



Comparison of reverse osmosis and nanofiltration membranes in water purification process

Su arıtma prosesinde ters osmoz ve nanofiltrasyon membranlarının karşılaştırılması

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Abstract

Water is indispensable for the continuation of life. However, fresh water resources in the world are decreasing day by day. For this reason, effective management of existing water resources and treatment of wastewater becomes important. In this study, two prominent membrane filtration processes in water treatment, reverse osmosis and nanofiltration processes, were compared. In this comparison, commercial NF90 nanofiltration membrane and BW30 reverse osmosis membrane from the same manufacturer were used. Tap water, 2000 ppm NaCl and ultrafiltration filtrate of synthetic oily solution were chosen as the feed solution. Only pretreatment was carried out by ultrafiltration process. Although the total dissolved solids rejection rates of both membranes are close to each other, the flux obtained with the NF90 membrane is approximately twice the flux obtained with the BW30 membrane at all pressure values. Since both membranes consume the same amount of energy in water treatment, it has been concluded that water treatment with NF membranes is more economical.

Keywords: Water purification, Reverse osmosis, Nanofiltration, Membrane filtration.

Öz

Su yaşamın devamı için vazgeçilmezdir. Ancak dünyadaki tatlı su kaynakları her geçen gün azalmaktadır. Bu nedenle mevcut su kaynaklarının etkin yönetimi ve atık suların arıtılması önem kazanmaktadır. Bu çalışmada su arıtımında öne çıkan iki membran filtrasyon prosesi, ters osmoz ve nanofiltrasyon prosesleri karşılaştırılmıştır. Bu karşılaştırmada aynı üretici firmaya ait ticari NF90 nanofiltrasyon membranı ve BW30 ters osmoz membranı kullanıldı. Besleme çözeltisi olarak ise musluk suyu, 2000 ppm NaCl çözeltisi ve sentetik yağlı çözeltinin ultrafiltrasyon süzuntüsü seçildi. Ultrafiltrasyon işlemi ile yalnızca ön arıtma gerçekleştirildi. Her iki membranın toplam çözünmüş katı madde reddi oranları birbirine yakın olmakla birlikte, NF90 membranı ile elde edilen akı tüm basınç değerlerinde BW30 membranı ile elde edilen akının yaklaşık iki katıdır. Su arıtmada her iki membran da aynı miktarda enerji tükettiğinden NF membranlarla su arıtmanın daha ekonomik olduğu sonucu elde edilmiştir.

Anahtar kelimeler: Su arıtma, Ters osmoz, Nanofiltrasyon, Membran filtrasyonu.

1 Introduction

Water is an indispensable element for the continuation of life on Earth. However, the amount of fresh water available in the world is limited. Approximately 3% of the water in the world is fresh water, while the rest is saline. The amount of fresh water suitable for the use of humanity is only 1% of all water in the World [1]. According to the drinking water report published by the World Health Organization (WHO) in 2019, half of the world's population will be living in water stressed areas by 2025 [2],[3]. Only 11% of fresh water is spent for domestic use, while 19% is used for industry and 70% for agriculture. Considering its place in agriculture, it can be said that water also means food. Although water is such an important component for the world, increasing population, overuse/misuse of water, improper management of water, pollution of water resources and climate changes show that humanity may face water scarcity in the future [4]. In order to prevent the predicted water shortage, existing water resources should be managed effectively and wastewater should be treated and reused. Wastewater can be treated by various

physical, chemical and biological methods [5] such as flotation [6], precipitation [7], oxidation [8], solvent extraction [9], evaporation [10], carbon adsorption [11], ion exchange [12], phytoremediation [13], electrochemistry [14], biodegradation [15] and membrane filtration [16]-[18]. The membrane filtration method stands out among these methods due to its features such as high removal efficiency, compactness, simple design, ease of use, not needing additional chemicals and being economical [19]-[21]. Membrane filtration processes can be divided into 4 main groups according to the differences that create the driving force across the membrane. These differences; pressure, temperature, concentration and electric potential differences [22],[23].

