided by the quantity of water to be produced during the year to determine the overall water cost [23].

In our case, the replacement rate taken is 5% [24]. The membranes life is evaluated to 5 y for the two processes.

3. Results and discussion

3.1. Performances of ED, NF and RO in nitrate removal

3.1.1. Case of ED

Nitrate removal by ED from a groundwater of Boujaad depends strongly on the water quality and the characteristics of the membranes. Fig. 3 gives the variation of the electric conductivity of the dilute and the nitrate removal, hardness, alkalinity and sulphate as function of the demineralization rate (DR).

The results show that the electric conductivity of the dilute compartment decreases with the increase of demineralization rate (DR) and reaches the minimum value of 850 μS/cm corresponding to 60% of DR. During this ED operation, and for the same, demineralization rate (DR), the removal rate obtained of alkalinity, hardness and sulphate are 70%, 68% and 20%, respectively. At the same time, the nitrate removal rate achieved 90%. This result is attributed essentially to the nature of the AEM used which is preferably selective to monovalent anions. Based on these results and taking care of respecting the limit of nitrate content to not exceed the threshold set by WHO, the optimal operating conditions of ED are demineralization rate (DR) of 35% which allows a nitrate removal rate of 65%, which corresponds to the reduction of nitrate concentration in the feed water from 119 to 50 mg/L in the permeate compartment, these values were optimised in a previous work [25].

3.1.2. Case of NF

Fig. 4 shows the rejection of nitrate, chloride and sulfate as function of the TMP. Fig. 4 shows also the evolution of some physicochemical parameters (alkalinity, *E*

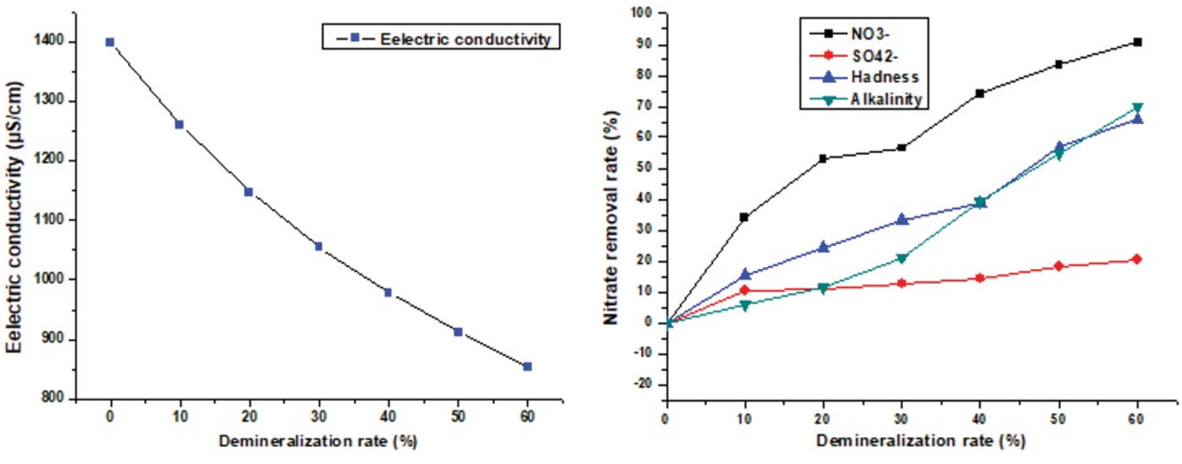

Fig. 3. Variation of the electric conductivity of the dilute and nitrate removal rate, alkalinity and sulfate as a function of demineralization rate (DR).

and hardness) with TMP. Results reveal that the rejection of nitrate and chloride have increased slightly with the increase of TMP and reached a maximum value of 94% and 95% at TMP of 24 bar, however, sulfate is totally rejected (R = 100%). Results of E, hardeness and alkalinity show that the rejection reache is close to 98%. According to these results, the quality of the produced water by NF90 water is not satisfactory, since all the water parameters are lower than the standards. The optimization of TMP leads to set the TMP at 8 bar with remineralisation step using a lime saturator, this is obligatory in order to obtain a water quality according to WHO.

