DOI: 10.7251/QOL2103093M
UDC: 678.01:541.66]:621.774

Original scientific paper

Performance Evaluation of Polymeric Membrane Permeability Characteristics Using Different Aqueous Solutions

Erhan Mustafa, Katerina Atkovska, Stefan Kuvendziev, Mirko Marinkovski, Kiril Lisichkov
Ss. Cyril and Methodius University in Skopje, Faculty of Technology and Metallurgy,
erhanmustafa1978@gmail.com

ABSTRACT: In the last decade the application of membrane separation technology is more increasing. The membrane in water purification and wastewater treatment is essential separation process used for water reclamation. The production of new membrane types with different permeable characteristics and performances allows them to be fitted in different membrane modules that can be used in the membrane filtration.

The water characteristics are important for the membrane performance. It can seriously affect the permeability characteristics and increase the fouling on the membrane surface. In wastewater treatment, the characteristics of the aqueous influent can reduce the permeability of the membrane and the process efficiency of the membrane bioreactor (MBR). The aim of this paper is to explore the effect of different aqueous solutions on membrane permeability using dead end filtration process. For this purpose, NaCl solution with different concentration were prepared and the effect of the concentration polarization on the membrane was observed. The constructed membrane module was also tested with real water sample and the membrane permeability was analyzed.

In this experiment a polymeric membrane produced from polyether sulphonate (PES), with diameter of 5.0 cm and pore size of 0.04 µm was assembled in a constructed module for dead-end filtration. The module was constructed in a way that would allow turbulence of the solution on the membrane surface. The following working parameters were examined: transmembrane pressure (TMP), the types of solutions, the working temperature, and the influence of agitation on the feeding to the specific membrane flux and permeability. The results showed that the membrane permeability is affected by the water organic and inorganic constituents and in the process of design of membrane reactor for wastewater treatment, the water composition should be taken in consideration.

Keywords: membrane permeability, PES, dead-end filtration module, TMP

INTRODUCTION

The need for clean water is increasing every day. The pollution of a water resources and need for treatment is important key driving factor for development of new purification technologies. The ultrafiltration process uses a membrane as a selective barrier (Abdessemedet al. 2000). The pore size can be in range of 0.01 µm to 0.1. µm (Fane et al. 2010). Choosing the right membrane configuration and its material is crucial for each application, since it depends on obtaining good separation and yields. Ultrafiltration (UF) membranes have been challenged to maintain their flux stability (Bolton et al. 2006). The UF membranes can be used in wastewater treatment, constructed in a way to be integrated in a membrane bioreactor (MBR). The MBR is a novel approach in producing effluent with superb quality. But one of the main problems of this technology is that UF membrane is prone to severe flux decline (Boerlage et al. 2002; Jin et al. 2015). The main reason for the membrane flux decline canbe connected to accumulation of organic material on the membrane surface or the effects of inorganic constituents and concentration polarization of the membrane surface (Brauns et al. 2002). In addition, due to fouling impact and maintaining constant flux of the permeate, the operational pressure is increasing, which further increases the energy consumption as well as operating cost with need for higher membrane area (Gao et al. 2011).

There are two main membrane pressure driven filtration technics. The first one is that produce permeate and retentate (rejected part) and is known as crossflow filtration, and the second one that only produce permeate without retentate and is known as dead-end filtration (Lee et al. 2003).In dead-end filtration, because of the retention of feed water constituents during ultrafiltration at constant transmembrane pressure, the flux changes in time (Poele and Van der Graaf, 2002; Li et al. 2020). For the design of the membrane processes it`s important to know how the flux will develop for a given time, especially in the design of MBR, where the effect of the solid has a heavy toll on the membrane permeability and fouling (Wuet al. 2006). But before determination of the solids effect, the effect of the water constituents on the membrane permeability and fouling should be known.

