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Nitrate removal from Moroccan brackish water by nanofiltration: configuration modes and energetic assessment

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ABSTRACT

This study deals with the comparison of the performances of nanofiltration (NF) membrane (NF90-40-40) in nitrate removal from brackish water using three configurations: continuous mode, batch recirculation mode and tube pressure. The experiment is conducted on an industrial pilot plant, having two modules equipped with a spiral commercial membrane with an area of 7.6 m^2. First, process configurations are introduced, by highlighting particularly the fundamental difference between different configurations in terms of recovery rate, scaling risk and permeate quality to achieve a nitrate concentration required by the standards (<50 mg/L). Then, the potential of energy efficiency of the three configuration modes is systematically compared using analytical expressions under real conditions. Lastly, other factors such as energetic costs, operational experience, and process robustness that may particularize the different process configurations are discussed. Finally, through this analysis, it appears that practical batch configuration and pressure vessels mode offer comparable and significant energy savings, it around 0.66 and 0.58 kWh/m^3 respectively. Nevertheless, in terms of recovery rate and nitrate content, the pressure vessel is more technologically attractive with a target recovery rate of 82% in comparison to batch configuration 52%. At the same time, batch mode and pressure tube configuration minimize the fraction of brine disposal dumping to sewage, making it environmentally friendly.

Keywords: Nitrate; Nanofiltration; Recovery rate; Configuration mode; Energy assessment

1. Introduction

The nitrate concentration level in groundwater for human consumption is one of the most important problems related to the quality of the groundwater in many countries and also in Morocco [1,2]. The pollution of the groundwater by nitrates affects nearly all the Moroccan territory with approximately 6% of resources having nitrate content more than the national standards [3]. The World Health Organization (WHO) has set maximal admissible nitrate concentration on drinking water at 50 mg/L [4]. Likewise, the Moroccan legislation established a maximum allowed concentration of nitrates at 50 mg/L in drinking water.

Nanofiltration (NF) membrane process has been shown to be a feasible solution for different water treatments including brackish water (BW), surface water and

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wastewater reclamation [5,6]. The key to using NF in these applications depends on the selection of the proper NF membrane and the design of a suitable process. The design approach is generally conditioned by the type of feed water to be processed and the configuration modes. When large feed streams are intended to be treated, the continuous configuration is usually chosen. In this case, several membrane elements are connected in series inside pressure vessels organized in stages with a different number of pressure vessels in parallel. The number of concentration stages will depend on the recovery rate and the number of membrane elements per pressure vessel [7].

In some industrial applications, the volume of feed water is relatively low and the production of water is discontinuous. Technically and economically, the preferred operating mode, in this case, is the batch mode [8,9]. The principle of this configuration is based on the fact that the feed water is collected in a tank and then treated with total recycling of the concentrate obtained. Hence, the operating conditions of each membrane element in this case change throughout the treatment and the concentration of the retained solute by membrane increases over time as the permeate is removed [10,11]. Another application of the batch mode is usually done at the lab-scale and pilot-scale levels to obtain the desired volume concentration ratio. However, since all of the concentrate is recycled, the components retained by the membrane have a high average residence time in the system [12]. Another option offered by the pressure increasing staging which is applicable for either reverse osmosis (RO) and NF plant, is becoming more attractive. In this configuration, the brine of the first stage becomes the feed of the second stage. Conceived to overcome the above drawback of single-stage, increasing staging has shown theoretically that this is a promising way to improve energy efficiency [13,14]. Consequently, pressure tube configuration can mitigate the unnecessary energy dissipation and reduce the overall energy consumption despite the significant increase in capital cost induced. Based on the previously discussed configurations, various studies have been carried out to propose a method for improving energy efficiency [15,16]. NF treatment system can be adopted in various configuration modes to improve its performance and to save energy.

