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Nanofiltration and reverse osmosis membrane for nitrate removal: performance study and economic evaluation

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Abstract

Contamination of water with nitrates is a global problem that poses a serious threat to human health. In Morocco, a country characterized by limited water resources, groundwater accounts for almost 32% of total resources. However, 6% of these water resources are estimated to have nitrate levels greater than 50 mg / L, the level recommended by the world health organization for drinking water (WHO) and in some areas of high agricultural activity the nitrate content exceeds 250 mg / L. In the first part of this study, the efficiency of two commercial membranes, Reverse Osmosis and Nanofiltration membranes (Dow-FilmTec) is experimentally compared and assessed for nitrate removal by using real brackish water, containing 850-1100 mg/l of total dissolved solids (TDS) and 119-130 mg/l of nitrate. To accomplish this goal, the influence of operating conditions (pressure, recovery rate) on nitrate removal is discussed. The salt rejection from each membranes and the energy consumption are also calculated to choose the best membrane for nitrate removal and drinking water production. The pilot used in this study (supplied by the French company TIA) is an industrial pilot plant having two modules equipped with various spiral commercial membranes with an area of 7.6 m². The second part of this study estimates the total cost of the produced water by the application of the two studied membranes (NF and RO) for a plant of nitrate removal having a capacity of production of 2400 m^3/d (100 m^3/h) corresponding to a water consumption for 50000 capita following the Moroccan standards in rural medium. The design of the plant was carried out based on the experimental results from the first part of the study corresponding to a recovery rate of 83% and 10 bars of pressure. Technically and economically, the work shows that in this case, the NF process appears more suitable than the RO for nitrate removal

Keywords: *Nanofiltration, reverse osmosis, nitrate, ground water, economic evaluation*

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1. Introduction

The problem of ground water pollution by nitrate ions has been reported above the drinking water standards in different region in Morocco, especially in the irrigated areas that are experiencing intense agricultural activities [1–9]. This trend has raised concern because nitrate causes a deterioration of water quality and generates a risk in rural areas, where most people use the aquifer as their daily drinking water [10]. The ingested nitrate turns to nitrite, leading to child methaemoglobinaemia and old age cancer forms. This is why the World Health Organization (WHO) fixed at 50 mg/l the maximum acceptable concentration of nitrate in drinking water [11], the same norm is adopted in Morocco. Several treatment processes including adsorption [12–14], ion exchange, biological denitrification, chemical denitrification, reverse osmosis, nanofiltration, electrodialysis, and catalytic denitrification can remove nitrates from water with varying degrees of efficiency, cost, and ease of operation, also chemical methods are some of the conventional techniques implemented [15]. 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 [16]. Ion exchange techniques are sensitive to various contaminants present in the water [17] and require post-treatment. Reverse osmosis and nanofiltration membrane are also sensitive to contaminants other than nitrate. It requires a specific amount of pressure and is also susceptible to biofouling. Biological and chemical methods cause toxicity in the water. Membrane processes, such as nanofiltration (NF) and reverse osmosis (RO), can be considered promising technologies to treat groundwaters with such nitrate contents in terms of high efficiency, easy operation, high effluent water quality, modularity, and flexibility [18,19]. These methods consist in forcing the water to pass through a membrane under pressure, the ionic species, such as nitrate, being rejected in the waste stream. Up to now, various research studies have been performed to remove nitrate from drinking water by NF and RO membrane modules [20,21]. Data from a plant using full-scale reverse osmosis to remove nitrate indicate that a concentration of 61 mg NO_3^-/L can be reduced to 2.6 mg NO_3^-/L using composite membranes consisting of a thin film of spiral polyamide. The plant has a capacity of 630 m3/ h of permeate, which is supplemented with 209 m3 / h of mixed water, the final target nitrate concentration being less than 35 mg NO_3^-/L. A 2-phase unit is used to provide 80% recovery with a feed pressure of 11.72 bar [22]. Schoeman et al. (2003) cite data for a small plant where reverse osmosis is used; this plant is capable of removing high concentrations of nitrate in the water at the source. With a capacity of 55 m^3 / day and a recovery rate of 50% at a pressure of 13,75 bar. 96% to 98% of the nitrate in this plant was removed; with permeate concentrations below 22 mg NO_3^-/L, while concentrations at the feed water were between 186 and 235 mg NO3-/ L [23]. Concerning nanofiltration technology, laboratory tests and pilot tests were also conducted to evaluate its effectiveness for nitrate removal. It has been established that, in general, membranes with small pores must remove more than 75% of nitrate ions [24]. In the same study, the effectiveness of four nanofiltration membranes for pesticide and nitrate removal was evaluated. It was found that a small pore membrane was able to remove 76% of the nitrate, reducing a concentration of 45 mg NO_3^-/L in the feed water up to 11 mg NO_3^-/L in permeate [24]. Moreover, El-Ghzizel et al. (2018) published results by using a 12 m3/day Nanofiltration (NF90) pilot plant capable of reducing the nitrate concentration by 68 mg NO_3^-/L to 18 mg NO_3^-/L, with a recovery rate of 75 %. To obtain these results, a feed pressure of 5 bar was applied, and the energy consumption was 0.2 kWh/m3 [5]. In this context, the aim of this study is twofold; firstly, the objective is meant to compare two membranes, nanofiltration (NF) and reverse osmosis (RO), for nitrate removal by using real brackish water, containing 850-1100 mg/l of total dissolved solids (TDS) and 119-130 mg/l of nitrate. Secondly, it's focused mainly on the estimation of the total cost of the produced water by using the two studied membranes (NF and OI) for a plant of nitrate removal. The production capacity of this plant is 2400 m^3/d (100 m^3/h) corresponding to a water consumption for 50000 capita following the Moroccan standards in rural medium. The design of this plant is carried out for the predetermined optimized conditions corresponding to the results obtained from the first part of the study.

