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Use of Nanofiltration for Nitrate Removal from Gaza Strip Groundwater

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Abstract— Due to excessive usage of nitrate fertilizer in agriculture and discharging of wastewater from treatment plants, and leakage of wastewater form cesspools, nitrate level in the groundwater has increased. Elevated nitrate in water resources could lead to serious problem including eutrophication, and potential hazards for human and animal health. The aim of this study is to investigate the use of Nanofltraiton for nitrate removal in Gaza Strip as case study. One commercial membrane (NF90) was used in this study. The stirred dead end flow model was used. In addition, two types of water were used: Aqueous solution and real water. The performance of the tested membrane was measured in terms of flux rate and nitrate rejection under different operation conditions: nitrate concentration was varied between 50-400mg/L, applied pressure (6-12) bar and TDS concentration (500-3570) mg/l. The percentage of nitrate removal was in the range of 0.62% and 66.68% and the flux rate ranges between 2.61 and 30.12 L/m^2.hr. These values depend on operation conditions such as nitrate concentration, TDS compostion and operation pressure. In real water, the percentage of nitrate removal was influenced by TDS value in general, but to be more specific, it was found that the concentration of sulphat has a great effect on nitrate removal, as the sulphat concentration increased the nitrate removal decreased. NF90 was observed to be an effective membrane for nitrate removal of Gaza Strip at higher permeate flux and lower applied pressure, especially in North Gaza Strip were low TDS and Sulphat concentration were observed.

Index Terms— Nanofiltration, Nitrate, Rejection, Flux Rate, Well, Total dissolved solids and Pressure.

I INTRODUCTION

Water is essential to sustain life, and a satisfactory (adequate, safe and accessible) supply must be available to all. Improving access to safe drinking water can result in tangible benefits to health.

The Gaza Strip is a highly populated, small area in which the groundwater is the main water source. During the last few decades, groundwater quality has been deteriorated to a limit that the municipal tap water became brackish and unsuitable for human drinking consumption in most parts of the Strip. The aquifer is intensively exploited through more than four thousands of pumping wells. As a result of its intensive exploitation, the aquifer has been experiencing seawater intrusion in many locations in the Gaza Strip; In addition high nitrate is measured in many places in Gaza strip aquifer [1].Nitrate in the groundwater in the Gaza Strip has become a serious problem in the last decade. As a result of extensive use of fertilizers, discharging of wastewater from treatment plants, and leakage of wastewater form cesspools, increased levels of nitrate, up to 400 mg/L, have been detected in groundwater. Nitrate concentrations more than 50 mg/L are very harmful to infant, fetuses, and people with health problems.

To overcome this serious situation, the reverse osmosis (RO) technology is used to replace the tap water or to improve its quality. Several private Palestinian water investing companies established a small-scale reverse osmosis (RO) desalination plants to cover the shortage of good quality drinking water in the whole Gaza Strip.

Desalination is a considerable alternative for water supply in order to improve the quality of water in the area. So, desalination plants began to be established in Gaza strip using RO technique. The shortage of energy source become a big constrain facing desalination plants of which these plants are operating at limited operational hours, The need to find more choices to develop water sector in Gaza Strip become an essential priority. Thinking of innovative actions for desalination sector needs balance and acceptable decisions [2].

New technologies including nanofiltration membrane (NF) application will be considered and experimentally investigated to measure the possibility of enhancing the performance of the desalination plants and increasing production in the near future. In addition, effluent brine treatment technology prior to disposal may be studied and recommended [3].

Nanofiltration (NF) is a suitable method for the removal of a wide range of pollutants from groundwater or surface water. The major application of NF is softening, but NF is usually applied for the combined removal of NOM (Natural organic material), micropollutants, viruses and bacteria, nitrates and arsenic, or for partial desalination. Industrial full-scale installations have proven the reliability of NF in these areas [4].

In the Gaza Strip there is no desalination plant using nanotechnology, the aim of this research to test if Nanofiltration membrane is suitable for nitrate removal from groundwater.

II Experimental Setup

A Materials

NF90 (DOW Filmtec) nanofiltration element is a high area, high productivity element designed to remove a high percentage of salts, nitrate, iron and organic compounds such as pesticides, herbicides and THM precursors. The high active area membrane combined with low net driving pressure of the membrane allows the removal of these compounds at low operating pressure. The system consists of HP4750 stainless cylindrical cell purchased from Steirlitech - UK with volume of 300mL. The cell is pressurized via Nitrogen Gas supplied by Gas cylinder with a manual pressure regulator. The experiments are conducted at room temperature and at pressure range of (6 – 12) bar; Figure 1 shows the system component.