The objective of this study is to compare the performances of NF membrane (NF90-40-40) in nitrate removal from BW using three configurations: continuous mode, batch recirculation mode and pressure vessels. These performances will be discussed in terms of recovery rate, scaling risk and permeate quality to achieve a nitrate concentration required by the standards (<50 mg/L). Then, the potential of energy efficiency of all the three configuration modes is systematically compared using analytical expressions under real conditions. Lastly, other factors are discussed such as energetic cost, operational experience, and process robustness in order to evaluate the performances of different tested configurations.

2. Experimental

2.1. Characteristics of the feed water

The experimental is applied in a case study to estimate the performance of a NF plant treating BW containing nitrate which exceeds slightly the concentration limit for drinking water (50 mg/L). The ionic composition of the feed BW is shown in Table 1.

The NF90-4040 spiral-wound membrane (Dow-FilmTec) is selected for the study as the feed water slightly exceeds the recommended nitrate concentration. The experimental setup is an industrial pilot NF/RO(E3039) provided by the French company TIA (Applied Industrial Technologies). The pilot is equipped with two identical modules in series described in a previous study [18,19].

Table 2 gives the characteristics of the used membranes. The pilot-scale experiments are conducted in three different operational configuration modes: simple pass, batch mode and pressure vessels. Fig. 1 gives a design of different configuration modes.

2.2. Analytical methods

The experiments are performed at 29°C. Samples of permeate are collected and the water parameters are determined analytically following standard methods previously described [20]. The followed parameters are:

Table 1
Characteristics of the feed BW

Parameters	Feed BW	Moroccan guidelines [17]
Temperature (°C)	30	35
Turbidity (NTU)	<3	<5
Electric conductivity (μS/cm)	2,010	<1,000
TDS (ppm)	1,280	
pH	7.80	6.5–8.5
pHs	7.41	–
TH (°F)	44.2	50
Alkalinity (ppm)	100	–
Ca^{2+} (ppm)	108	270
NO_3^- (ppm)	90	50
Na^+ (ppm)	283	200
Mg^{2+} (ppm)	41.96	<50
Cl^- (ppm)	560	250
SO_4^{2-} (ppm)	126	200
LSI	–0.53	–0.2 < LSI < 0.2

Table 2
Characteristics of the used membranes

Membrane	NF90-4040 (FilmTec)
Surface (m^2)	7.6
P_{max} (bar)	41
pH	3–10
Max. temp. (°C)	45
MWCO (Da)	100–200
Material	Polyamide
[Cl_2] tolerance (ppm)	0.1