Fig. 5 shows that for a constant TMP (8 bar), increasing the recovery rate leads to an increase in electric conductivity. At a recovery rate of 83% and the feed water concentration of NO_3^- at 119 mg/L, the nitrate concentration in the permeate does not exceed 24 mg/L. Based on these results, the optimal operating conditions using NF90 are the pressure of 8 bar and the recovery rate of 83%.

3.1.3. Case of RO

Figs. 6 and 7 show that the performances of the RO membrane BW40-40 exhibits similar behaviour than that of NF90 in terms of variation of ions rejection as function of TMP. The rejection of nitrate, chloride, sulphate, electric conductivity, hardness and alkalinity is in the range 83%–100%. However, for recovery rate ranged in 35%–83%, electric conductivity of the RO permeate and rejection of nitrate is ranged between 36–74 μS/cm and 93%–97% respectively. According to these results, the optimal operating conditions of RO using BW40-40 membrane are TMP of 8 bar and the recovery rate of 83%. Comparatively with previous studies, the results demonstrated that the

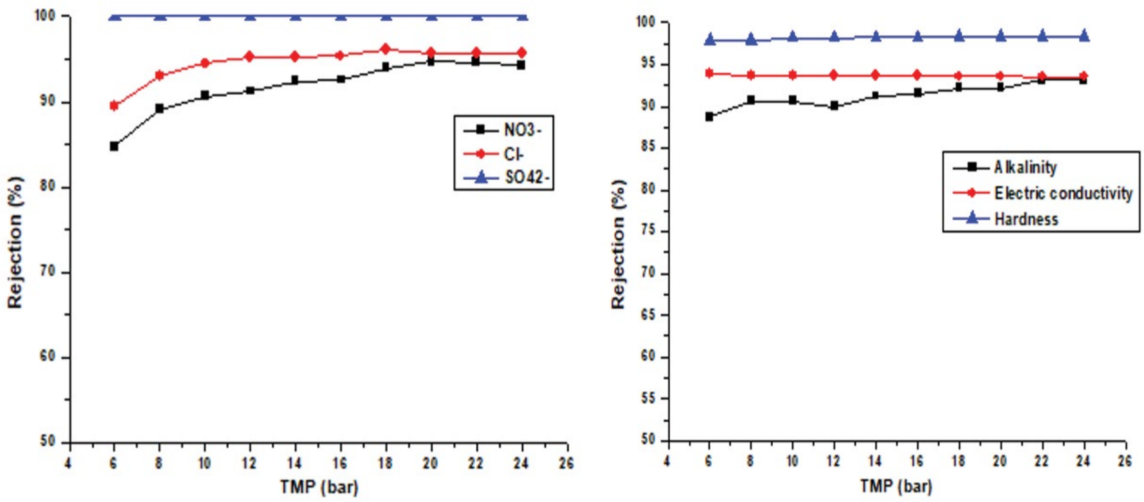

Fig. 4. Rejection of nitrate, chloride, sulfate, electric conductivity alkalinity and hardness vs. TMP.