2. Materials and methods

The experiments were performed on an NF/RO pilot plant (E 3039) supplied by TIA Company (Technologies Industrielles

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Appliquées, France). Fig. 1 gives the scheme of the pilot plant used. The pressure applied over the membrane is varied from 5 to 24 bar with manual valves. The pilot plant is equipped with two identical spiral wound modules operating in series.

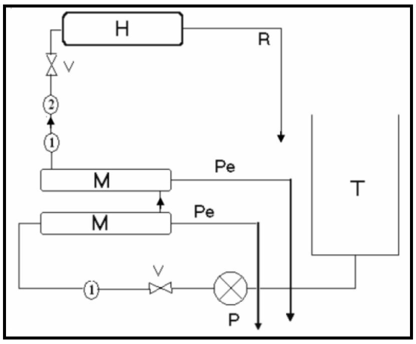

Fig. 1. Diagram of the nanofiltration pilot plant. T: tank;

P: feed pump; V: pressure regulation valves; M: nanofiltration module; Pe: permeate recirculation; R: retentate recirculation; H: heat exchanger; 1: pressure sensor; 2: temperature sensor.

Each module contains one element. The pressure loss is about 2 bar corresponding to 1 bar of each module. The configurations tested are: simple pass, and supplied batch configuration. Table 1 gives the characteristics of the commercial membranes used.

Table 1 Characteristics of the used membranes

Membrane	**Area (m^2)**	**P_{max}(bar)**	**pH**	**Max temp (°C)**	**Materials**	**Cl_2 tolerance ppm**
RO	7.5	41	2-11	45	Polyamid	0.1
NF	7.6	41	3-10	45	Polyamid	0.1

The Experiments of this study are performed at 29°C. Samples of permeate are collected and the water parameters are determined analytically following standard methods.

In addition, the performances of the plant were followed in terms of the salt rejection, permeate flux, recovery rate and the specific energy consumption parameters which are defined as:

$$R(\%) = \frac{(Gf-Gp)}{Gf} \times 100 \quad (1)$$

where Gp and Gf are respectively the conductivity of permeate and feed water;

- Permeate flux (L/h. m^2) :

$$Flux = \frac{Qp}{S} \quad (2)$$

where S (m2) and Qp (L/h) are respectively the surface area of the membrane and permeate flow;

- Recovery rate (%):

$$Y = \frac{Qp}{Qf} \times 100 \quad (3)$$

where Qf and Qp are the feed and the permeate flow rate respectively;

- Specific energy consumption (KWh/m^3):

$$E = \frac{\Delta P \times 100}{(\eta \times Y \times 36)} \quad (4)$$

where ΔP, η and Y are the transmembrane pressure (bar), η is the global pumping system efficiency (0.85) and the recovery rate (%), respectively.