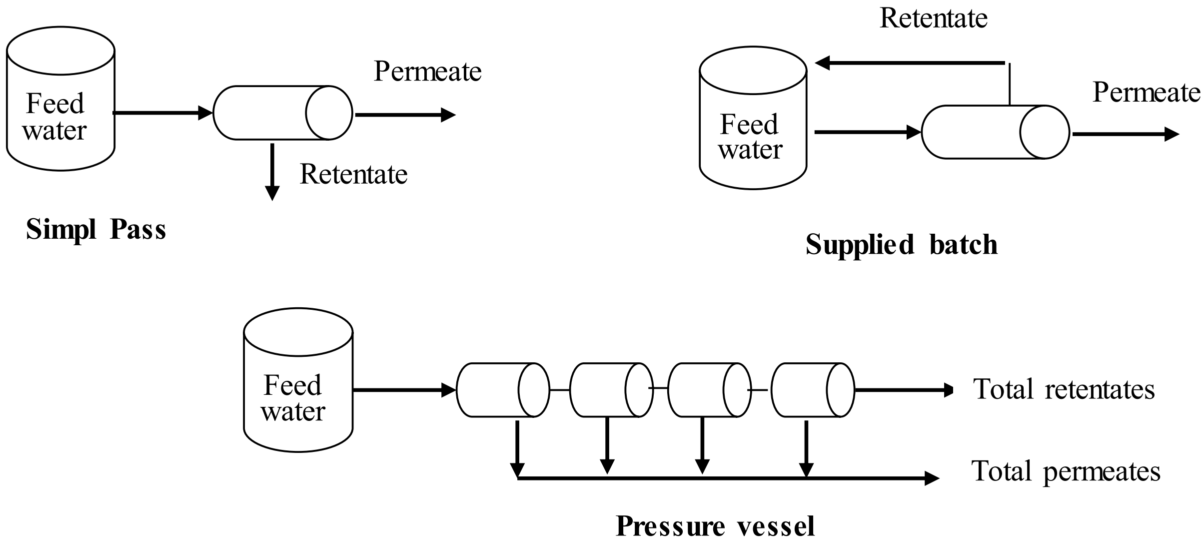

Fig. 1. Design of different configurations modes.

- Electric conductivity, pH, sulfates, calcium and nitrates contents.
- Salt rejection (R%) which is defined as:

$$R = \frac{C_0 - C_p}{C_0} \times 100 \quad (1)$$

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

- Recovery rate (Y) which is defined as:

$$Y = \frac{Q_p}{Q_0} \times 100 \quad (2)$$

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

- Langelier Saturation Index (LSI): is the corrosion index, which has been widely used in the water industry to control the corrosion of pipes. LSI is simply defined as follows [21–23]:

$$\text{LSI} = \text{pH} - \text{pH}_S \quad (3)$$

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 energy consumption (SEC) (kWh/m^3): is proportional to transmembrane pressure (TMP). SEC is calculated by the following relation [24]:

$$E\left(\frac{\text{kWh}}{\text{m}^3}\right) = \frac{\Delta P}{Y \times \eta \times 36} \times 100 \quad (4)$$

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

3. Results and discussions

3.1. Simple pass configuration

The applied TMP is one of the most significant factors affecting the NF process. In this study, the experiments are carried out at four TMP, namely 8, 10, 15 and 20 bar. The obtained results (Fig. 2) show that the nitrate removal efficiency and the permeability of the membrane are improved with increasing applied TMP. Thus, for a linear fit of the permeate flux, the R^2 value is equal to 0.992, which indicates that concentration polarization is negligible. This improvement was expected since the increase of driving force which corresponds to the pressure gradient, promotes solvent transfer, while the transfer of the solute by diffusion remains constant, as reported in previous studies [25,26]. In addition, for all applied TMP the nitrate and sodium contents in the permeate are below the Moroccan recommended standards. The same behavior is observed for chloride content, except at 8 bar. Thereafter, the operations are carried out using a TMP of 10 bar to evaluate the performances of the proposed configurations.

3.2. Supplied batch configuration

In this part, the supplied batch configuration is conducted with a TMP of 20 bar, corresponding to 1,468 L/h of feed flow. The permeate is recuperated while the retentate is recirculated to the tank of alimentation. Water supply equal to the flow of the permeate is maintained continuously. The volume of the tank of alimentation is fixed at 50 L. The recovery rate is calculated on the basis of Eq. (2).

Fig. 3 gives the evolution of flow rate of permeate, retentate and recovery rate as a function of time. In this figure, the observed decline in flux is a consequence of both decreases in the effective pressure caused by the

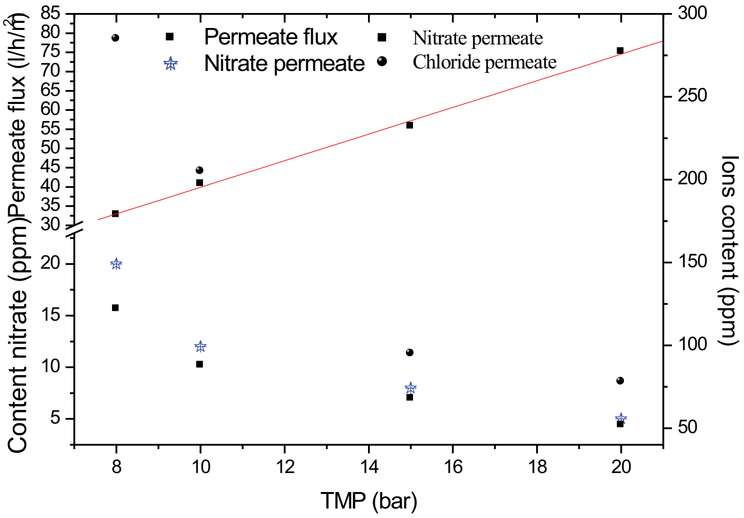

Fig. 2. Effect of TMP on permeate flux, nitrate, chloride and sodium content in the permeate.