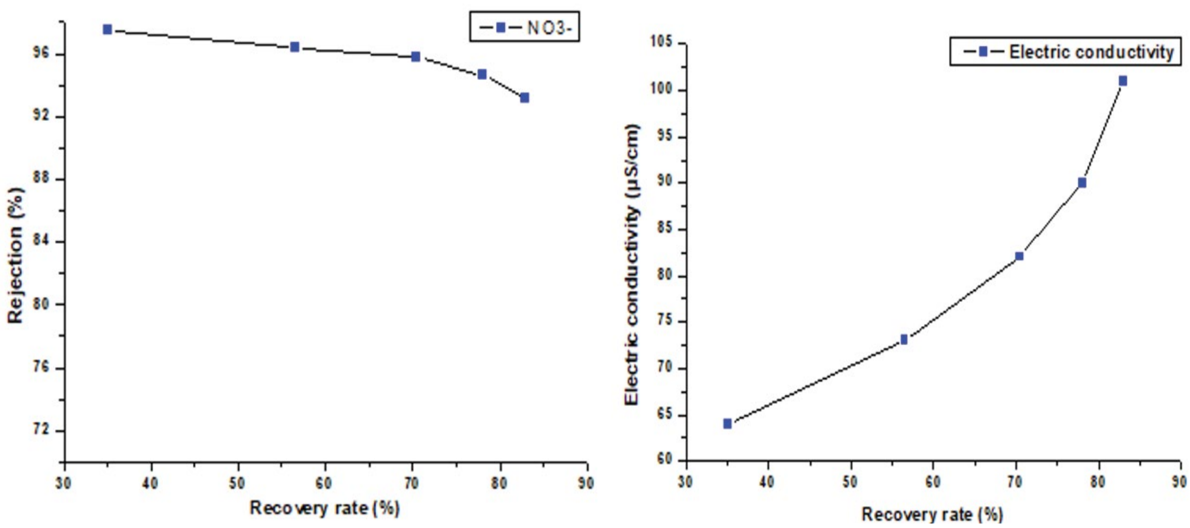

Fig. 5. Rejection of nitrate and electric conductivity as function of recovery rate.

operating TMP and the recovery rate in the RO stage has a crucial impact on the quality of the produced water [7]. In addition, to obtain the water quality according to WHO a remineralisation step of RO permeate is obligatory. Table 4 shows the comparison of the water quality obtained by the three technologies under optimal conditions.

3.2. Comparison of the performances of ED, NF and RO

The performances of the three technologies in nitrate removal from groundwater of Boujaad are described and compared briefly. The characteristics of the produced water before and after the treatment by ED, NF and RO (with and without remineralization step using a lime saturator) are

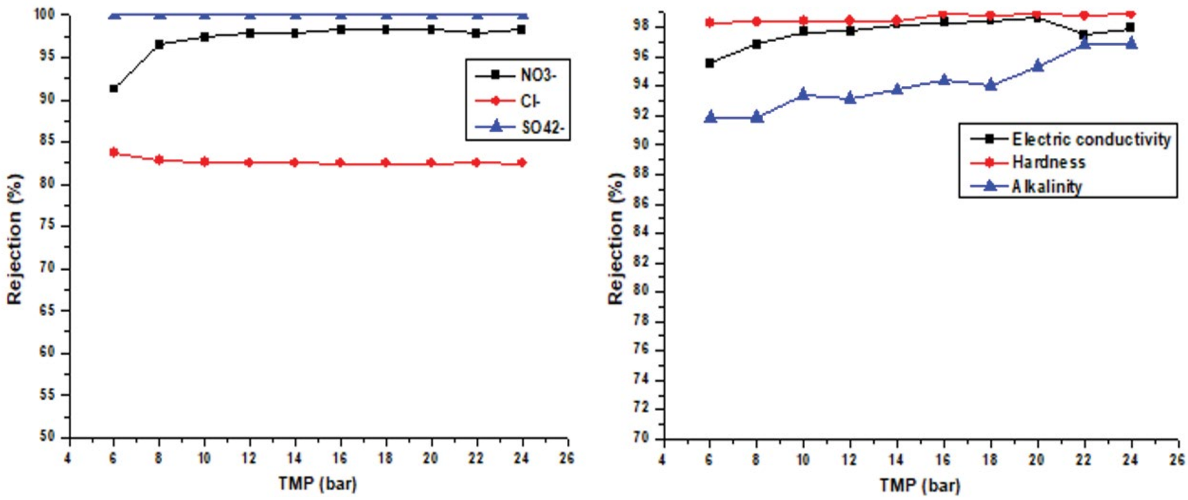

Fig. 6. Rejection of nitrate, chloride, sulfate, electric conductivity, alkalinity and hardness as function of TMP.