Today, pressure driven membrane processes (PDMPs) are recognized worldwide as a key element in sustainable water management systems due to their unique advantages and versatility [24],[25]. PDMPs are classified as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) according to the pore size of the membrane used and applied pressure ranges [26]. Pore sizes and pressure

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ranges of membranes used in MF, UF, NF, and RO can be found in Table 1.

Table 1. Pore size and pressure range informations of PDMPs [22].

Process	Pore Size (μm)	Pressure (bar)
Microfiltration (MF)	10 – 0.05	0.1 – 2
Ultrafiltration (UF)	0.05 – 0.002	1 – 10
Nanofiltration (NF)	0.002 – 0.001	5 – 20
Reverse osmosis (RO)	< 0.001	10 – 100

Microfiltration is a process frequently used to concentrate, purify or separate macromolecules and suspended particles from solution [27]. MF is one of the oldest commercially available PDMPs. Since MF membranes have low hydrodynamic resistance, they require low hydrostatic pressure in separation processes. However, the wide pore size range allows MF processes to be used in many different areas [28]. Ultrafiltration is also a low pressure membrane filtration method like MF. The bacteria and most of viruses can be removed using ultrafiltration membranes [29]. UF processes are used in areas such as the recovery of chemicals, waste water purification, fruit juice concentration, as well as in the medical field [30]. In addition to all these different areas of use, there have been many recent studies using MF and UF as pre-treatment in NF and RO processes [31]-[34].

Dissolved substances containing singly charged ions such as Na^+ , Cl^- are separated from water using a semi-permeable membrane in RO processes. Separation is achieved by the passage of water through a semi-permeable membrane as a result of the application of an external pressure to overcome the osmotic pressure [35],[36]. RO can be defined as a diffusion controlled process. This is because the mass transfer in RO membranes is controlled by the solution diffusion mechanism. In this mechanism, the filtrate dissolves in the membrane material and then diffuses through the membrane [36],[37]. RO membranes are very hydrophilic so that water can quickly diffuse into and out of the polymer structure. In addition, they effectively have a non-porous structure and exclude even any low molar mass species [35],[36]. Water flux and salt rejection rate are two main parameters for RO membranes. It is desirable that both of these parameters be as high as possible. In addition to these parameters, an ideal RO membrane should have good chlorine and fouling resistance, mechanical strength and low cost [35]. Local governments and other water suppliers are turning to RO to meet the ever-increasing demand for fresh water. In recent years, the application of RO technology has increased noticeably [38]. By 2009, RO plants comprised almost half of the world's water treatment plants [39].

Nanofiltration, defined in the late 1980s, was defined as a process that rejects molecules whose size is on the order of one nanometer. This caused NF to be positioned between UF and RO [40]. Due to the advantages of nanofiltration such as low processing pressure, high flux, high retention rate of multivalent anion salts and an organic molecular above 300 Da and relatively lower cost, interest in its applications has increased worldwide [41]. Typical NF membranes have a pore size of 1 nm. This pore size corresponds to a molecular weight limit of 300-500 Da. Similar to RO membranes, NF membranes are highly effective at separating inorganic salts and small organic molecules. Besides, they have higher flux, lower rejection of monovalent ions and higher rejection of divalent ions compared to RO membranes [42]. These properties have allowed NF to be used in many fields, especially in water

treatment, biotechnology, and food engineering [42],[43]. NF, the most recently developed PMDP, has already replaced RO in many areas with the developed membranes [44]. Compared to RO, success of NF is usually due to the selective separation of one solute over another [45].

In this study, 3 different solutions; municipal tap water, 2000 ppm NaCl solution and permeate of synthetic oily solution from UF process were purified using NF and RO membranes and the results were compared. UF was used as the pretreatment process for the synthetic oily solution. In order to find an answer to the question of whether higher purity water can be obtained using the two-stage membrane process, the permeates obtained from the NF and RO processes were used as feed solutions and treated with the RO membrane again and the results were examined.