Figure 1 system component.

B Sampling

The filtration experiments were carried out on different samples:

1) Pure sample: deionized water with EC=7μS/cm

2) Synthesis standard solutions: (50-10-150-200-250-300-350-400) ppm as NO_3 Solution.

3) Real sample: Water samples were collected from different municipal wells distributed on all Gaza Strip governorates and divided based on the concentration of Nitrates, the sample Nitrates concentrates are chosen every fifteen mg/L, the concentrations of Nitrates varied between (32-364) mg/L. The water samples were collected based on Palestine Water Authority (PWA) chemical tests results in 2011.

C Methods

After collecting the samples, major chemical analysis were performed for these samples such as (pH, TDS, and NO_3).

Nitrate Measurement

4500-NO3 nitrogen (nitrate) method was used in nitrate measurement. Nitrate concentration was determined by CT-2600 Spectrophotometer.

TDS Measurement

Concentration of TDS was determined by Conductivity meter (Microprocessor conductivity meter BODDS-307W, which measures the EC. To get the approximare TDC value we multiply EC by (0.6).

PH Measurement

PH is a logarithmic notation used to measure hydrogen activity (i.e., whether a solution is acid or basic).

$$pH = - \log [H+] \quad (1)$$

As a simplification, it is assumed that pH is a function of the hydrogen ion concentr tion {[H+]} when in reality it is related to the hydrogen ion activity H+. Since pure water is slightly ionized, it is expressed as an equilibrium equation termed the ion product constant of water. The concentration of these two ions is relatively small and is expressed as a simple logarithmic notation. pH is the negtive log of the hydrogen ion[5] .

The pH was measured with (pH/ORP/ISE Graphic LCD pH Bench top Meter, HANNA instrument) pH meter.

D Tested parameter

Flux Rate

Flux rate Represent the volume of liquid passing through specific area of membrane at certain operating pressure during a period of time.

The flux rate of a filter is important in determining how rapidly filtration can be completed. If there is nothing in the sample stream to clog the pores, the flux rate should remain constant.

$$\text{Flux rate} = V/A.t \ (l/m^2.hr) \quad (2)$$

Where;

V: volume of water permeated at the time (t) (l).

A: surface area of membrane (0.00146 m^2).

t: time of filtration(hr).

Note that these tests were carried out at different pressures (6, 8, 10, 12 bar), because this pressure ranges are lie in the operation pressure range of NF membrane (Filmtec membranes product information).

Rejection

The same meaning of removal efficiency, represent the ability of membrane to reject salts and impurities from feed water. This is one of the most important characteristics of membrane; that's depended on the feed water characteristics, membrane characteristics and applied pressure. The ability of membrane to reject TDS & NO3 was measured using the following equation:

$$\%R= (1-Cp/Cf)*100 \quad (3)$$

Where;

Cp: salt concentration in permeate (mg/l).

Cf: salt concentration in feed water (mg/l).

III Result and discussion

A Flux rate

1 A queues solution

Many factor influence the flux rate such as operation pressure and ionic concentration Figure 2 illustrate the relation between flux rate and operation pressure for pure water sample. Flux rate dos not only depend on the operating pressure but also on the influent concentration.as ionic concentration increase the flux rate will be decrease as show in Figure 3 the effect of operating pressure and ionic concentration on flux rate in nitrate solution sample. For each pressure, a linear relation can be obtained for flux rate against the feed nitrate concentration with high correlation ranges between (0.94 to 0.97). This reduction in flux crossing is increased when the ions is added, probably due to increasing solution osmotic pressure.

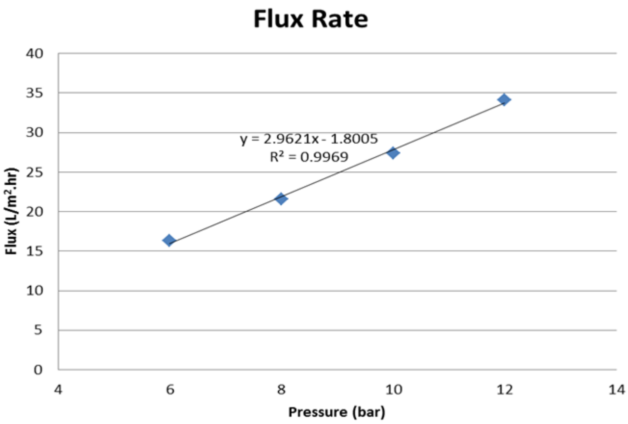


Figure 2 pure water flux rate with different pressure.