In terms of membrane material, membranes are classified as either organic or inorganic. Organic membranes are made from organic polymers. These include polyethylene (PE), polyethylene sulphate (PES), polytetrafluorethylene (PTFE), polypropylene (PP) and cellulose acetate among others (Aliyuet al. 2018). All of them have different permeability and hydrophilic properties. Inorganic membranes are made from ceramics, metals, zeolites, or silica carbide. They are chemically and thermally stable and used widely in industrial applications in the processes of ultrafiltration and microfiltration (MF) (Mallada and Menéndez,2008; Parket al. 2020).

Today, the membranes separation processes as microfiltration, ultrafiltration, nano filtration and reverse osmosis, are generally made from synthetic organic polymers (Cetinkaya and Bilgili, 2019). The wastewater treatment of membrane bioreactor is a combination of biological processes with activated sludge and membrane separation processes with UF or MF. The MBR are used for wastewater treatment purposes or for resource recovery from wastewater (Singh and Hankins, 2016; Judd, 2010). Over the past couple of decades, MBRs have emerged as efficient wastewater treatment technology as they fill in the gaps left by conventional activated sludge processes such as their inability to cope with fluctuations in effluent flow rates and composition as well as their failure to meet higher effluent discharge limits for reuse purposes or the tighter water legislatives. MBRs having much smaller footprint, also save much space compared to conventional treatment systems (Judd, 2016). Currently there are two configurations that are used, the side stream MBR and immersed MBR (Wanget al. 2020). The side stream MBR was the first to be developed. With the side stream MBR, the membranes or filtration element are installed outside the bioreactor, needing an intermediate pumping system which transfers the biomass to the filtration module and the concentrate from the filtration set up back to the bioreactor (Loet al. 2015). This set up is advantageous, in that the membrane module is easily accessible for cleaning, however, due to the high energy and pressure requirements, the side stream MBR have had limited application (Yanget al. 2006).

The aim of the paper is to investigate the membrane filtration permeability characteristics with different water solutions. It is experiment made to determine the effect of the water inorganic constituents and turbulence to the filtration characteristics of the membrane that is used for separation of the active sludge in bioreactor. There are studies for the membrane permeability when is used for separating the active sludge in the membrane bioreactor, but there a few experimental set-up regarding the composition of the inorganic constituents and filtration characteristics of the membrane where the active sludge effects on the membrane is neglected. In this work we used the water composition and turbidity, without the presence of the active sludge, to determine the permeability characteristics of a flat sheet membrane that is used as submerged membrane module in membrane bioreactor for wastewater treatment.

MATERIALS AND METHODS

The polyethylene sulphate membranes (PES) that were used were obtained from Microdyne-Nadir. The average pore size of the PES membrane was 0.04 μm (150 KDa). The PES membrane is used in the removal of macromolecules or concentration of large organic solutes in process applications and can be used for membrane bioreactor (MBR). NADIR PES flat sheet membrane is available in an A4 sample size of 210 mm x 297 mm and in a nominal roll size of 150 m x 1016 mm. As supporting layer for the PES membrane, polypropylene backing material is used. The membrane material was formed to have net membrane area of 0.009 m2. The module was constructed using polyvinyl chloride (PVC) parts and fittings that can withstand 8 bar pressure and was designed to fit the membrane of 9.07 cm^2. Using a build in membrane surface mixer, turbulence could be created on the membrane surface. It was connected to high pressure dosing pump and pressure gauge with pressure transmitter. The PLC was connected and the pressure in the system was maintained at 1bar. On the side a manual pressure gauge was installed. The valves were installed for controlling the process and if needed, for a pressure correction. The material used for the module and the system was tested at 5 bar pressure. The module was design for dead -end filtration. For the control of the process LOGO SOFT PLC was used. The constructed membrane module is shown in Figure 1.