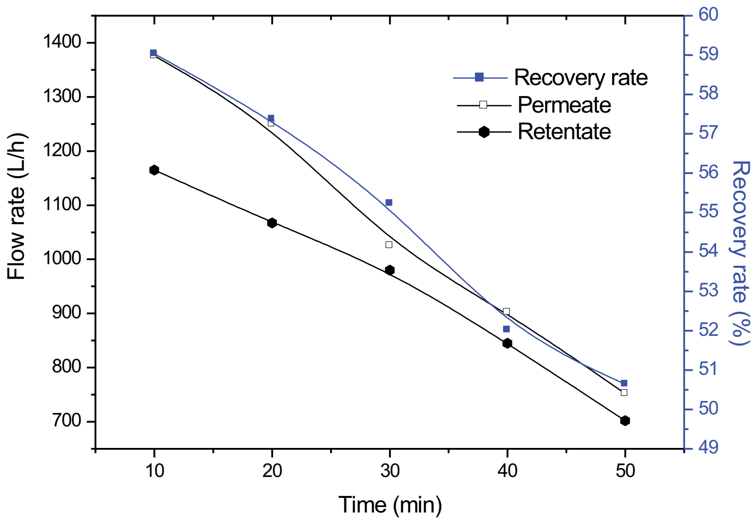

Fig. 3. Evolution of flow rate (permeate, retentate) and recovery rate as a function of time, TMP = 10 bar.

pressure drop and an increase in the osmotic pressure due to the concentration rise. In this case, the osmotic pressure increases from 0.042 bar for the initial feed solution to 0.18 bar for the final brine at the outlet of the eighth membrane module. Moreover, the effect of concentration polarization is still low. As shown in Fig. 3, the recovery rate of NF90 membrane drops from 59.2% to 50.9% for each moment. While the overall recovery rate of the system increases to reach 52% after 40 min of the treatment.

Fig. 4 shows that all the followed parameters increase practically over time in the feed, retentate and permeate, but they are still below the standards required by WHO until 40 min: nitrate (35 ppm), chloride (185 ppm) and sodium (98 ppm).

Thus, after 40 min of treatment, the water quality of the permeate becomes worse when electric conductivity of the permeate reaches 1,180 µS/cm, and the growth trend is accelerated while the content of nitrate, sodium and chloride in permeate exceeds the standards at 50 min of treatment, while the electrical conductivity increases and reaches 2,230 µS/cm in the feed water and exceeds 4700 µS/cm in the retentate.

Fig. 5 shows the evolution of the concentration ratio Ca^{2+}/Mg^{2+}, LSI and pH in the NF retentate as a function of time, it can be seen that all parameters increase with time and after 20 min a decrease was observed for these parameters indicating the onset of precipitation phenomena. After 50 min, the precipitation is observed with the naked eye in the retentate.

