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Comparison of diverse direct and hybrid membrane processes for nitrate removal from brackish water

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ABSTRACT

The purpose of this paper is to evaluate the efficiency of nitrate removal from ground water with membrane processes, including direct: electrodialysis (ED), reverse osmosis (RO), nanofiltration (NF), and hybrid process coupling NF with RO/ED. Both energy consumption and water product quality were considered to assess process efficiency of direct and hybrid systems. The blending strategy was adopted only for permeates that are lower than the standards for drinking water in terms of nitrate by mixing the permeate with brine. Results indicate that the nitrate removal efficiency in RO system and ED was about 95%, 3% and 91%, 48% more than that of the NF system. Thereby, the energy and cost efficiency of ED is relatively higher than RO. Considering the nitrate removal efficiency of NF-RO and NF-ED hybrid system, the results show that the NF-ED quality of nitrate content is better than that of NF-RO. In addition, the water recovery of NF-ED is relatively higher. However, the energy consumption and energy cost of both NF-ED and NF-RO hybrid systems are similar with a particular benefit of NF-ED. However, the portion of blending rate with NF-RO permeate is equal to 65% and to 45% of RO. Having said that, the NF-RO hybrid system is technically more attractive than others to obtain water with 50 ppm of nitrate and a target recovery rate of 95.42%, despite their moderately higher energy cost. At the same time, NF-RO hybrid minimizes the fraction of brine disposal dumping to sewage, making it environmentally friendly.

Keywords: Nitrate; Nanofiltration; Recovery rate; Hybrid system; Energy consumption; Electrodialysis

1. Introduction

Ensuring appropriate quality of drinking water is a major task nowadays because of the expansion in pollution of water bodies. Nitrate is vital pollutants that undermine living life forms, especially people. Nitrate ions can enter into aqueous environment by weathering of nitrate-rich minerals and as through anthropogenic actions, for example, domestic wastewater and industrial drains or agricultural surface run-offs [1–3]. However, common nitrate values in contaminated waters is in the range of 10 to 600 mg/L, reported in different locations, such as China [4], India [5], Spain [6], and Morocco [7] According to World Health Organization (WHO) guidelines, the nitrate concentration

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in drinking water cannot be higher than 50 ppm [8]. Likewise, the Moroccan legislation established a maximum allowed concentration of nitrates at 50 mg/L in drinking water [9].

As was already mentioned in the literature, advanced physical–chemical treatment techniques such as reverse osmosis (RO), nanofiltration (NF), electrodialysis (ED), and electromembrane processes have the potential to be suitable techniques for removing nitrates and drinking water production [10–15].

However, these techniques have many limitations, including brine production, membrane life time, and permeate fluxes that is primarily affected by fouling at the membrane surface under normal operating conditions [10,16–18]. To minimize chemical fouling and to operate with higher water recovery ratios, the use of chemicals (antiscalents) was largely practiced [19].

For the same reason, hybrid systems have been recently used for different applications. In particular, several hybrid NF-RO filtration schemes in a specific configuration have been proposed to take advantage of the intrinsic properties of NF and RO membranes (i.e., ion-ion and water-salt selectivity, respectively), leveraging the process in terms of product and waste stream composition, water recovery, and other parameters [10,20–23]. For example, it was shown that selective nitrate removal from contaminated groundwater can be achieved by applying a preliminary NF stage that favorably permeates nitrate over chloride and divalent-based minerals [10]. The NF nitrate-rich permeate is filtrated by the following RO stage and the salt-free RO permeate is mixed with the NF retentate to produce a product water with a balanced mineral composition. In another study, the authors demonstrated how recycled water suitable for irrigation can be produced from high-salinity wastewater by using the preceding NF stage to extract useful nutrients that are then mixed with the permeate of the subsequent RO stage [24]. More specifically, these studies have demonstrated how the ion–ion selectivity of the preliminary NF stage can be utilized to separate and “store” important ingredients in the feed water, which later can be mixed with the permeate of the following RO stage to achieve an overall selective process towards specific constituents.