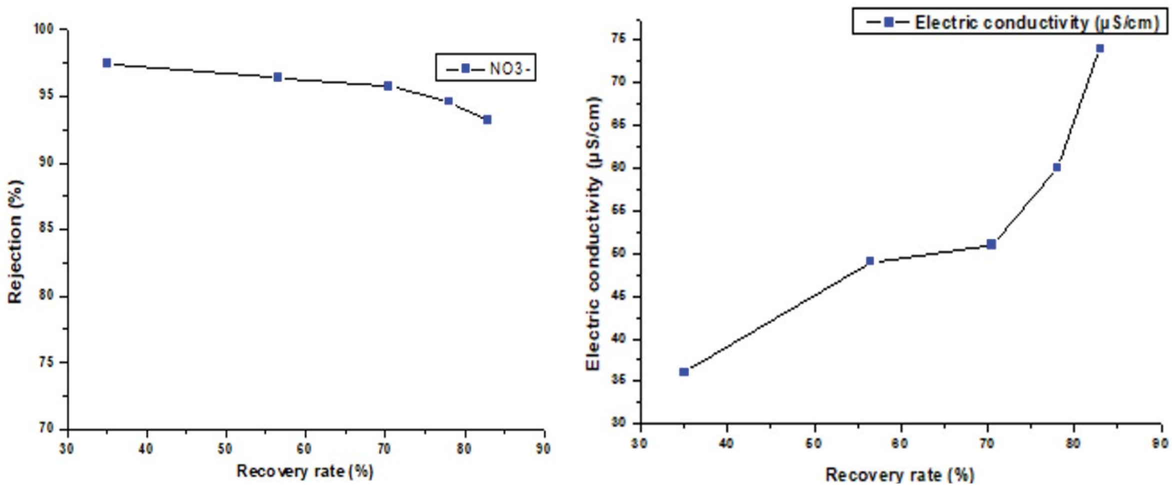

Fig. 7. Nitrate rejection and electric conductivity vs. recovery rate.

Table 4
Characteristics of the produced water before and after treatment by the three technologies

Parameters	Raw water	Treated water (ED)	Treated water (NF)	Mixed water (NF)	Treated water (OI)	Mixed water (OI)
E (µS/cm)	1,330	902.7	90	218.24	45	225.2
Hardness (°F)	55.5	43.95	1.58	4.35	1.53	7.356
Alkalinity (°F)	30	22.7	3.7	6.20	2.7	8.16
NO_3^- (mg/L)	119	50	14.2	33.33	4.3	30.28

presented in Table 4. The results show that the water quality obtained by ED complies with the Moroccan standards and doesn't need a remineralisation. However, the water quality obtained by RO and NF in the optimal conditions (TMP of 8 bars; recovery rate of 83%), even the nitrate content meets the standards, the water quality is not satisfactory and a remineralisation step is needed (Fig. 8). After remineralization step with raw water, the produced water quality is satisfactory in terms of electric conductivity and nitrate content.

3.3. Proposed design

The proposed design for the NF and the RO unit is shown in Fig. 8. The main treatment unit comprises:

- *Pre-treatment*: Composed of sand filters of 5 μm, with capacity of 125 m^3/h;
- *NF and RO group*: The calculation of the NF/RO group was carried out on the basis of the experimental results obtained;
- *Post-treatment*: Includes the remineralisation with raw water.

3.4. Cost comparison of the three technologies plant: ED, NF and RO

3.4.1. Description of ED, NF and RO plants

For the three technologies ED, NF and RO the design of the plant is performed for a capacity of 2,200 m^3/d (100 m^3/h) corresponding to a water consumption for 50,000 capita following the Moroccan considerations in rural medium (consumption for domestic use of 50 L/cap/d).