2 Material & Method

2.1 Material

NaCl used in 2000 ppm NaCl solution was purchased from Sigma Aldrich company and used without any treatment. Dow Filmtec's NF90-2540 and BW30-2540 membranes were used in NF processes and RO processes, respectively. The operating conditions of the membranes used, shared by the manufacturer, are presented in Table 2.

Table 2. Operating conditions of the membranes used in NF and RO processes.

Property	Membrane	
	BW30-2540	NF90-2540
Maximum Operating Temperature	45 °C	45 °C
Maximum Operating Pressure	41 bar	41 bar
Maximum Feed Flow Rate	1.4 m ³ /h	1.4 m ³ /h
Maximum Pressure Drop	1.0 bar	0.9 bar
pH Range (Continuous Operation)	2-11	2-11
Free Chlorine Tolerance	< 0.1 ppm	< 0.1 ppm

2.2 Method

In this study, pure water was obtained from 3 different solutions by using NF and RO processes. While the municipal tap water and 2000 ppm NaCl solution were used directly, the synthetic oily solution was subjected to ultrafiltration process to remove the oil. Tap water does not require pre-treatment and does not contain contaminants that would cause plugging up in the membrane pores. Therefore, it is a good feed solution option to test membrane performance. However, in order to see the salt rejection rates of the membranes, 2000 ppm NaCl solution was used as the feed solution. In addition, a synthetic oily solution was chosen as the feed solution in order to examine the effects of UF pretreatment on the obtained ultrapure water. As mentioned in the following sections, pure waters are also classified within themselves. Finally, a two-stage treatment process was carried out in order to see whether more than one membrane treatment affected the purity level of the resulting water. Accordingly, the permeates obtained from RO and NF processes were treated with the RO membrane again as the second stage. Two-stage NF/RO hybrid systems are frequently used, especially in wastewater treatment [46]-[48]. In order to see the difference, in this study, the RO/RO process was also examined in addition to the NF/RO process.

The feed solution was transferred from the tank to the membrane by a pump. The solution transferred to the membrane is purified and the purified fraction is called

permeate while the remainder is called concentrate. The permeate is stored in another tank, and the concentrate is sent back to the feed tank for further purification. For the oily synthetic solution, the permeate of the UF process was used as the feed solution for the NF and RO processes. The schematic representation of the systems used is shown in Figure 1(a) and Figure 1(b).

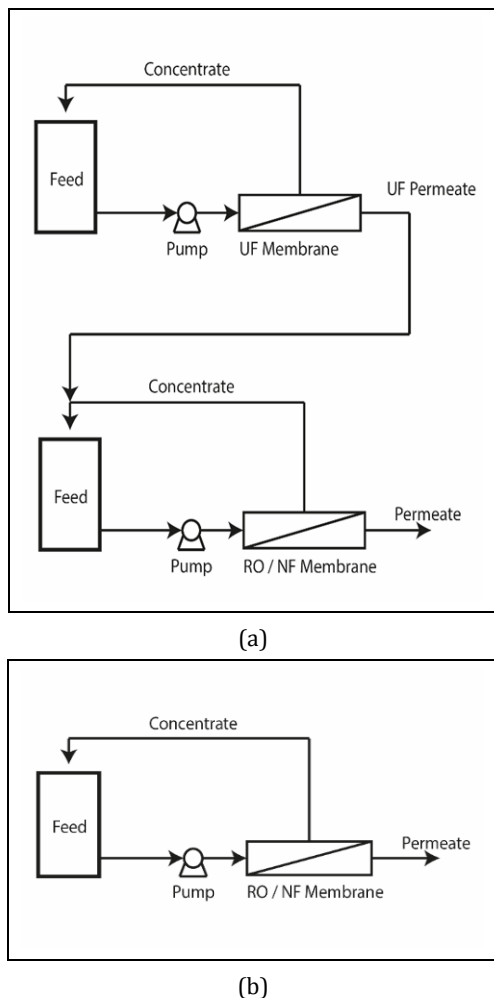


Figure 1. Schematic of the experimental system. (a): System using UF pretreatment, (b): System without pretreatment.