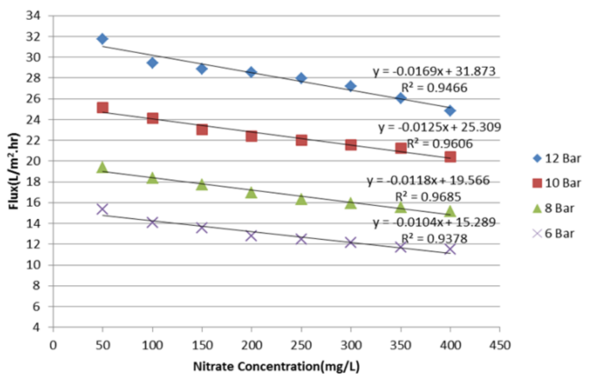


Figure 3 Effect of feed nitrate concentration and opreating pressure on flux rate (nitrate sample (50,100,150,200,250,300,350 and 400 mg/l as NO_3.

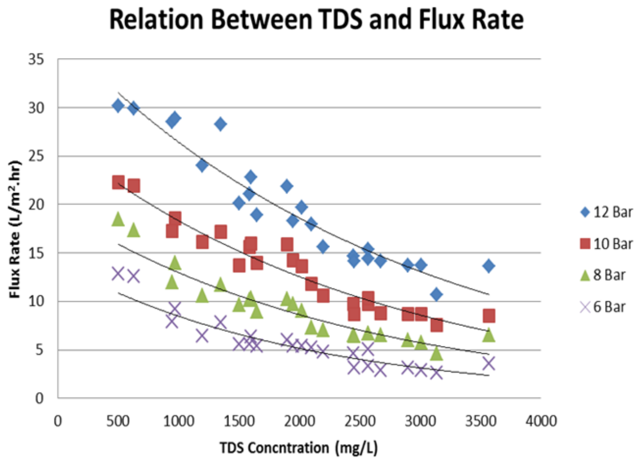
2 Real water sample.

As in case of a queues solution, the flux rate increases linearly with increase of applied pressure Figure 4 and Table 1 show the effect of TDS concentration and operating pressure on flux rate, the general trend is as TDS concentration increases the flux rate decrease

B Rejection of ionic component

1 A queues solution

Figure 4 Effect of TDS concentrations on flux.



The nitrate removal (rejection rate) of solution at different pressure were analyzed Figure 5 shows the effect of operation pressure and ionic concentration on nitrate rejection, as pressure increased nitrate rejection increased on the contrary as nitrate feed concentration increases nitrate rejection decreases. This can be explained by considering salt transport through the membrane as a result of diffusion and convection, which are respectively due to a concentration and a pressure gradient across the membrane. At low trans-membrane pressure

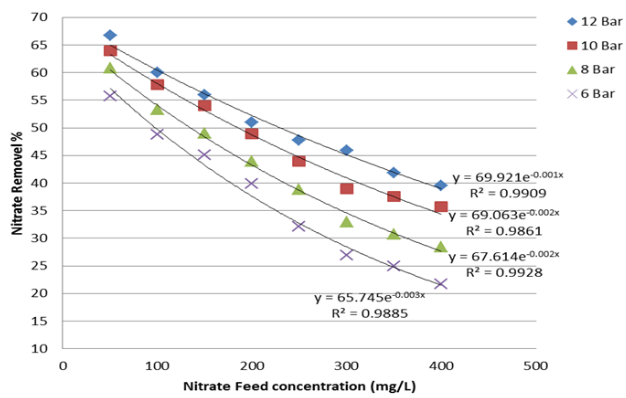


Figure 5 Effect of feed nitrate concentration and opreating pressure on nitrate rejection rate(nitrate sample (50,100,150,200,250,300,350 and 400 mg/l as NO_3).

(TMP), diffusion contributes substantially to the salt transport resulting in a lower retention. With increasing TMP, the salt transport by diffusion becomes relatively less important, so that salt retention is higher [6] [7].