The proposed design for ED unit is shown in Fig. 9. The characteristic of well feed water is shown in Table 1. The raw water is pumped with low pressure pumps from the storage tank to the pretreatment unit. The pre-treated water is pumped up with low pressure pumps to the ED unit. The treated water is stored in the product water tank T3 and then distributed to the city of Boujaad. No post-treatment is suggested here because of the good quality of the produced water. The brine is collected in the tank T4 and discharged on a surface for spreading and evaporation.

For the proposed design of NF and RO unit is shown in Fig. 10. The drilling water is pumped from the reservoir T1 (150 m^3) to the pretreatment section by low pressures pumps (P1 and P2). The pretreated water is pumped up with high pressure pumps to the NF unit. Due to its low mineralization, permeate is then remineralized according to the proposed design shown in Fig. 8. The treated water is stored in the product water tank T3 and then distributed to the city of Boujaad. The produced brine is collected in the tank and discharged on a surface for spreading and evaporation.

3.4.2. Cost comparison

The evaluation of the cost unit of the produced cubic meter is based on the calculation of the Capex and Opex, including cost of the membrane replacement under the optimal conditions set of a production capacity estimated to 100 m^3/h with daily capacity of 2,200 m^3. Capex and Opex of three technologies ED, NF and RO are assessed and compared. Table 5 summarizes a comparison between the three technologies of the estimated Capex, Opex and SEC. Table 6 gives the required data cost for the three technologies.

According to these results, the cost of the three technologies are very close. Moreover, the NF annual Opex which is around €8,850.96 appears to be excessive with regard to RO (€12,760.362) and ED (€10,472.04) with regard