The purity of feed solutions and permeate is determined by measuring their electrical conductivity (EC) and total dissolved solids (TDS) value. There are various standards (ISO 3696 (1987), ASTM (D1193-91), NCCLS (1988), etc.) around the world to classify waters according to their purity. EC and TDS are two parameters that are frequently used and indicate the quality of water. The value of EC and TDS are correlated. EC is the measure of the liquid's capacity to conduct electric charge, while TDS is the measure of the dissolved ion concentration. The ability of a liquid to conduct an electric charge depends on the dissolved ion concentration, ionic strength and temperature of the measurements [49]. The universal standard for assessing the salinity of aqueous systems is electrical conductivity at 25 °C [50]. For this reason, TDS and EC measurements were carried out at 25 °C by keeping the solution temperatures constant at this temperature. TDS and EC measurements were performed using Myron L 4PII Conductivity, Resistivity, TDS, Temperature Meter.

Before starting the experiments, RO and NF systems were operated at 10 bar pressure for 1 hour to stabilize them. Then, the pressure was increased by 5 bars, to maximum of 25 bars. After the system was operated for 30 minutes at each pressure value, samples were taken from feed and permeate solutions and measurements were made. This process was applied to all feed solutions at constant conditions.

In the two-stage purification process, NF and RO permeates of municipal tap water were used as feed solution for the RO process and subjected to a second purification process. Thus, the differences in the use of permeates of RO and NF processes as feed in two-stage purification processes were examined.

3 Results & Discussion

3.1 Municipal tap water

The first measurement was taken at 10 bar pressure. Then, the pressure was increased by 5 bars and the measurement was made up to 25 bar, and the results presented below were recorded. As can be seen in Figure 2(a), higher fluxes were achieved with NF membranes than RO membranes at all pressure values. The highest flux with both membranes was reached at the highest pressure of 25 bar, as expected. While this value was 145 dm³/h for the BW30 membrane used in the RO process, it was measured as 310 dm³/h for the NF90 membrane used in the NF process.

As can be seen in Figure 2(b), the removal rate of both membranes increased as the pressure increased. The TDS rejection percentages of both membranes are very close to each other, although there is more than two fold difference between the fluxes. The highest rejection rate was achieved with both membranes at 25 bar pressure, 99.32% for the NF membrane and 99.55% for the RO membrane.

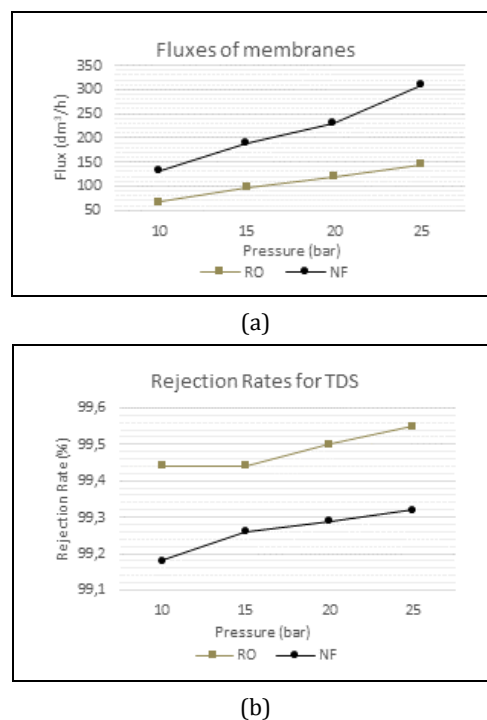
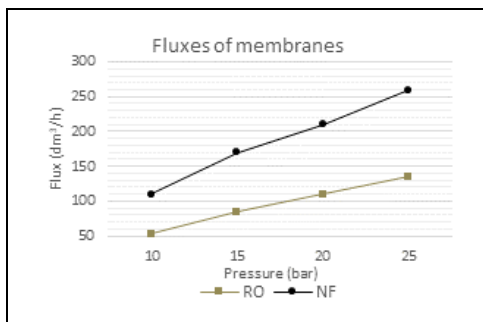