Table 1 Flux rate and TDS concentration.

Well ID.	*TDS(mg/L)*	**Pressure (Bar)**			
		6 Bar	***8 Bar***	***10 Bar***	***12 Bar***
		Flux rate (L/m²．hr)			
A211	500	12.82	18.45	22.29	30.12
D75	630	12.57	17.37	21.95	29.87
D60	950	7.94	12.01	17.24	28.48
W2	970	9.22	13.93	18.54	28.86
Darage	1200	6.42	10.6	16.17	23.98
Hera	1350	7.79	11.69	17.13	28.24
S69	1506	5.57	9.6	13.72	20.16
R306	1587	5.55	10.18	15.64	21.1
C79A	1600	6.36	10.37	15.94	22.8
P145	1650	5.4	8.98	13.95	18.92
R25A	1900	6.04	10.28	15.85	21.82
L127	1950	5.35	9.69	14.19	18.26
R25B	2020	5.35	9.07	13.59	19.65
L198	2100	5.18	7.27	11.82	17.96
R74	2200	4.78	7.06	10.56	15.62
L87	2450	4.61	6.53	9.73	14.7
H104	2454	3.11	6.4	8.7	14.12
R311	2570	3.35	6.7	9.73	14.4
Shoot	2574	5.03	6.72	10.33	15.38
Seka	2673	2.92	6.53	8.81	14.12
Astath	2900	3.11	5.97	8.7	13.7
G49	3010	2.9	5.78	8.66	13.7
E124A	3140	2.61	4.65	7.55	10.71
L190	3570	3.61	6.49	8.53	13.61

2 Real water samples.

As observed in aqueous solutions the effect of operating pressure was evaluated. In real water there were many factors that influenced the rejection percentage such as TDS concentration and other chemical concentration.

The result show that as operation pressure increases the removal of nitrate increases. However, for other wells, the operating pressure was not the main influencing factor. TDS concentration plays an important role. Table 2 shows the results of nitrate removal and operating pressures, the maximum rejection percentage at 12 bar was 55.56% at well A211 and the minimum nitrate rejection was zero at many wells when operating pressures was 6 bars depending to TDS concentration and composition and nitrate concentration in feed water

The result in Figure 6 showed that in general that a relation between TDS concentration and nitrate rejection, when we fixed the nitrate concentration in feed water. As shown in Figure 6 there are drop in curve, but when the effect of nitrate concentration is fixed and plot the nitrate removal and sulphate concentration, a strong relation between sulphate concentration and nitrate rejection was found (Figure 7).

Figure 6 and Figure 7 show the nitrate rejection results against TDS and sulphat concentration. To show this relation, Nitrate concentration must be fixed. For example E124A well have 3140 mg/l as TDS concentration and S69 well has 1506 mg/l as TDS concentration, but nitrate rejection in E124A is higher than S69, although nitrate concentration in E124A is higher than in S68. This was due to that the sulphate concentration in E124A was 149 mg/L but in S69 was 240 mg/L. That means the sulphate concentration plays important role in NF90 nitrate rejection percentage.

Because of high removal of sulphate, because of their valance, nitrate is forced to pass through the membrane. The removal of monovalent such as nitrate was greatly decreased under the presence of sulphate ions. Retention of the negative sulphate ion in concentration water disturbed the electrical equilibrium on both sides of the membrane that the nitrate ions was forced through the membrane in permeate water to maintain electric equilibrium [8].

It was also observed that an increase of sulphate concentration generally decreases the chloride rejection. The retention of chloride anion is lower for the salt mixtures than for single salts experiment. It seems that the presence of high valance anion (SO4) drives more chloride into membrane, thus decreasing its retention [9].

The sequence of rejection of monovalent anions can be written as R (F)> R (Cl)> R (NO3), the observed retention of the three ions is similar to the ionic order and opposite to the hydration energy order for the monovalent ions, the F which has higher hydration energy is better retained than Cl and NO3 [10] [11].

From the above two paragraph it can be conclude that the chloride is better than nitrate in rejection according to rejection sequence, while sulphat has negative effect on chloride rejection so sulphate has negative effect on nitrate rejection.