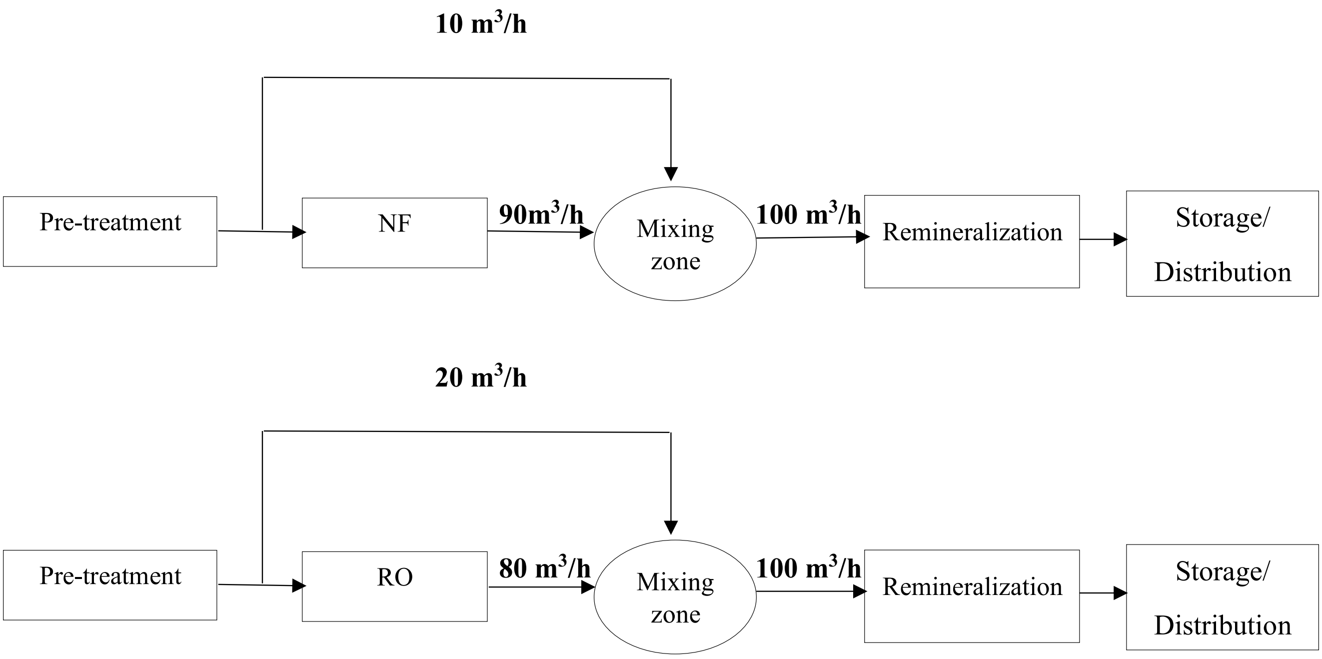

Fig. 8. Proposed design for nitrate removal by NF and RO.

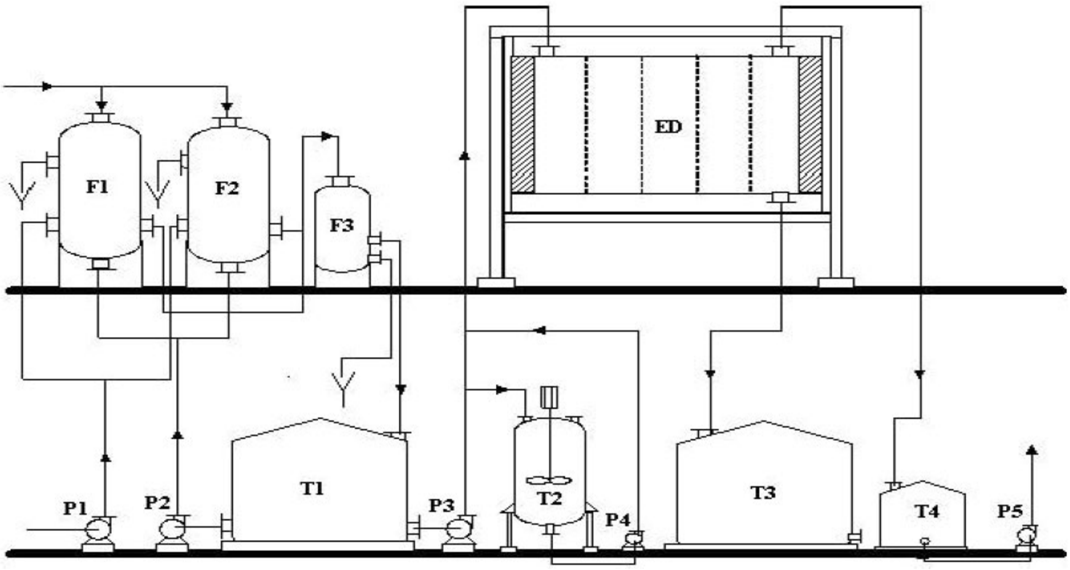

Fig. 9. Scheme of ED plant for nitrate removal. F1 and F2 Sand filters, F3 Cartridge filter, T2 Sequestering preparation tank, T3 Treated water tank, T4 Brine tank.