Thus, in Morocco, until recently, no practical hybrid membrane applications have been adopted for the nitrate removal from brackish water. Further, very few study or field experiments employing NF/RO combination have been reported from Morocco [20,23].

Therefore, the aim of this paper is to compare separately the efficiency of diverse direct and hybrid membrane process of electrodialysis (ED), nanofiltration (NF), reverse osmosis (RO), nanofiltration-reverse osmosis/electrodialysis (NF-RO/ED) for the treatment of the complex ground water containing both high nitrate and salinity. The performances efficiency of both systems was evaluated in terms of improving the overall water recovery and minimizing the specific energy consumption of pilot scale ED, NF, RO and hybrid NF-RO/ED. In addition, the blending strategy was designed to stabilize the water quality in accordance with drinking water standards and to achieve a near to zero discharge.

2. Experimental

2.1. Characteristic of the feed water

The experiment is applied in this case study to individually compare the performances of ED, RO and NF membrane treating complex brackish water (BW) containing both high nitrate and salinity, which greatly exceeds the standards of Moroccan regulation for drinking water and the WHO standars. The ionic composition of the M’nasra feed BW is shown in Table 1.

2.2. Experimental setup

In this study, the pilot of RO/NF (Fig. 1) and ED (Fig. 2) were employed for nitrate removal of brackish water in M’nasra in the production mode. Pretreatment was performed using sand filter for removing suspended

Table 1
Characteristics of the M’nasra feed BW

Parameters	Feed BW	Moroccan guidelines [9]	WHO [7]
Temperature (°C)	30	35	25
Turbidity (NTU)	<3	<5	<5
Conductivity (µS/cm)	4240	<1,000	<1,000
TDS (ppm)	1,280	<500	900
pH	7.80	6.0–9.0	6.5–8.5
Alkalinity (ppm)	100	–	–
TH (°F)	85	50	–
Ca^{2+} (ppm)	237	<500	<270
NO_3^- (ppm)	470	50	50
Na^+ (ppm)	550	200	200
Mg^{2+} (ppm)	120	100	<50
Cl^- (ppm)	629	250	<250
SO_4^{2-} (ppm)	454	200	200
LSI	–0.45	–0.2 < LSI < 0.2	–

Fig. 1. NF/RO pilot.

Fig. 2. ED pilot.

solid and the possible existence of organic matter in raw water (NTU < 3). Then, after the pretreatment process, chloric acid (HCl) and antiscalant A-NET RO (100 ppm) were mixed with raw water to adjust pH and avoid precipitation. The resulting solution was proceeded to RO, NF and ED system individually. Afterward, the concentrated water of each process system in the first step entered the second stage of every system with further recycling of water and reducing the amount of disposing concentrated water.

2.2.1. ED experiment

The ED operation was carried out on a laboratory pilot plant. This apparatus was a batch ED unit composed of ten compartments alternatively separated by cation and anion exchange membranes. The two electrode compartments are separated from the others to prevent a modification of the composition of the solution, which could be caused by electrode reactions. The circulation of water through the, concentrate and electrode rinse compartments was assured by pumps. The used membranes were a conventional cationic exchange membrane CMX and an anionic exchange membrane ACX manufactured by Tokuyama Corp. The stack design characteristics of the ED pilot plant are given in Table 2.

2.2.2. NF/RO system

NF/RO experiments are carried out on a semi-industrial pilot NF/RO provided by Firmus. The pilot is equipped with one pressure tube that can receive spiral organic

Table 2
Stack design

Membrane area (cm²)	200
Cation exchange membrane	CMX Tokuyama Corp.
Anion exchange membrane	ACX Tokuyama Corp.
Number of cell pair	10
Separator	PE + PP
Electrode	DSE
Flow of and concentrate compartments (L/h)	180–200
Flow of electrode compartment (L/h)	150
Current max (A)	9
Maximum voltage (V/Cell)	1.5
Polarity reversal	Manual

membranes 2.5 inches in diameter by 40 inches long. The experiments were conducted using nanofiltration membranes (NE2540-90) and reverse osmosis membrane (RE2540-BN). After the run, membranes were cleaned with alkaline and acidic cleaning solutions according to the manufacturer's recommendations. The main characteristics of these membranes are shown in Table 3.