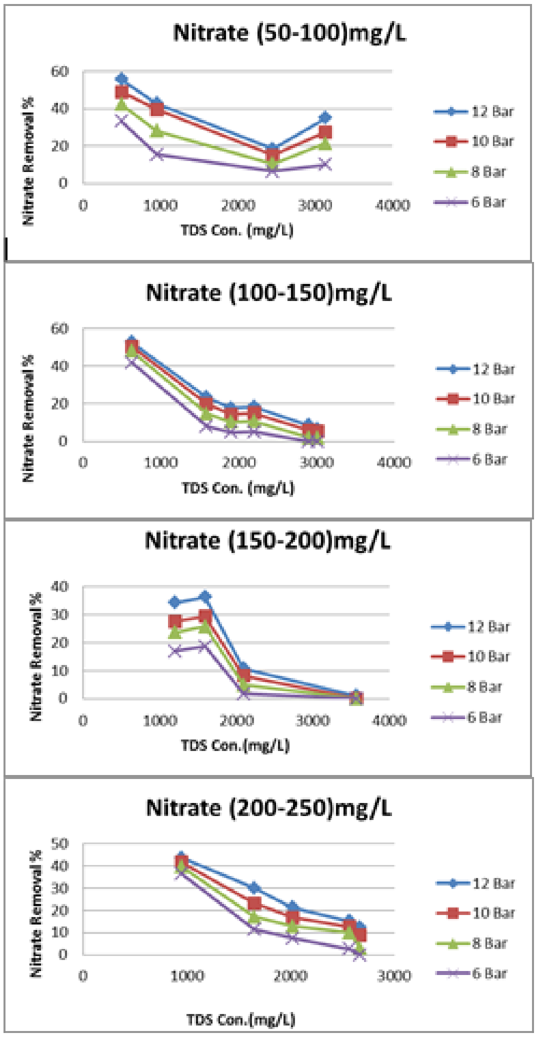


Figure 6 Relation between TDS concentration and nitrate rejection.

Table 2 Nitrate rejection result with sulphate and TDS concentration.

Well No.	*TDS*	*Nitrate NO3 (mg/LNO3)*	*Sulphate SO4 (mg/L)*	*6 Bar*	*8 Bar*	*10 Bar*	*12 Bar*
A211	500	45	22	33.33	42.22	48.89	55.56
W2	970	71	108	18.31	28.17	39.44	42.85
E124A	3140	80	149	75	21.25	27.5	35
S69	1506	32	240	8.23	15.63	22.32	28.13
H104	2454	76	394	5.35	10.6	15.15	18.5
D75	630	133	41	42.87	48.12	50.38	52.63
R306	1587	136	155	8.09	14.71	19.85	23.53
R74	2200	120	219	5	10.83	15	18.33
R25A	1900	146	269	4.79	10.27	14.38	17.81
Astath	2900	140	407	0.71	2.14	5.73	8.57
G49	3010	138	550	0.96	1.79	5.17	6.79
C79A	1600	190	105	16.84	19.47	24.21	36.63
Darage	1200	178	111	16.85	23.6	27.53	34.27
L198	2100	185	375	1.62	4.86	8.11	10.81
L190	3570	193	628	0.52	1.04	1.55	2.07
D60	950	211	90	36.49	39.81	41.71	43.6
P145	1650	206	213	6.31	13.59	23.3	30.1
R25B	2020	226	280	7.52	12.83	16.81	21.24
Seka	2673	230	359	2.17	10	12.61	15.22
R311	2570	217	444	1.38	3.23	8.76	12.44
Hera	1350	273	135	21.61	31.87	36.26	45.42
L127	1950	364	157	18.68	23.9	29.67	32.97
L87	2450	304	271	4.61	9.21	14.47	17.76
Shoot	2574	332	356	0.60	1.2	7.83	14.16