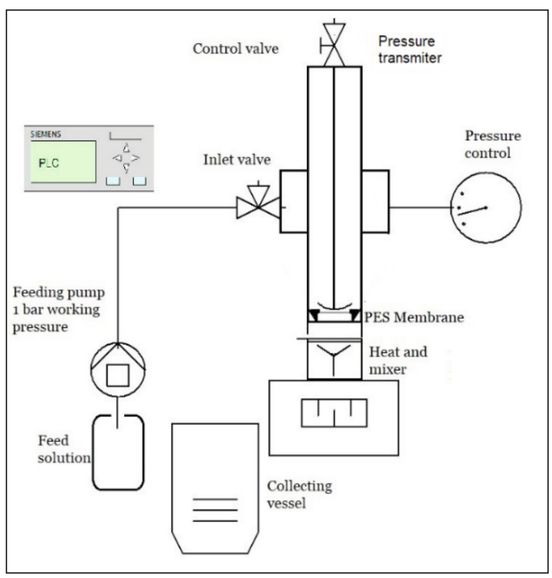

Figure 1. Schematics of the laboratory set up for dead-end membrane filtration

The membranes were shaped in circles with same dimensions. For the filtration, using different aqueous solutions, one membrane at a time was placed in the membrane module. The mechanical magnetic stirrer above the membrane surface was be used to promote the turbulence. The mixing speed was controlled with the magnetic stirrer below the module. For every new experiment, the membrane was replaced. The used membranes were not chemically cleaned or used again, instead, a new membrane was applied. The prepared aqueous solutions were placed in 5 L feed container. The container could be easily replaced every time when a new solution was used. The dosing pump feeds the solution into the module and maintains operational pressure of 1 bar. The permeate from the membrane filtration, was collected in a container that could measure the flow as mg/min or ml/min. The membranes that shaped from NADIR and used are shown in Figure 2.

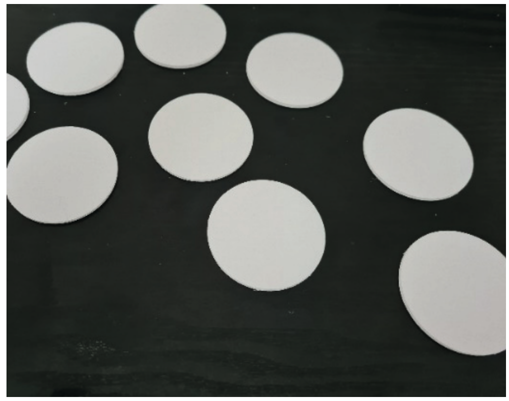

Figure 2. Shaped and prepared PES membrane for dead end filtration

The aqueous solutions used for this experiment were prepared with permeate water from reverse osmosis (R.O.) that has conductivity (EC) of 20 µS and pH=6.7. In this investigation, tap water with the characteristics shown in Table 1, obtained from Skopje city public water system and treated effluent from wastewater treatment plant at the airport in Skopje was also used. The analysis was made in the laboratory of the center for sanitary control, public enterprise for water supply and sewage of Skopje.

Table 1. Characteristics of tap water from Skopje

Parameter:	Value
pH	7.3
EC [µS]	540
COD_{KMnO4}	1.30
NO_3[mg/l]	6.50
SO_4^{2-}[mg/l]	6.80
Na^+ [mg/l]	7.10
Cl^- [mg/l]	7.10
HCO_3^-[mg/l]	434

The effluent that was obtained from Sequencing batch reactor (SBR), treating wastewater from airport terminal in Skopje, had the properties shown in Table 2. The analysis was made in the laboratory of the center for sanitary control, public enterprise for water supply and sewage of Skopje.

Table 2. Characteristics of effluent SBR from airport terminal in Skopje

Parameter:	Value
pH	7.3
BOD_5[mg/l]	12
COD $_{Cr207}$[mg/l]	42
NO_3^-[mg/l]	8.25
SO_4^{2-}[mg/l]	23.8
TSS [mg/l]	3.6

The aqueous solutions were prepared using reverse osmosis water with different concentrations of NaCl. The analysis of the water was conducted with LUTRON YK-2005 WA and Spectroquant Prove UV/VIS spectrophotometer. The reverse osmosis produced water was used for preparing 10% NaCl solution that would be used in the membrane filtration. The prepared solution was placed in a new container and feed to the system. During the experiment the membrane was replaced with a new one and the membrane module was cleaned.