2.3. Analytical methods

The experiments were performed at 29°C. Samples of permeate of various tested process were collected and the water parameters were determined analytically following standard methods previously described [25]. The followed parameters are electric conductivity, pH, sulfate, chloride, sodium, calcium, magnesium and nitrates contents.

2.4. Performances of calculation

The performances of various tested process were quantified with:

- Salt rejection (R%), which is defined as:

$$R = \frac{\left(C_0 - C_p\right)}{C_0} \times 100 \tag{1}$$

where C_0 and C_p (mg/L) are ion concentrations respectively in the initial solution and in the permeate.

- Recovery rate (Y) which is defined as:

$$Y = \frac{Q_p}{Q_0} \times 100 \tag{2}$$

Table 3
Membranes characteristics

Membrane	Manufacturer	Type	MWCO (Da)	Configuration	Area (m²)
NE2540-90	CSM	NF	200D	Spirale	2.5
RE2540-BN	CSM	RO	–	Spirale	2.5

where Q_p is the permeate flow (L/h) and Q_0 the feed flow (L/h).

- Langelier saturation index (LSI) which is defined as:

$$\text{LSI} = \text{pH} - \text{pHs} \tag{3}$$

where pH is the pH value of the solution and pHs is the saturation pH.

If LSI < 0, the water is under saturated and most of the $CaCO_3$ is dissolved in the water; while LSI > 0 indicates a supersaturated water and $CaCO_3$ precipitation. Finally, water is in equilibrium with $CaCO_3$ when LSI = 0.

- Specific power consumptions for ED which is defined as:

$$\text{SPC} = \frac{I}{V}\int U dt \tag{4}$$

where I (A) is the enforced current intensity, U (V) the potential, V (L) the weaken stream volume and t is the time. The energy needed to pump the diluate and concentrate streams through the ED stack was not considered.

- Demineralization rate for ED:

$$\text{DR}\% = \frac{E_f - E_p}{E_f} \tag{5}$$

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

- Energy consumption of RO/NF:

$$E = \frac{\left(P_{\text{RO/NF}}\right) \times 100}{36 \times Y \times \eta} \tag{6}$$

where $P_{\text{RO/NF}}$, η and Y are the applied pressure in RO or NF process (bar), the global pumping system efficiency and the overall recovery rate (%), respectively.

3. Results and discussions

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

The performances of ED, RO and NF in nitrate removal were performed individually using a feed solution containing high nitrate level (450 mg NO_3^-/L). It is also characterized by a higher level of salinity (TDS = 4,240 ppm) that exceeds greatly the standards of Moroccan regulation for drinking water and the WHO standards. ED experiments were carried out by applying a voltage of 8 V corresponding to 72 L/h of dilute flow and 75% of DR. Concerning the RO and NF membrane, the experiments were conducted underproduction mode at applied pressure of 10 and 8 bar of TMP corresponding to 34 and 42 L/h of permeate flow respectively. Results of permeated and concentrated water quality in terms of nitrate and conductivity, recovery rate for the three tested systems are depicted in Table 4.

Results of the NF system showed that the average concentration of nitrate in permeated water was greater than the standard rate (50 mg/L) and 73.8% of the nitrate removal efficiency was achieved in this system. This means that further treatment of the NF product should be considered before disinfection and distribution for human consumption. However, nitrate removal efficiency in RO system and ED was about 95.3% and 91.48%, more than that of the NF system. Results also indicates that average concentration of nitrate in permeated water in the RO system was 23 ppm, which was less than the ED system with the concentration of 40 ppm. Moreover, the obtained conductivity for the ED and NF process are typically below (1,000 mS/cm),which are within the WHO guidelines and substantially lower than the standards for drinking water quality in Morocco [7,9]. However, in the case of RO, the conductivity is relatively lower than the standards. Nitrate concentration in the brine water was 1,023; 1,859 and 1,531 μS/cm in the ED, RO and ED systems respectively. Thereby, there was no significant difference between the amounts of nitrate concentration in the concentrated water of the various tested systems. It needed to be disposed into sewers or converted into nitrogen dioxide through biological denitrification. Concerning the brine conductivities of ED, RO and NF, there were systematically higher than the Moroccan standards limit of conductivity content discharged in sewage dumping (<2,700 μS/cm). Moreover, the direct ED system can product a permeate without scaling phenomenon (LSI > 0), which present no risk to the installed equipment. While for the direct NF and RO process the scaling potential is significant under working conditions, which requires preventive action aimed at adjusting the calco-carbonic balance with remineralization step [26].