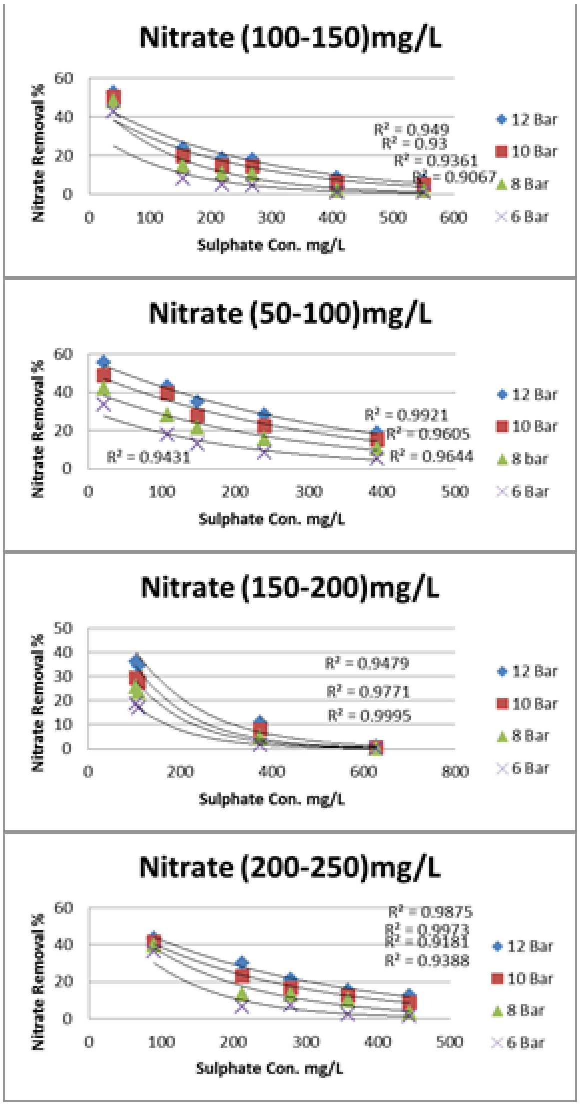


Figure 7 Relation between nitrate rejection and sulphate concentration.

IV Comparison between Real water and Aqueous Solutions:

1) Flux Rate

The performance of NF90 membrane varied in terms of flux rate. Consequently, the pure water flux rate was higher than the real water flux rate.

As the water contains more salts or other substances, the flux rate decreases. At this pattern the membrane performance, so the pure flux rate was higher than of real water flux. Also complexity of water character play a good role in membrane behavior and that is why the queues solution flux rate is higher than real water flux rate.

The maximum flux rate for aqueous solution was obtained at 12 bar (34.13 L/m^2.hr) for pure water and minimum flux rate was obtained at 6 bar (16.31 L/m^2.hr).

The maximum flux rate for real water was obtained at 12 bar (30.12 L/m^2.hr) for A211 and minimum flux rate was obtained at 6 bar (2.61 L/m^2.hr) for E142A.

2) Nitrate Rejection

Generally, the overall rejection percentages of the NF90 membrane of aqueous solutions were found to be higher than the rejection of real water. For aqueous solution the maximum and minimum nitrate rejection of aqueous solution was 66.68% and 21.67% respectively, while for real water the maximum and minimum nitrate rejection of were 55.56% and 0 % respectively.

The characteristics of feed water significantly affect the membrane rejection such as the content of sulphate and hardness. This explains the difference of rejection between real water and aqueous solution. In addition, real water may contain some colloids and many other substances that can negatively affect the membrane rejection.

Conclusion

NF90 membrane showed good result for nitrate removal in real water, which varied between 0.62% and 55%, and flux rate between 2.61 and 30.12 L/m^2.hr, when the operating pressure varied between 6 and 12 bar.

It can be concluded that the sulphate has negative effect on chloride rejection and on nitrate rejection. As the real water contains more salts or other substance, the flux rate decrease. At this pattern the membrane performance, so the pure flux rate was higher than of real water flux. Also complexity of water character play a good role in membrane behavior and that is why the nitrate solutions flux rate are higher than real water flux rate.

NF90 was observed to be an effective method to nitrate removal of Gaza Strip at higher permeate flux and lower applied pressure, especially in North Gaza Strip were low TDS and Sulphat concentration were observed. In other Gaza Strip places TDS and sulphat should be removed before using nanofiltration to nitrate removal.

The characteristics of feed water significantly affect the membrane rejection such as the content of sulphate and hardness. This explains the difference of rejection between real water and aqueous solution.

Sensitivity of the system to the circumstances like temperature, quality of deionized water used in system flushing, regular insurance of zero leakage of pressure , the period of using membrane, using tools washed by deionized water , all these restriction make the test harder.

The importance of testing Nanofiltration membranes as new emerging technology in Gaza strip is to improve the overall desalination quality with acceptable cost; carrying out tests helps to understand the behavior of NF90 for nitrate removal.

Desalination of brackish water using Nanofiltration technique is seen as one of the promising solution that can assist Gaza in filling the gap between the growing needs for water, limited water recourses, limited energy resource, the standard of domestic water and unacceptable water quality.

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