3.3. Pressure vessel configuration mode

In order to obtain more quantity of freshwater and to minimize the brine volume, the experiments are carried out on pressure tube configuration with nitrate concentration equal to 90 ppm at a pressure of 10 bar. In this configuration mode, the retentate of the first module serves as the feed of the second module, and all modules were arranged

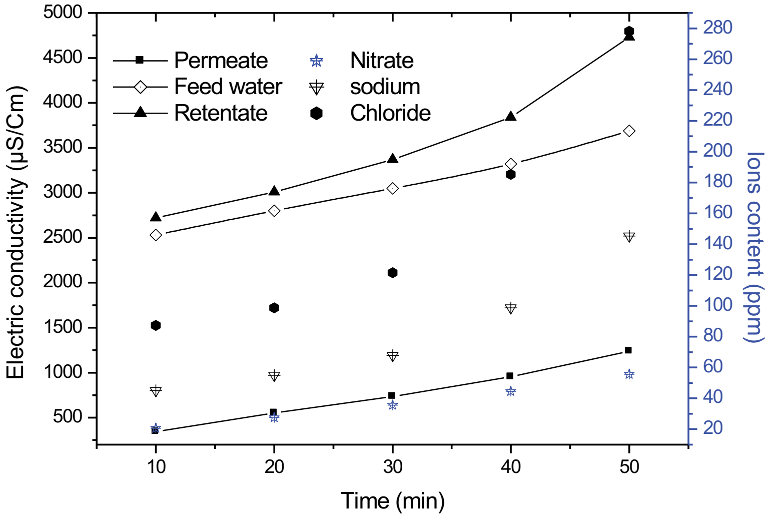

Fig. 4. Evolution of electric conductivity (of permeate, retentate and feed water) and ions content in the permeate as a function of time.

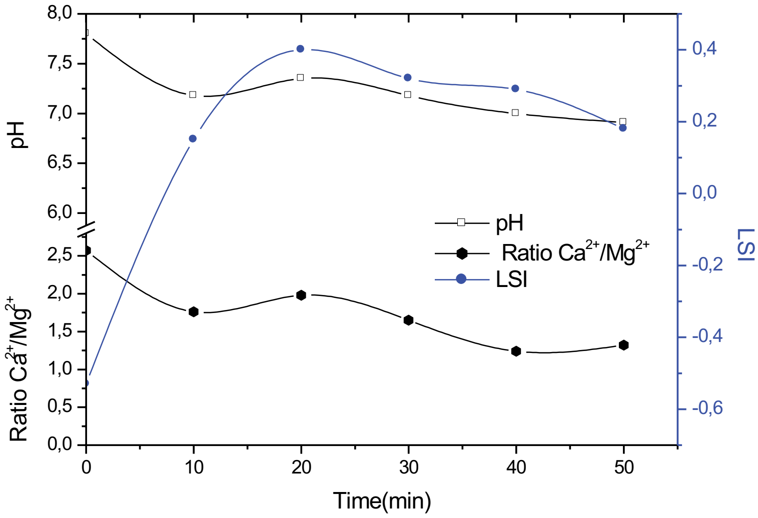

Fig. 5. Evolution of concentration ratio Ca^{2+}/Mg^{2+}, LSI and pH in NF retentate as a function of time.

in the same fashion. The permeates of all modules are collected as product water. Fig. 6 shows the evolution of recovery rate and electric conductivity permeate and retentate as a function of the number of modules. The results indicate that the number of tubes increases, the overall water recovery rate increases due to the greater membrane area deployed (Fig. 6). There is a sharp increase in the recovery rate at the beginning with a slow increase as a number of modules goes higher. This is due to the increase of the salt concentration of the brine (feed water) which feeds the modules of the pressure tube. In the last modules of the tube, the higher salt content generates higher osmotic pressure, which counteracts the effect of the applied pressure. Consequently, the concentration polarization phenomenon that follows is severe. This phenomenon is responsible for the decrease in permeate flow which drops from 1,253 to 825 L/h between the first and the eighth module of the pressure tube. In the same way, the electric conductivity in brine and permeate increase throughout each module due to the reduction of volumetric flux and salts rejection.

All the permeate parameters increase significantly with a number of modules but remain slightly above the standards for a number of modules equal to 6. This configuration leads to the following performances: production capacity of 856.23 L/h, the overall recovery rate of 82.4%, permeate conductivity of 860 µS/cm, nitrate and chloride concentration of 35 and 218 mg/L respectively. Fig. 7 gives the variation of several characteristics of the permeate vs. number of modules (electric conductivity, pH, nitrate, chloride, sodium, calcium and sulfate contents).