In dead-end filtration, the relation between the permeate flux gravimetrically measured at different time intervals and TMP is defined by the fallowing Equation 1, that is the modified Darcy`s equation for describing the role of different fouling resistances that cause flux decline on the membrane (Rushton et al. 1995; Sousa et al. 2020).

$$J_p = \frac{1}{A_m \rho} \frac{d\, m_p}{dt} = \frac{\Delta P}{\mu\,(Rt+Rf)} \quad (1)$$

where J_p is the permeate flux (l/m^2h), A_m is the effective membrane area (m^2), m_p is the total mass of permeate (kg), ρ is the volumetric mass density (kg/m^3), t is the filtration time (s), ΔP is transmembrane pressure drop (Pa), μ is the filtrate viscosity (Pa·s), Rm is the intrinsic membrane resistance (1/m) and Rf is the fouling resistance (1/m).

The membrane flux can be calculated using Equation 1 and the fallowing derived Equation 2 (Judd, 2010).

$$J = \frac{Q_{permeate}}{A_{membrane}} \quad (2)$$

where J (l/m^2 h) is the membrane flux and Q is the amount of permeate produced from the installed active membrane area A of 1 m^2 in one hour. The standardized membrane permeability (l/m^2h bar) at 20˚C was calculated using the Equation 3 (Judd, 2010).

$$J_{20} = \frac{J \cdot 1.024^{(20-T)}}{\Delta P} \quad (3)$$

where $J_{20(l/\ (m}{}^{2}$h bar)) is the normalized permeability of the membrane at 20˚C and (bar) is the transmembrane pressure.

RESULTS AND DISCUSSION

The different types of aqueous solutions were prepared and set for continuously feeding to the membrane module. The membrane flux and the TMP were measured. Then the normalized membrane permeability at 20 ˚C was calculated. During the filtration period back flush of the membranes was not applied.

The membrane permeability with tap water is shown in Figure 3. In the first hour there is a permeability decrease, that is a result of the water constituent, concentration polarization and membrane resistance to the filtration process. The filtration was carried out without promoting turbulence on the membrane surface. The initial permeability with tap water was calculated as 277 l/ m^2 h bar. In the research made by Li et.al (2019), using the filtration cell Amicon 8400 and ultrapure water, the initial flux for 150 KDa PES membrane was calculated in the range of 410 l/ m^2h bar in. According to the producer of the membrane, the initial clean water flux of the membrane is 285 l/ m^2h (Microdyn-Nadir, Germany).

In the test results obtained by the producer using stirred cell 700 RPM with 0.7 bar pressure at 20°C the membrane clean water permeability is characterized as more than 200 l/ m^2h bar (Debien et al. 2013).