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3. Results and discussion

3.1. Pure water flux

Figure 2 shows permeate flux versus applied pressure for both membranes. The slopes of the straight lines give the water permeability values for each membrane. The slope is a measurement of the resistance exerted by the membranes as a diffusion medium, when a given force (pressure) is acting on a component [25]. The increase of the permeate flux with applied pressure indicates that NF90 presents the largest pore size with a permeability of 6.7 L/h.m².bar while reverse osmosis membrane which is a dense membrane shows a lower permeability of 3.15 L/h.m².bar.

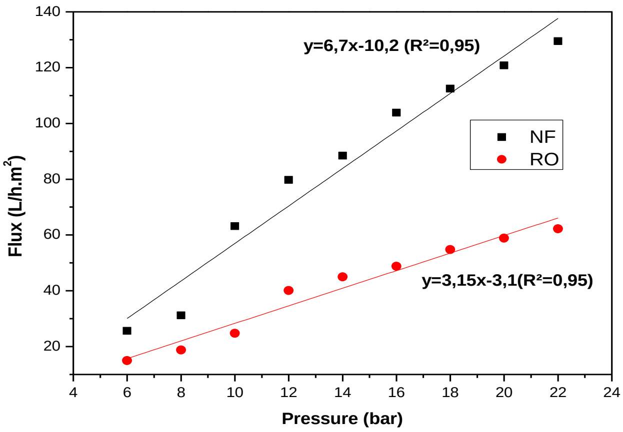

Fig.2 water permeability of nanofiltration and reverse osmosis membrane

3.2. Effect of pressure on flux, nitrate removal and energy consumption

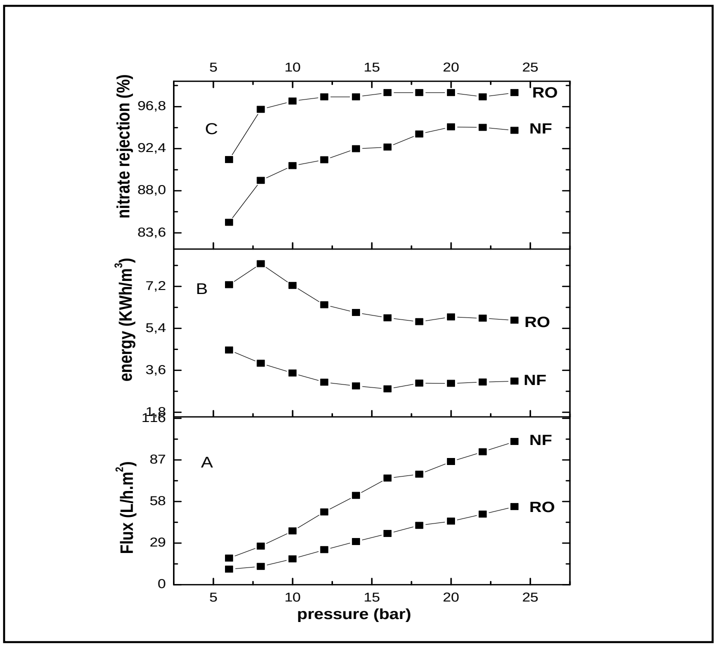

Fig. 3 Variation of the flux, energy consumption and nitrate removal versus applied pressure for the two tested membranes (NF90 and RO)

3.3. Effect of recovery rate on the nitrate removal

The effect of recovery rate on nitrate removal at 119 mg/l of initial nitrate concentration for NF and RO membranes is shown in Fig. 4. Applied pressure and temperature are fixed at 10 bar and 29°C, respectively. The quality of the

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permeate in terms of nitrate concentration respects the WHO standard (50mg/L) for the two membranes. For this reason, it is preferable to work with the highest recovery rate. Highest recovery rate allows a large productivity, a good quality in terms of nitrate content, lower energy consumption and in the end a small quantity of rejection stream. At this recovery rate (83%), NF and RO membranes gave a nitrate removal percentage of 98.23 and 80 % respectively.