Fig. 8 gives the variation of LSI in the permeate and the pH in the retentate, the permeate, and the feed water as a function of a number of modules. It can be seen that with all number of modules, the pH value remains practically stable, while the LSI increases with increasing the number of modules. This behavior is due to the slight increase

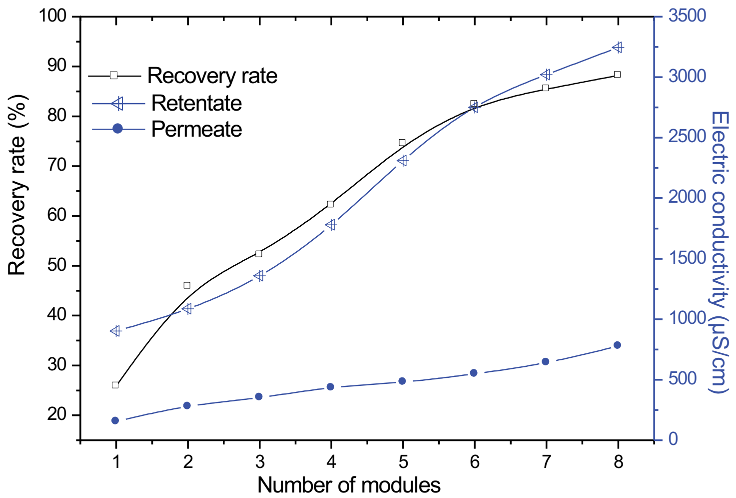

Fig. 6. Evolution of permeate flow and permeate and retentate electric conductivity vs. a number of modules.

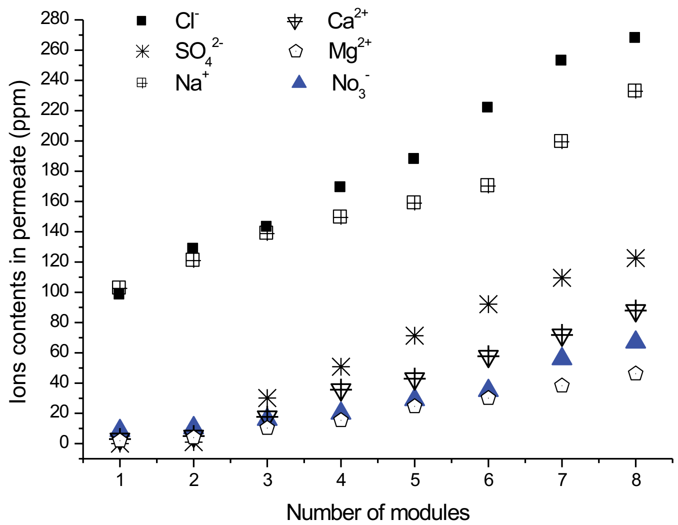

Fig. 7. Variation of ions content in the permeate as a function of a number of modules.

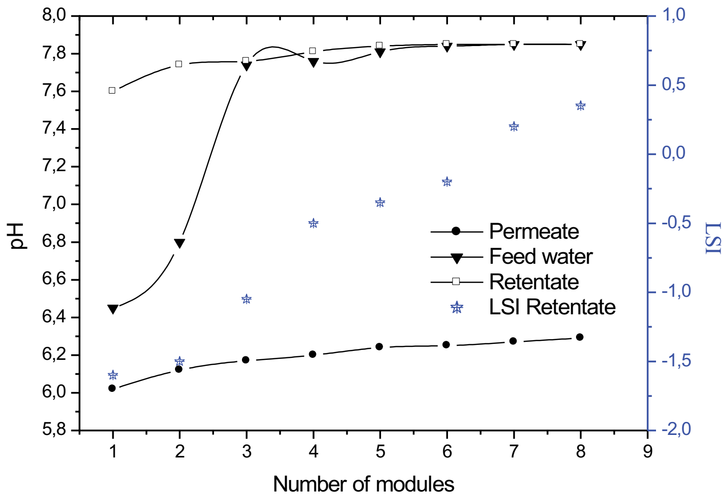

Fig. 8. pH and LSI variation vs. a number of modules.