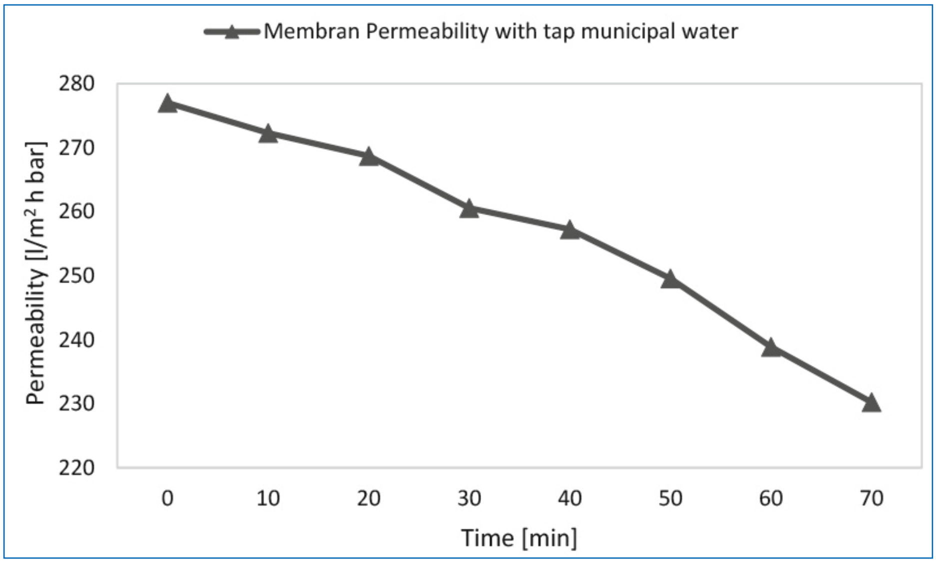

Figure 3. Permeability characteristics of PES membrane with tap municipal water

Then, the aqueous solutions were prepared using permeate water from reverse osmosis with conductivity EC=20 μS and pH=6.7. The water from reverse osmosis was placed in the container and feed to the designed module. The surface mixing was not activated and no turbulence on the membrane surface was promoted. The parameters were monitored for 2.5 hours. Then the membrane was replaced with a new one and the solution again was feed to the module. This time, during the dead-end filtration, the mixer in the module was activated and turbulence was promoted.

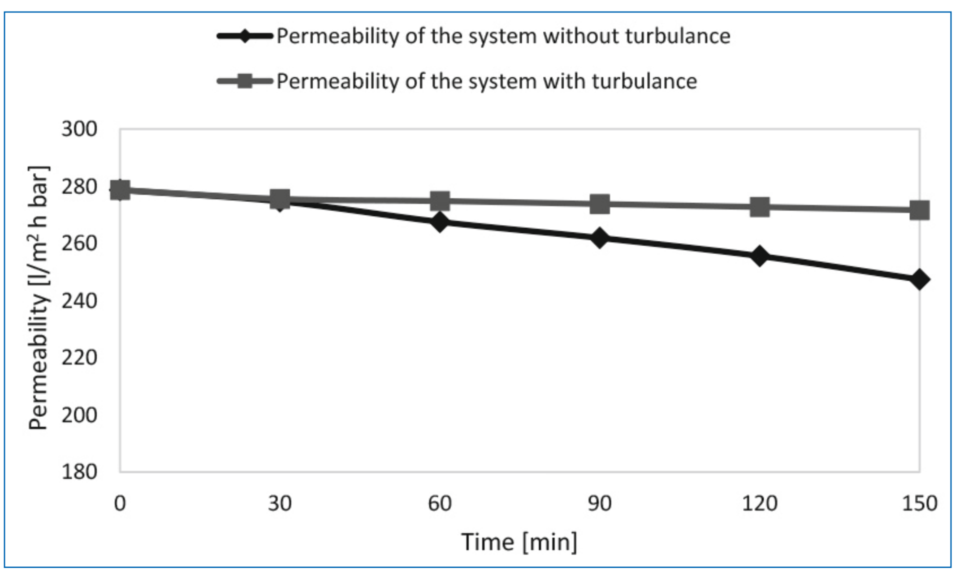

Figure 4. Permeability characteristics of PES membrane with R.O. water and surface mixing

The results are shown in Figure 4 and can be seen that the permeability characteristics of the membrane are better when turbulence is promoted. There is a permeability decrease with both aqueous solution but with adding turbulence to the membrane surface, the permeability reduction is steadier. This is due to the effect of reducing the surface concentration polarization and the membrane fouling effect, making the membrane flux more sustained. Figure 5 shows the permeability characteristics of the PES membrane with 10% NaCl solution, where there is no mixing on the membrane surface to promote the turbulence. Adding NaCl to the system increases the concentration polarization of the membrane and reduces the membrane permeability characteristics withing minutes during the dead-end filtration.