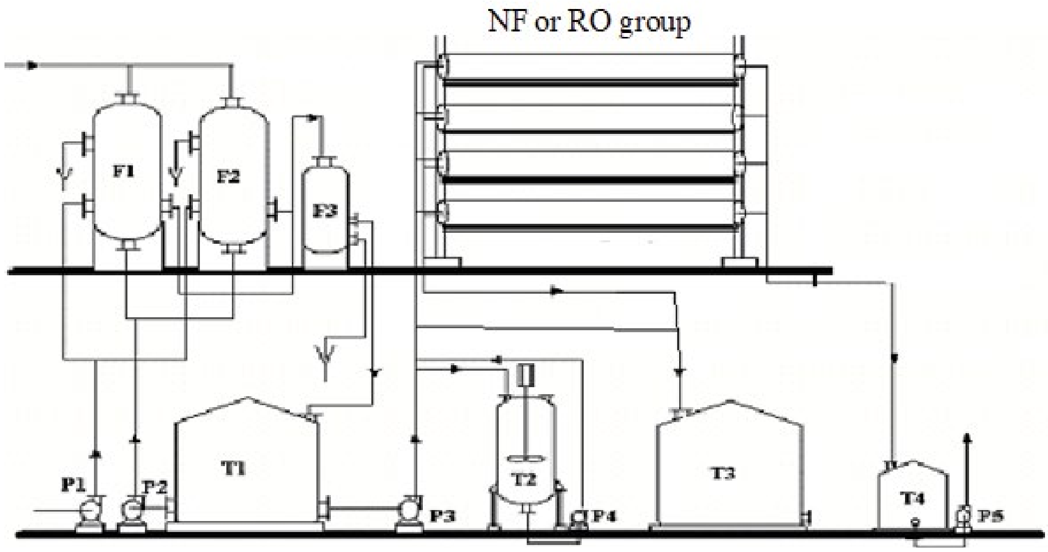

Fig. 10. Design of NF and RO plant for nitrate removal. F1 and F2 Sand filters, F3 Cartridge filter, T2 Sequestering preparation tank, T3 Treated water tank, T4 Brine tank.

Table 5
Capex, Opex and SEC/m^3 for the three technologies

Process	Capex (€)	Opex (€/m^3)	SEC (€/m^3)
ED	116,356	10,472.04	0.0361
NF	98,344	8,850.96	0.0247
RO	141,781.8	12,760.362	0.0247

Table 6
Data cost for the three technologies

	NF	RO	ED
Membrane module cost (€)	192,375	182,590	171,000
Replacement of membrane cost (€)	47.5	47.5	8,550
Production capacity (m^3/d)	2,400	2,400	2,400

to the energy in Morocco, where the average price of energy is 0.085 €/kWh. In our case, the energy consumption for the three technologies NF, RO and ED is in the range of 0.0247 and 0.0361 €/m^3 respectively. The difference can be attributed to the configuration of the membrane modules used which certainly will decrease the costs. However, it remains difficult to analyse and compare the costs for the three technologies for nitrate removal from the groundwater of Boujaad. Nevertheless, practically and experimentally the ED unit has the advantage of flexibility with regard to seasonal variations in the nitrate content, the robustness of ion-exchange membranes compared to NF and RO membranes, the low-cost pretreatment and limited post-treatment in most cases to chlorination, which is not the case for NF and RO that use the complicated pre- and post-treatment. Among the advantages of NF and RO compared to ED is its simplicity which is of particular importance for small installations, as well as the easy change of membranes than that of ED which appears more difficult. However, comparing the advantages and the disadvantages of these three technologies we conclude that the ED is the more suitable for nitrate removal than NF and RO.

4. Conclusion

The nitrate removal by ED, NF and RO was conducted on a groundwater of Boujaad (centre of Morocco) using two pilot plants. The study confirms the performances of the three technologies in nitrate removal and these performances appear to be comparable. A drinking water with outstanding quality can be easily produced by these three technologies. The total cost of ED, NF and RO was estimated by calculation of the Capex and Opex taking into account the depreciation factor, membrane replacement rate and plant capacity. The Capex for a plant of production capacity 2,200 m^3/d is estimated to be €116,356, €98,344 and €141,781.8 for ED, NF and RO respectively. The calculated annual Opex is 10,472.04, 8,850.96 and 12,760.362 €/m^3 for ED, NF and RO respectively and the SEC calculated is 0.036 €/m^3 for ED and 0.024 €/m^3 for NF and RO. The expected potable water production cost is calculated to be 0.019 €/m^3 for ED, 0.16 €/m^3 for NF and 0.10 €/m^3 for RO unit. Therefore, it's difficult to analyse and compare the costs for the three processes. However, comparing the cost of nitrate removal according to the performance of the three technologies, and taking into account various other aspects specific to each technology, ED unit seems to be more suitable for nitrate removal than NF and RO.

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