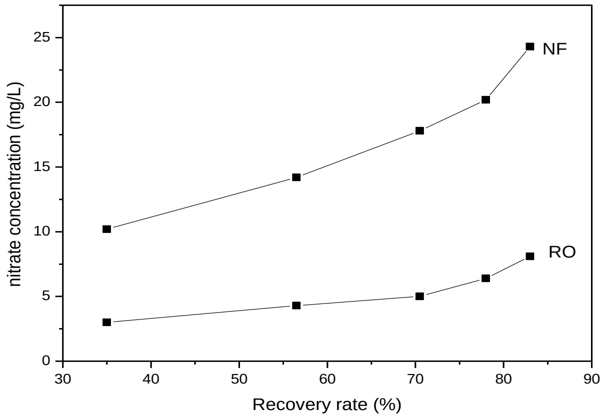

Fig.4 Effect of recovery rate on nitrate concentration in the permeate of the two tested membrane (NF, RO)

3.4. Effect of recovery rate on conductivity

Figure 5 gives the permeate conductivity at 10 bar at different recovery rates. The obtained permeate quality with NF90 and RO membranes are not satisfactory. However, the produced water by RO and NF membrane is practically demineralized, a reminéralisation step is needed if water is destined for consumption. According to these results, it is reasonable to conclude that NF and RO membranes are almost equally for nitrate removal and salinity in the range of the studied operating conditions (pressure and recovery rate).

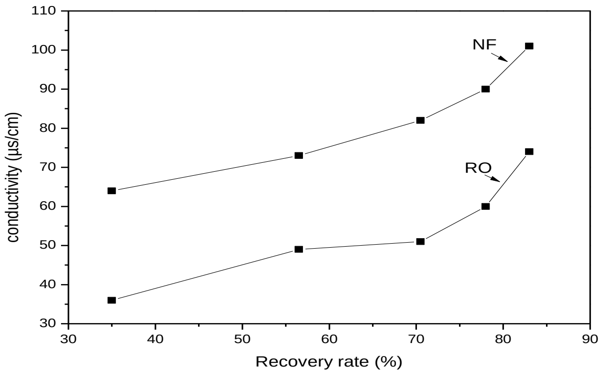

Fig.5 Permeate conductivity vs recovery rate for NF and RO membranes

3.5. Proposed design

The proposed design for the NF and the RO unit is shown in Fig. 6. The main treatment unit comprises:

- Pre-treatment post
- NF and RO group
- Post-treatment (remineralization with raw water)

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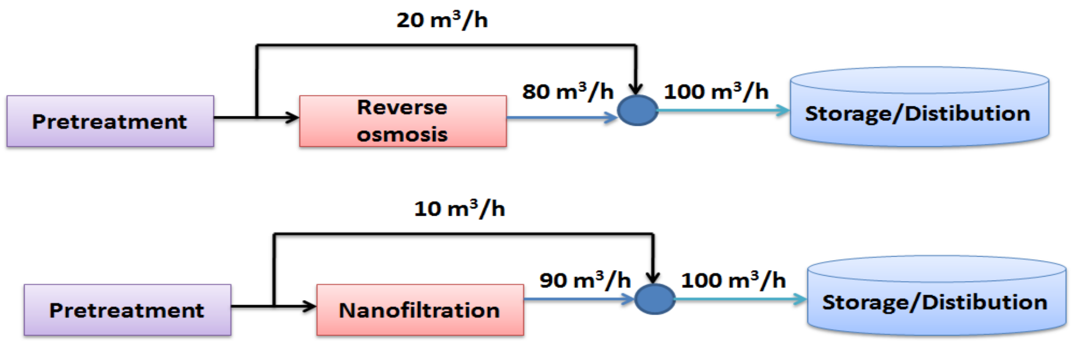

Fig.6 Proposed design for the NF and RO unit

3.5.1 Pre-treatment post

A simple pre-treatment system is carried out; it is composed of sand filters of 5 µm, with capacity of 125 m^3/h filled with sand, to retain the suspended solids that might be present larger than 10 µm in order to provide a final protection and to conform the pre-treated water to the NF and RO membranes requirements.

3.5.2. NF and RO group

The calculation of the NF/RO group was carried out on the basis of the experimental results obtained. Table 2 gives the results of the design of the NF/RO group. The number of modules and the number of pressure tubes are calculated by the following relation [26].

A. Number of modules

$$N = \frac{Fp}{S*Jw} \quad (5)$$

Fp: flow of permeate (L/h), S: membrane area per module (m^2), Jw: permeate flux ($L/h.m^2$) and N: number of modules.