Recovery rates for the three membrane processes are almost similar; they reached 71.16%, 72.51%, 74.79% for the ED, RO and NF systems, respectively, while the energy consumption of the various system ED, RO and NF reached 1.15, 0.99 and 0.61 kWh/m^3, respectively. Knowing that the

Table 4
Performances evaluation of ED, RO and NF in nitrate removal

	NO_3^- permeate		pH permeate	LSI	NO_3^- brine	Conductivity product		Brine conductivity (μS/Cm)
	ppm	R%			ppm	(μS/Cm)	R%	
ED	40	91.48	7.64	0.22	1,023	876	79.33	8,459
RO	23	95.3	6.25	–1.57	1,859	82	97.99	9,835
NF	123	73.8	6.89	–1.86	1,531	485	88.56	9,108

public cost of electricity in Morocco is close to \$0.1/kWh [27], the cost of ED system was slightly higher than RO in the present study. It was around of 0.115, 0.099 and 0.021 (\$/m^3) for ED, RO and NF. Fig. 3 describes the comparison of energy consumption and recovery rate of the ED, NF and RO membrane processes.

3.2. Effectiveness of nitrate removal by two hybrid NF-RO and NF-ED systems

To improve permeate water quality in terms of nitrate content of NF technology, NF membrane can be combined with other membrane technologies to form different combined process systems. For this, two hybrid systems NF-RO and NF-ED were investigated in nitrate removal as shown in Fig. 4. In the hybrid NF-RO and NF-ED system, the permeate produced by NF membrane with 123 ppm of nitrate content and 8 bar in the TMP feeds RO and ED membrane in the second stage.

To highlight in greater detail the behavior of the different studied combination, the ED operations were conducted under a current intensity of 5 A corresponding to 40% of DR and dilute flow of 126.72 L/h. The RO operation was carried out at 8 bar of TMP corresponding to 168.85 L/h and recovery rate of 81.25%. Fig. 5 presents the permeate ions composition for two hybrid NF-RO and NF-ED systems. Table 5 gives the values of some parameters of the treated water.

It can be observed that the quality of water produced from the NF-RO and NF-ED were in compliance with the WHO and Moroccan guideline for the majority of the parameters. In fact, the obtained nitrate content for two hybrid systems is below the recommended value (<50 ppm), while the salinity is relatively lower than the standards for NF-RO system. The nitrate content was equal to 10.45 and 45.08 ppm for NF-RO and NF-ED corresponding to TDS value around 98, 76 and 301, 15 ppm respectively. Though, the rejection of monovalent ions was strongly increased when the ED and RO were applied in the second stage. Fig. 6 illustrates the rejection rate for all ions composition in the permeate of hybrid NF-RO and NF-ED.

According to the mentioned results, the removal efficiency of all ions in the RO was higher than in the ED system. Moreover, higher rejection of monovalent and divalent ions leads to lowering LSI values for both hybrid systems. It was equal –2.1 and –0.8 for NF-RO and NF-ED corresponding to pH value of 6.92 and 8.01. As result, the quality of the NF-RO permeate acquires serious corrosive character,