Figure 2. (a): Flux values obtained for municipal tap water with NF and RO membranes, (b): Rejection rates obtained with NF and RO membranes for TDS value of municipal tap water.

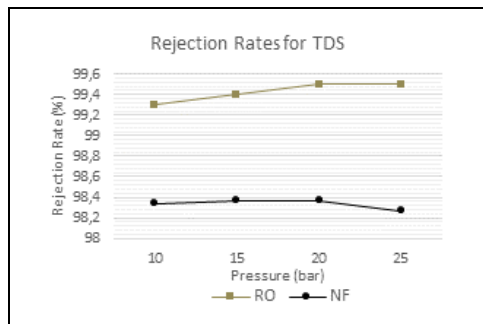
3.2 2000 ppm NaCl solution

The first measurement was taken after the system was operated at 10 bar for one hour to stabilize. The pressure was increased by 5 bars up to a maximum of 25 bar and results was recorded. When the results were examined, it was observed that the fluxes of the NF90 membrane were higher for the NaCl solution at all pressure values. As expected, the increase in pressure also increased the fluxes. However, the fluxes were lower when compared to the results obtained with municipal tap water, as seen in Figure 3(a).

On the other hand, the difference between TDS rejection rates of NF90 and BW30 membranes was higher in this feed solution compared to municipal tap water. While the maximum difference between the TDS rejection rate of NF90 and BW30 membranes for municipal tap water was 0.26 %, the maximum difference was found to be 1.23 % for 2000 ppm NaCl solution. As can be seen in Figure 3(b), while rejection rate was almost the same for BW30 membrane, the small difference in TDS rejection rate of NF90 membrane caused this. The biggest difference between TDS rejection percentages of NF and RO membranes was seen at 25 bar pressure. It was observed that the TDS rejection value of the NF90 membrane decreased when the pressure increased above 20 bar. This is due to the fact that the pore sizes of NF membranes are larger than the pore sizes of RO membranes. While this is an advantage for permeate flux, it creates a disadvantage for rejection rate. The increased pressure causes the salt to pass through the membrane pores along with the water. As the pressure increases, the amount of salt passing through also increases, thus the rejection rate of the membrane decreases.



(a)



(b)

Figure 3. (a): Flux values obtained for 2000 ppm NaCl solution with NF and RO membranes, (b): Rejection rates obtained with NF and RO membranes for TDS value of 2000 ppm NaCl solution.

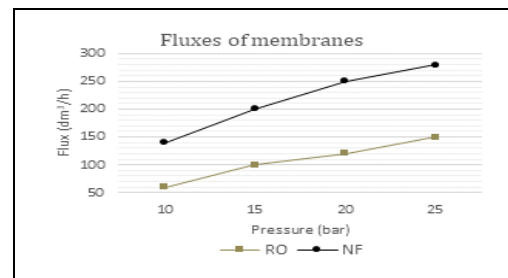
There is an average decrease of 1.09% in the percentage of TDS rejection of the NF90 membrane. The difference can be

attributed to the larger pore sizes of nanofiltration membranes compared to reverse osmosis membranes. Li et al. [51] tested the NF90 membrane with 2000 ppm NaCl solution and measured the TDS rejection as 92.78% at 15 bar pressure in their study. This value is very close to the 99.33% value obtained in the current study.