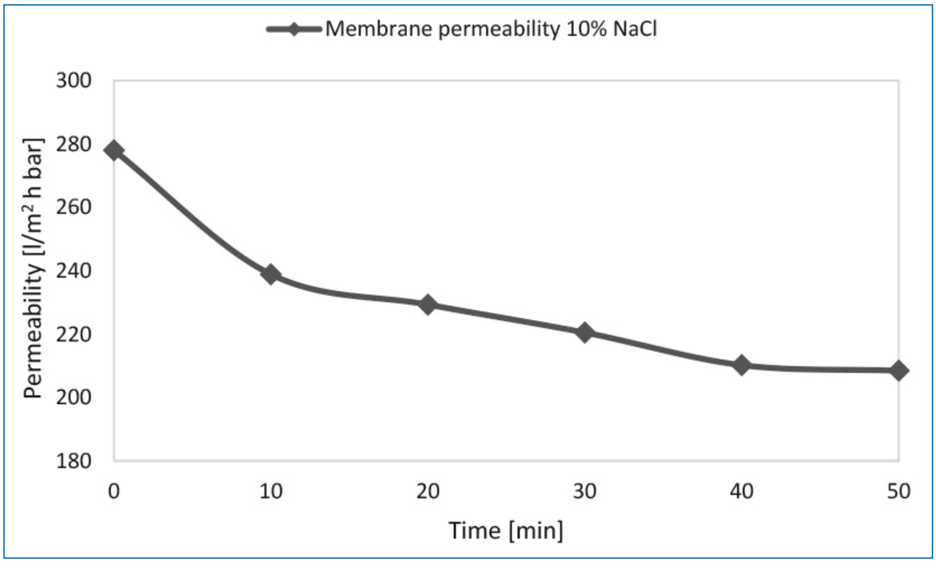

Figure 5. Permeability characteristics of PES membrane with 10% NaCl without mixing

Then the membrane was replaced with new one and the filtration continued with activating the mixing unit and promoting turbulence on the membrane surface. The results of the membrane permeability are shown in Figure 6. In Figure 6 are shown the permeability characteristics of PES membrane with 10% NaCl and promoting surface turbulence. In the beginning of the experiment, when using 10% NaCl aqueous solutions as feed to the module as shown in Figure 5 and Figure 6, the promotion of turbulence did not affect the permeability of the membranes. But after some time, as the dead-end filtration was continuing, the difference in permeability characteristic were clearly visible.

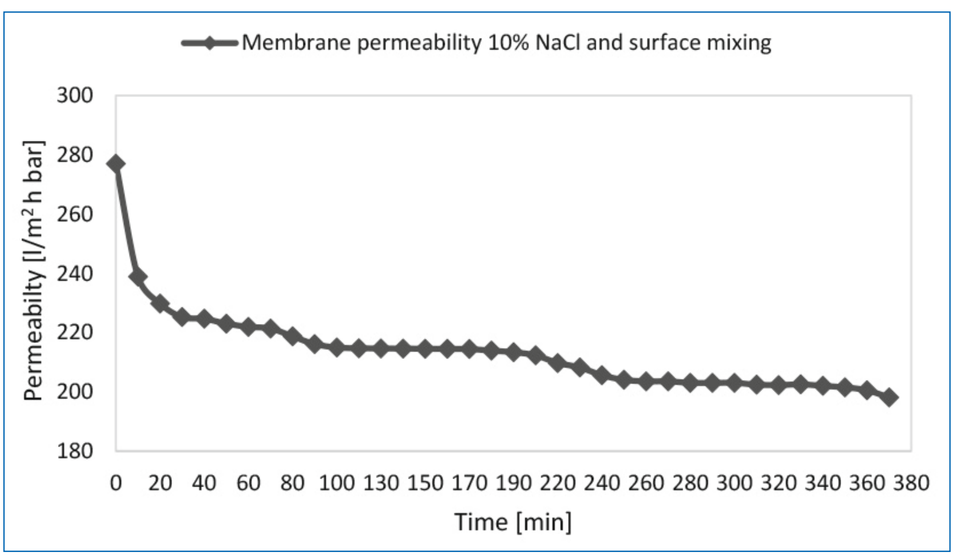

Figure 6. Permeability characteristics of PES with 10% NaCl with mixing

The permeability of the membrane is clearly affected by the water compostion and the type of filtration.Adding turbulance to the filtration proces gives better permeability caracteristcs of the membrane.