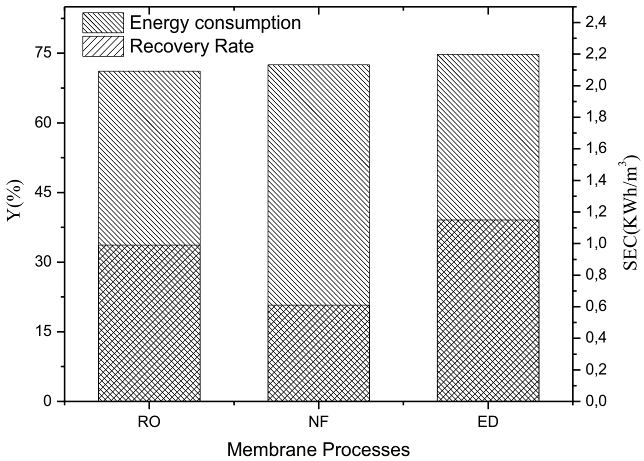

Fig. 3. Energy consumption and recovery rate for RO, NF and ED membrane systems.

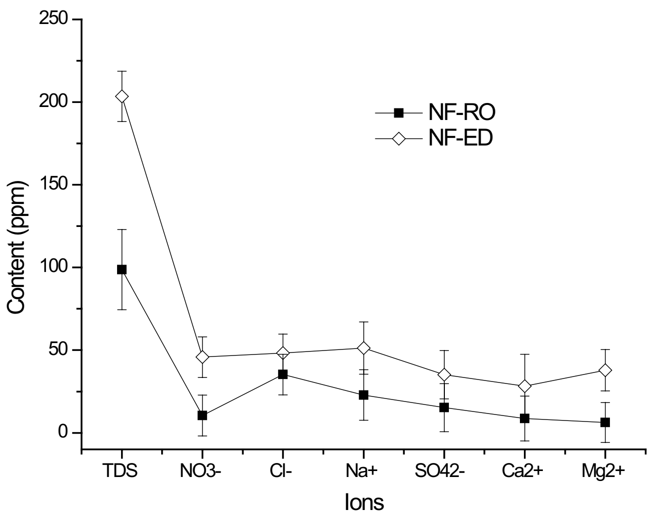

Fig. 5. Ions composition of the two hybrid NF-RO and NF-ED systems.

Table 5
Values of some parameters of the treated water

	NO_3^- (ppm)	TDS (ppm)	pH	LSI
NF-ED	45.08	203.5	8.01	–0.8
NF-RO	10.45	98.97	6.92	–2.1

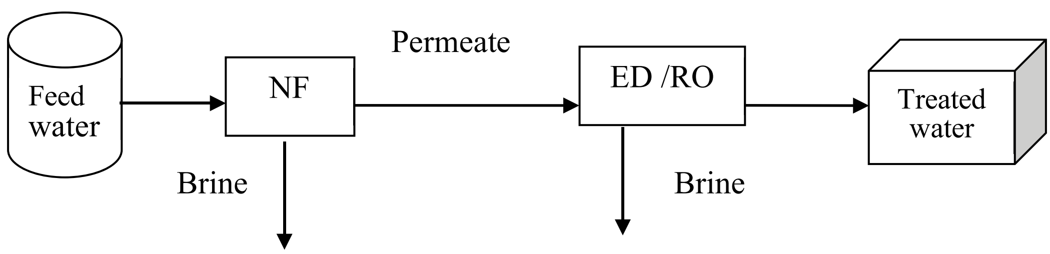

Fig. 4. Procedure of the NF-RO and NF-ED hybrid systems.

which requires a correction before distribution [26], while the quality of NF-ED permeate appears non-scaling.

The recovery rate of NF-ED reached approximatively 93.25% compared to 90.48% for NF-RO. Thus, the preliminary NF stage reduced substantially the scaling potential in the RO and ED stage, extending the life expectancy of the membrane in second stage. This also led to an increase of the water recovery of the overall hybrid system; thus, RO and ED membrane can ensure good water purity.

However, the energy consumption of both NF-ED and NF-RO hybrid systems was similar, around 1.13 and 1.24 kWh/m³ respectively, On the basis of the above analysis, it can be concluded that the energy consumption was reduced by increasing the number of stages [23]. Thereby, the cost of energy NF-ED and NF-RO was practically comparable with a particular benefit of NF-ED. Table 6 shows a comparison of water production and energy consumption of hybrid NF-ED and NF-RO systems. The calculations of the overall water recovery rate and energy consumption values for the various hybrid systems were based on the formulas below:

- Overall water recovery rate [23]:

$$Y_T = \frac{Y_{\mathrm{NF}}}{1 - Y_{\mathrm{RO/ED}}\left(1 - Y_{\mathrm{NF}}\right)} \tag{7}$$

where Y_{NF} and Y_{RO} are the recovery rate (%) in each stage, respectively.