Table 3
Comparison of energy consumption, energy cost and recovery rate for each tested configuration

	Simple pass	Supplied batch T = 40 min	Pressure tube n = 6
η (%)	80	80	80
TMP (bar)	10	10	10
Y (%)	35	52	82
NO_3^- content (ppm)	12	44.45	35.23
Cl^- content (ppm)	135.89	185.45	218
Na^+ content (ppm)	84.75	98.26	170
Electric conductivity (µS/cm)	234	985	860
Concentrate flow (L/h)	1050	623	478
Energy consumption (kWh/m^3)	0.99	0.66	0.58
Energy cost ($/m^3)	0.099	0.066	0.060

of calcium and carbonate concentration in the permeate. The results indicate that LSI is higher than 0 from the 7th module. Therefore, no scaling phenomenon is observed in the retentate. Previous works on BW indicate that for LSI close to 1.8, no serious scaling problems were observed [27].

3.4. Comparison of energy requirements in different configuration modes

Energy and its cost are one of the most crucial factors in water treatment since the energy required will determine the viability of the treatment process. This last part will focus on the energetic consideration for the three studied configurations in this paper: simple pass supplied batch and pressure tube. The calculation of the overall water recovery rate and energy consumption values for the three configurations is based on the analytical expressions. Knowing that the public cost of electricity in Morocco is close to $ 0.1/kWh [28], Table 3 summarizes energy consumption; energy cost estimates and recovery rate for each tested configuration.

From the results reported in Table 3, it appears that both configurations supplied batch and pressure tube are more technologically attractive thanks to their remarkable improvement over the conventional simple pass in terms of recovery rate, energy consumption and nitrate content in permeate when removing nitrate from brackish water. Through this analysis, it appears that practical batch and pressure tube configuration offer comparable and significant energy savings and good permeate quality. As a matter of fact, on the basis of the above analysis, it can be concluded that the energy consumption depends on the used configuration mode.

4. Conclusion

Altogether, the results confirm the performances of NF to remove nitrate for BW at different configuration modes under Moroccan Standards. Therefore, the quality of the produced water depends on the configuration mode and the working conditions. In a simple configuration, the quality of the produced water is poor in terms of nitrate content and all other physico-chemical parameters, which are below the Moroccan standards. Hence, remineralization of this water is obligatory. In supplied batch, All the permeate followed parameters increase practically over time while the feed water parameters increase considerably. In the retentate, precipitation occurs at 20 min, the precipitation is observed with the naked eye after 40 min of treatment, as well as the nitrate, chloride and sodium contents exceeded the Moroccan recommended values. While, in pressure vessel configuration, the satisfactory permeate water quality that meets the WHO standard, is obtained at the output of the sixth module. Hence, the produced water requires a slight post-remineralization to rebalance it again before disinfection. Also, the results provide that the potential of energy efficiency of batch and tube pressure configuration modes are very close, 0.66 and 0.58 (kWh/m^3) respectively. For the simple configuration, the energy consumption is 0.99 kWh/m^3. Nevertheless, in terms of recovery rate and nitrate content, the pressure vessel is more technologically attractive with a target recovery rate of 82% in comparison to batch configuration (52%) and simple configuration (35%). Moreover, the pressure vessel configuration can produce a brine without scaling phenomenon (LSI > 0), which presents no risk to the installed equipment. While for the supplied batch configuration the scaling potential is significant under working conditions, which requires preventive action aimed at controlling and adjusting the calco-carbonic balance by correcting the pH of water in the feed tank. At the same time, batch mode and pressure tube configuration minimize the fraction of brine disposal dumping to sewage, making it environmentally friendly.

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