B. Number of pressure tubes

$$N(P) = \frac{N}{Ne} \quad (6)$$

N: number of modules, Ne: Number of modules per tube and N (P): number of pressure tubes.

Table 2: Design of the NF/ RO group

Configuration	**Simple pass**		**Flow rate of the mixed water**	**Production capacity**
Production capacity	RO	80 m^3/h	20 m^3/h	100 m^3/h
	NF	90 m^3/h	10 m^3/h	100 m^3/h
Membranes per tube of pressure: Ne	RO	6		
	NF	6		
Number of modules	RO	405		
	NF	473		
Number of pressure tube	RO	67		
	NF	78		
Recovery rate (%)	RO	83%		
	NF	83%		

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3.5.3. Post-treatment

The post-treatment includes remineralization by the raw water. This method is chosen because it appears the least-cost remineralization technique. It is accomplished by routing a portion of the raw groundwater around the NF-RO process and blending it with permeate. After reminéralization step, the produced water is stored in the product water tanks before distribution.

3.6. Performances comparison

In this part, the performances of the two technologies in desalination of brackish water are described and compared briefly. The experiments are carried out on two commercial membranes (RO and NF) in simple pass at pressure of 10 bars and 83% of recovery rate. The characteristics of the produced water after treatment are presented in Table 3 with and without remineralization step for the two studied membranes.

Table 3: Characteristics of the produced water after treatment with RO/NF

	Raw water	**NF**	**Mixed water (NF)**	**RO**	**Mixed water (OI)**
Conductivity (µs/cm)	1330	101	218,24	74	325,2
NO_3^-(mg/l)	119	24,3	33,33	8,1	30,28
Cl^-(mg/l)	536	61,1	106,40	51	148
F^-(mg/l)	1,2	0,04	0,15	0,03	0,264
TH (F°)	30,66	1,58	4,35	1,53	7,356
TAC(F°)	30	3,7	6,20	2,7	8,16

The analysis of the results given in Table 3shows that: The obtained permeate quality with the NF90 and OI membrane is not satisfactory. However, the produced water by RO and NF membrane is practically demineralized, a reminéralisation step is needed. After remineralization step with raw water, the produced water quality is satisfactory in term of conductivity and nitrate content.

3.7. Total desalinated water cost

Cost is a major factor in implementing membrane technologies. Several factors affect the cost for NF and RO process such as the raw water quality, the plant capacity, the recovery rate and the desired produced water quality. The economical evaluation of the cost of the produced cubic meter requires the calculation of the capital cost, the operating cost, membrane replacement costs and the energy cost. The capital cost and operating cost of nanofiltration and reverse osmosis membranes are assessed and compared. The economical analysis is carried out for production capacity of 2100 m^3/d corresponding to water consumption for 42000 capita following the Moroccan standards (consumption for domestic use of 50 L/per capita per day.

3.7.1. Capital cost

The capital cost includes the cost of system itself and its foundation on its place. In our case, the total capital cost includes:

- Cost of construction and building.
- Cost of pre-treatment step.
- Cost of NF and RO group.

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- 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 capital costs 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 years. Total capital costs 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[27]. Table 4 summarizes a comparison between the NF/RO membranes of the estimated capital cost.

Table 4: Capital cost for the two membranes processes RO/NF ($)

	NF90	RO
Membranes	236500	202500
pretreatment	88000	88000
Pressure tube	78000	67000
Construction and building	34600	34600
Auxiliary equipment	126000	185640
Various service	57000	83980
Total	620100	661720

With the analysis of the results and taking into account the limitations in the calculations that have been indicated, the capital cost of the NF90 is slightly than the RO membrane, this differences of the costs are due to the flow rate of the produced water delivered by NF90 compared to RO membrane.

3.7.2. Annual operating costs

Annual operating cost covers all expenditure incurred after plant commissioning and during actual operation. These include:

- Amortization or fixed charges

When the required funds for 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 [28]:

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

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

- 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 [29]. The annual O&M costs are estimated at 20% of the plant annual payment.

- Membrane replacement

For RO and NF processes, membrane replacement rate depends largely on raw water quality. Replacement rate may vary between 5% and 20% per year. The lower bound applies to low salinity brackish water and the upper would reflect the high salinity seawater [28]. Membrane replacement cost is estimated for a 1-year period and divided by the

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quantity of water to be produced during the year to determine the overall water cost [30]. In our case, the replacement rate has been found to be equal to 5% [31–33]. The membranes life was evaluated to 5 years for the two processes.