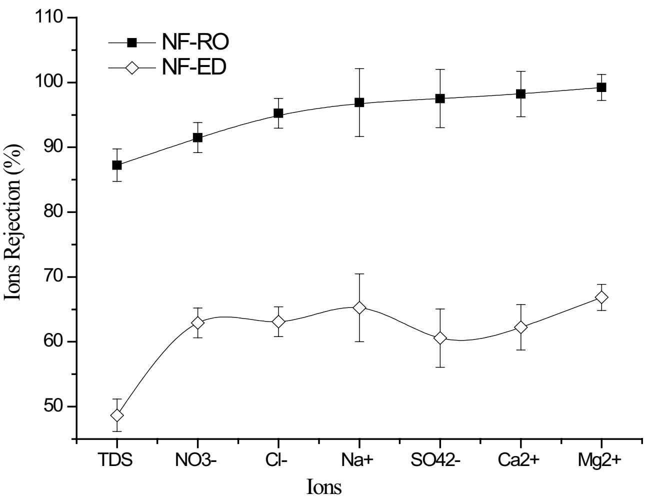

Fig. 6. Rejection rate ions composition of hybrid NF-RO and NF-ED.

- Specific energy consumption (SEC) for NF-RO [28]:

$$\mathrm{SEC} = \frac{\left(P_{\mathrm{RO}} + P_{\mathrm{NF}}\right) \times 100}{36 \times Y \times \eta} \tag{8}$$

where P_{RO}, P_{NF}, η and Y are the applied pressure in reverse osmosis and nanofiltration stage (bar), the global pumping system efficiency (80%) and the overall recovery rate (%), respectively.

- Specific energy consumption (SEC) for NF-ED was calculated by the sum of both the energy of NF and ED separately mentioned in part 2.4.

$$\mathrm{SEC} = \mathrm{SEC}(\mathrm{NF}) + \mathrm{SEC}(\mathrm{ED}) \tag{9}$$

3.3. Blended water product

As it shown in Tables 4 and 5, the water quality obtained by NF-ED hybrid system is practically equilibrated and is in accordance with the standards of corrosion [23,29]. However, the water quality obtained by direct RO and by NF-RO hybrid system is not equilibrated, unpalatable, corrosive and unhealthy. For these, the blending approach was adopted only for direct RO and hybrid NF-RO system in order to rebalance the produced permeates, to bring its characteristics in accordance with drinking water standards of nitrate level, and to minimize the brine volume.

The TDS of the permeate will be lower than 1,000 ppm, the nitrate content do not exceeded 50 ppm and the LSI value will be in range of $-0.2 < \mathrm{LSI} < 0.2$.

On the basis of these three conditions, the flow rate of the blended water was calculated by the following equation:

$$Q_{\mathrm{blend}} = \frac{Q_p\left(50 - C_p\left(\mathrm{NO}_3^-\right)\right)}{\left(C_{\mathrm{blend}}\left(\mathrm{NO}_3^-\right) - 50\right)} \tag{10}$$

where Q_p: flow of permeate; C_p: content nitrate ion in permeate; Q_{product}: product water; C_{blend}: content nitrate ion in brine

According to these conditions, Table 7 provides the impact of blending strategy on the permeate Hybrid NF-RO system and direct RO.

The results show that the blending approach with brine for each configuration had the beneficial effect of increasing nitrate content in the final product of water according to WHO and Moroccan standards (50 pm). In addition, it led to prevent corrosion, with improvement in the LSI value, and to minimize the brine volume to discharge.