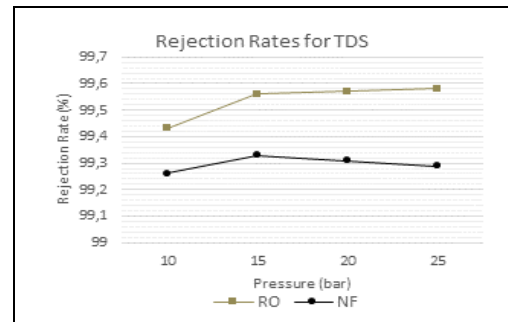
3.3 UF Permeate of synthetic oily solution

The first measurement was taken at a pressure value of 10 bar for the permeate of the synthetic oily solution pre-treated with the UF process. Then, by waiting for half an hour at each pressure value, the pressure was increased by 5 bars and reached up to 25 bar. When the results for the permeate of the UF process are examined, it is seen that the difference between the fluxes decreases relatively with the increase in pressure. At 10 bar pressure, the flux of the NF90 membrane is 2.33 times the flux of the BW30 membrane. However, as can be seen in Figure 4(a), this ratio decreased with the increase in pressure and decreased to 1.87 times at 25 bar.

When the TDS rejections of NF and RO membranes for UF permeate are examined, it is seen in Figure 4(b) that there is no significant change in the TDS values of the BW30 membrane compared other solutions. The percentage of TDS rejection of the BW30 membrane increased with the increase in pressure and reached its highest value at 25 bar pressure. Although the percentage of TDS rejection of the NF90 membrane at all pressure values was close to each other, it reached the highest value at 15 bar. However, there is a decrease in the percentage of TDS rejection of the NF membrane with the increase in pressure.



(a)



(b)

Figure 4. (a): Flux values obtained for UF permeate of synthetic oily solution with NF and RO membranes, (b): Rejection rates obtained with NF and RO membranes for TDS value of UF permeate of synthetic oily solution.

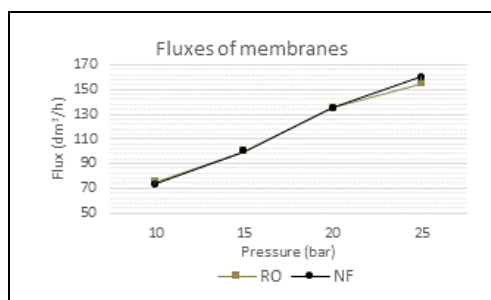
For these three different feed solutions, the flux of the NF90 membrane was higher than the flux of the BW30 membrane. Gündoğdu et al. [52] also used these two membranes in their study on industrial wastewater recovery. They conducted

experiments with NF membranes at 10 bar and with RO membranes at 20 bar. Despite this, they obtained a permeate flux of 50.4 L/m²h with the NF90 membrane and a flux of 65.9 L/m²h with the BW30 membrane. It is seen that the fluxes are relatively close to each other even though the RO membrane is operated at twice the pressure. In the study, it is seen that the permeate flux of NF90 increases nearly two-fold when the pressure increases from 10 bar to 20 bar. Therefore, it is obvious that the flux will increase when the experiment is conducted with the NF90 membrane at 20 bar in the study of Gündoğdu et al. When the TDS values of the obtained permeates are examined, it is seen that there is no big difference between them. While the TDS value of the permeate of the NF90 membrane is 64.0 mg/L, the TDS value of the permeate of the BW30 membrane is 31.3 mg/L. The results of Gündoğdu et al. similarly show that NF90 membranes are more economical for water treatment processes. Although there is a small difference between the rejection rates of the NF90 membrane and the BW30 membranes, much higher flux is obtained with the NF90 membranes.