3.7.3. Energy cost

To calculate the energy cost, such factors as the working pressure of the high pressure pump, power consumption of the metering pumps which is proportional to the pressure and inversely proportional to the conversion rate, and energy prices must be taken into account. In Morocco, the average price of energy is 0.085 €/kWh [26]. In our case, the energy consumption for the two technologies is in the range of 0.4KWh/m3 which depends on feed pressure (10 bar) and recovery rate (83%).

3.8. Cost comparison

Nanofiltration and reverse osmosis process cost data includes the following (table 5):

Table 5: Nanofiltration and reverse osmosis process cost data

	Nanofiltration	Reverse osmosis
Capital cost (CC) ($)	620100	661720
Membrane module cost (MC) ($)	236500	202500
Capacity (*M*) m^3/d	2400	2400
Plant availability (*f*) (%) [23]	90	90

3.8.1 Nanofiltration process

The calculations proceed as follows:

- Amortization factor(a)

$$a = \frac{i(1+1)^n}{(1+i)^n - 1} = \frac{0.067\,(1+0.067)^{20}}{(1+0.067)^{20} - 1} = 0.092203 \text{ year}^{-1}$$

- Annual fixed charges (A_{fixed})

A_{fixed}= a* CC= (0.092203)* (620100) =57175, 0803$/year

- Annual membrane replacement costs ($A_{replacement}$) :

$A_{replacement}$= (0.05)*(MC) = (0.05)*(236500) = 11825 $/year

- Operating and maintenance annual costs ($A_{O\&M}$):

AO&M = (0.2)*(Afixed) = (0.2)*(57175.0803) = 11435,01606$/year

- Annual energy cost (EC)

EC= E (KWh/m3)*0.085*M*365*f =0.4*0.085*2400*365*0.9= 26805.6 $/year

- Total annual cost A_{total}:

A_{total}= A_{fixed}+$A_{replacement}$+ $A_{O\&M}$+ EC= 107240.69636$/year

- Unit product cost A(unit)

$$\mathbf{A}(\text{unit}) = \frac{A(\text{total})}{f * M * 365} = \frac{107240.69636}{0.9 * M * 365} = 0.13\$/m3$$

3.8.2 Reverse osmosis process

The calculations proceed as follows:

- Amortization factor (a)

$$a = \frac{i(1+1)^n}{(1+i)^n - 1} = \frac{0.067\,(1+0.067)^{20}}{(1+0.067)^{20} - 1} = 0.092203 \text{ year}^{-1}$$

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- Annual fixed charges (A_{fixed})

Afixed= a* CC= (0.092203)* (661720) = 61012.56916 $/year

- Annual membrane replacement costs ($A_{replacement}$) :

$A_{replacement}$= (0.05)*(MC) = (0.05)*(202500) = 10125$/year

- Operating and maintenance annual costs ($A_{O\&M}$):

AO&M = (0.2)*(Afixed) = (0.2)*(61012,56916) = 12202.513832 $/year

- Annual energy cost (EC)

EC= E(KWh/m3)*0.085*M*365*f =0.4*0.085*2400*365*0.9= 26805.6 $/year

- Total annual cost A_{total}:

A_{total}= A_{fixed}+$A_{replacement+}$ $A_{O\&M}$+ EC=110145.682843 $/year

- Unit product cost A(unit)

$$A(unit) = \frac{A(total)}{f * M * 365} = \frac{110145.682843}{0.9 * M * 365} = 0.14\$/m3$$

4. Conclusion

In conclusion, the ultimate choice of RO or NF membranes for nitrate treatment is directly depending on operating conditions, nitrate rejection and economical cost. However, the three factors should be combined to arrive at the best decision. The economic evaluation of the two technologies is carried out for a production capacity of 2400 m^3/d corresponding to water consumption for 50000 capita. The expected potable water production cost is calculated to be 0.13$/$m^3$ and 0.14$/$m^3$ for nanofiltration system and reverse osmosis system, respectively. According to these results NF is more suitable for the desalination of this water due to the low energy consumption, the less membrane used and finally it's relatively less expensive compared to reverse osmosis membrane.

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