Table 6
Recovery rate, pressure, specific energy consumption, and cost of energy of hybrid processes NF-RO and NF-ED

Hybrid systems	Y (%)	Pressure (bar)		Specific energy consumption (kW/m³)			Cost ($/m³)
		1st stage	2nd stage	SEC 1st stage	SEC 2nd stage	SEC total	
NF-RO	90.48	8	8	0.61	0.63	1.24	0.124
NF-ED	93.25		–	0.61	0.52	1.13	0.113

Table 7
Impact of blending strategy on the permeate direct RO and NF-RO hybrid system

Hydraulic parametrs	Direct RO		NF-RO hybrid system	
Performances	Before blending	After blending	Before blending	After blending
NO_3^-	23	50	10.45	50
LSI	–1.54	0.15	–2.02	0.10
Recovery rate (%)	72.35	90.71	90.48	95.42%
Blending rate (%)	45.63%		65.21%	
TDS (ppm)	825.78		775.46	

Table 8
Comparison of diverse direct and hybrid membrane processes for nitrate removal

	Direct process		Hybrid process	
	ED	RO-blending	NF-ED	NF-RO-blending
NO_3^- (ppm)	40	50	45.08	50
TDS (ppm)	438	825.78	301.15	775.46
Recovery rate (%)	71.16	90.71	93.25	95.42%
Energy consumption (kWh/m^3)	1.15	1.25	1.13	1.86
Energy cost (\$/m^3)	0.115	0.125	0.113	0.186

Therefore, the flow rate of the blended water is established at 65 and 19 L/h for hybrid NF-RO and direct RO respectively with portion rate of brine blending equal to 65.21% and 45.63%, respectively, which helped to increase the recovery rate from 90.48% to 95.42% for NF-RO and from 72.35% to 90.71% for RO. Thus, blending strategy in the two cases makes it possible to achieve adequate level of TDS and all the others required parameters to prevent corrosion as well as to satisfy the water quality regulations. Moreover, blending strategy enables to reduce the ultimate brine volume as well as the management cost associated with the concentrate disposal, thus impacting marine life.

3.4. Comparison of diverse direct and hybrid membrane processes for nitrate removal

Table 8 summarizes a brief comparison of direct and hybrid process in nitrate removal with and without blended strategy. From these results, all the proposed system of treatment are possible to obtain water having a nitrate content corresponding to recommended concentrations (= <50 ppm) with a various content of salinity respecting Moroccan standards (<1,000 ppm), which that are increased in this order NF-ED < ED < RO-blending < NF-RO-blending. In terms of recovery rate, the order is as follows NF-RO-blending > NF-ED > RO-blending > ED, while the order of energy consumption and cost are decreased in the following order: NF-RO-blending > RO-blending > ED > NF-ED. However, the choice between the four options depends on the desired water quality in terms of salinity content.

4. Conclusion

The goal of this study is to compare separately the direct (ED, NF, RO) and the (NF-RO/ED) hybrid membrane processes for the treatment of the complex ground water containing both high nitrate and salinity.

Results of direct process (NF) indicate that the quality of the produced water is poor in terms of nitrate content, it is exceeded largely the Moroccan standards (>50 ppm), while the quality of direct RO and ED are better. In terms of salinity, the direct ED and NF process are typically below 500 ppm. However, in the case of RO, the salinity is relatively lower than the standards. Thereby, the energy and cost efficiency of ED is slightly higher than RO.

In order to face of direct NF disadvantage, a hybrid processes coupling NF and RO and ED stage were proposed. Considering the nitrate removal efficiency of NF-RO and NF-ED hybrid system, the results show that the NF-ED quality of nitrate content is better than that of NF-RO. Thus, the cost of energy NF-ED and NF-RO were practically comparable with a particular benefit of NF-ED. In addition, blending the permeate RO and NF-RO with their brine allowed to obtain product water at a maximum admissible of nitrate level, TDS and LSI. The portion of blending rate with NF-RO permeate is equal to 65% and to 45% of RO.

Furthermore, to achieve our goal, it appears that the NF-RO hybrid treatment is technically more attractive than others to obtain water with 50 ppm of nitrate and a target recovery rate of 95.42%, despite their moderately higher energy cost. At the same time, NF-RO hybrid minimizes the fraction of brine disposal dumping to sewage, making it environmentally friendly.

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