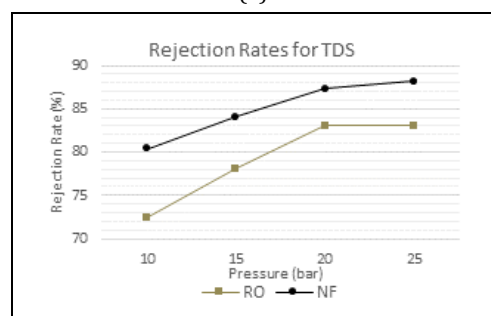
3.4 Two-Stage purification

NF and RO permeates were used as feed solution for BW30 membrane. When the flux of the BW30 membrane was measured for these two solutions, it was seen that there was not much difference between them, as can be seen in Figure 5(a). It was observed that there was a difference of 5 dm³/h between the fluxes obtained for the two feed solutions at only 25 bar pressure. With the BW30 membrane, the highest flux, 160 dm³/h, was obtained at 25 bar pressure when NF permeate was used as the feed solution.

When the percentages of TDS rejection are examined, it is seen that there are relatively large differences for the two solutions. The percentages of TDS rejection of the BW30 membrane for NF and RO permeates are presented in Figure 5(b).



(a)



(b)

Figure 5. (a): Flux values obtained with two-stage purification, (b): Rejection rates obtained with two-stage purification.

The highest difference between TDS rejection percentages for these two feed solutions was obtained as 7.92% at 10 bar pressure. As the pressure increased, this difference decreased slightly and reached an average of 5%. The closest TDS rejection value for these two feed solutions was obtained at 20 bar, the difference in this pressure value was only 4.25%. The reason for this difference being high at the beginning is that the NF permeate feed has a higher TDS value. As the NF permeate feed is treated with the RO membrane, total dissolved solids are removed and the TDS rejection rate approaches the TDS rejection rate of the RO permeate feed.

TDS rejection percentage of over 99% was achieved in one-stage treatment with both membranes. However, TDS rejection percentages of the BW30 membrane are slightly higher than those of the NF90 membrane at all pressure values. This is the reason for the difference in the second stage.

Although higher purity water is obtained by using the two-stage membrane process compared to the single-stage membrane process, the water obtained still does not have a Class 2 pure water degree. Although the values are very close to the Class 2 pure water values according to the ISO 3696 standard, the purity level of the upper class could not be obtained with the two-stage purification process.

4 Conclusion

In this study, different solutions were purified by using NF90 nanofiltration membrane and BW30 reverse osmosis membrane and these two membranes were compared with each other. In addition, NF and RO permeates of municipal tap water were used as feed solution in the RO process and a two-stage purification process was carried out.

The flux increased with the increase in pressure in all processes, as expected. For both membranes, the highest flux was reached at 25 bar pressure, while the highest flux was obtained with municipal tap water for the NF90 membrane, the highest flux for the BW30 membrane was obtained with NF permeate as the feed solution.

When the conductivity values of purified municipal tap water, UF pre-treated synthetic oily solution and two-stage membrane filtration permeates using NF90 and BW30 membranes are examined, it is seen that 3rd degree purified water can be obtained according to ISO 3696 (1987) [53] standard. It has been observed that the conductivity values of permeate in two-stage membrane filtration are quite close to the conductivity values of the second grade purified water class. However, conductivity values of NF and RO permeates of 2000 ppm NaCl solution were higher than these levels. Therefore, permeates of 2000 ppm NaCl solution require a second purification process in order to be included in the purified water grades of the ISO 3696 (1987) standard.

The same pump was used for water purification with BW30 and NF90 membranes and operated at the same pressure values for the same time. However, the flux obtained with the NF90 membrane at all pressure values is approximately twice the flux obtained with the BW30 membrane. Despite the big difference between fluxes, TDS rejection percentages of both membranes were found to be close to each other. It has been determined that it is more economical to use the NF90 membrane in water purification because it consumes the same energy but has approximately twice as much flux.

5 Author contribution statement

Ahmet Bora Yavuz contributed to the creation of the idea, the design, the experiments, the evaluation of the results, and the writing of the article, while Osman Nuri Ata contributed to the literature review, examination of the results, and spelling check.

6 Ethics committee approval and conflict of interest statement

"There is no need for ethics committee permission for the prepared article."

"There is no conflict of interest with any person/institution in the article prepared."

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