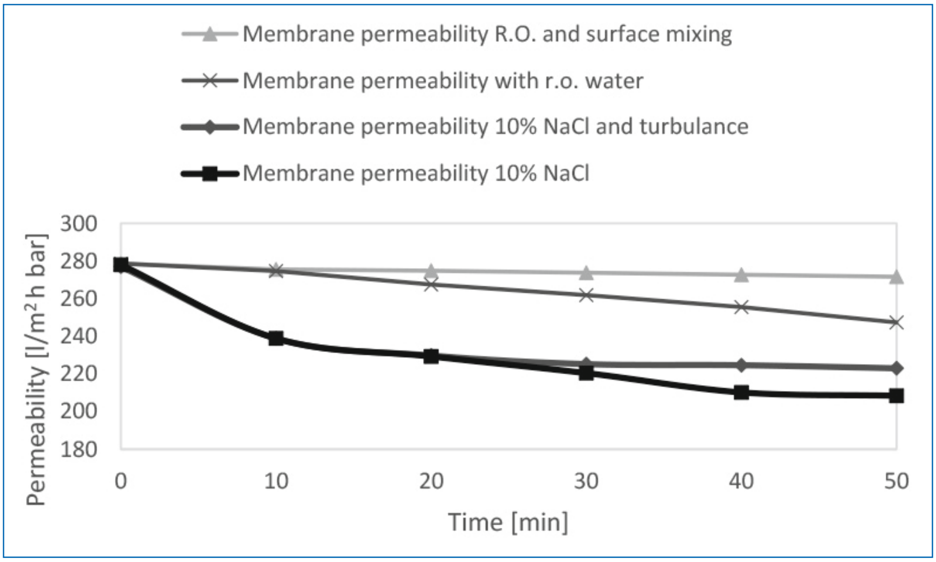

Figure 7. Permeability characteristics of PES membrane with different feed water characteristics and mixing

The differnces in the membrane permeability when using aqueus solutions with different characteristics and different filtration strategies with or without adding turbulance on the membrane surface is shown in Figure 7. At the end of the experiment a new membrane was used for dead-end filtration with 20% NaCl solution. The results from this filtration and membrane permeability are shown in Figure 8. During the filtration membrane surface turbulence was not promoted.

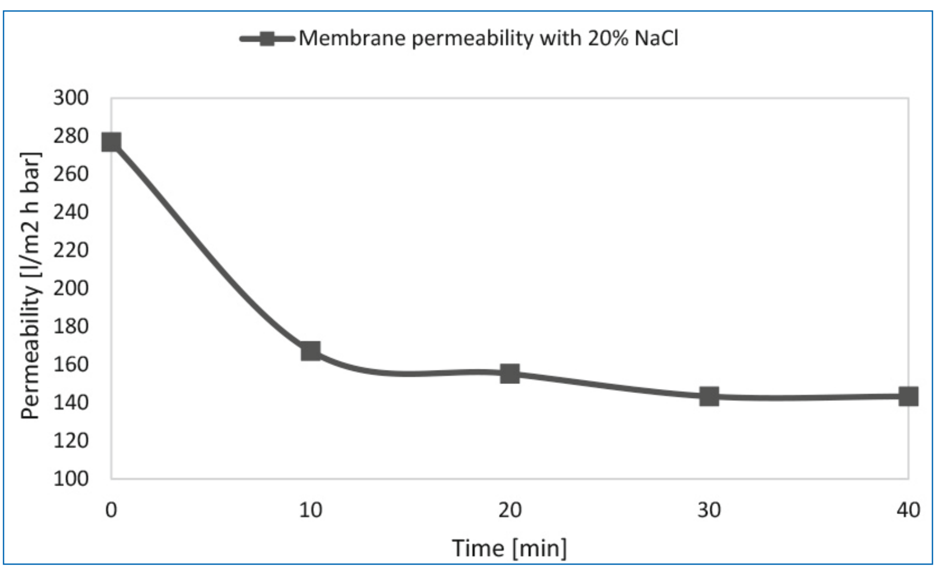

Figure 8. Permeability characteristics of PES membrane with 20% NaCl solution without mixing

When comparing the membrane permeability characteristics where there is no turbulence promoted and dead-end filtration is used, we can see as the concentration of the constituents is higher the permeability starts to drop faster. In Table 3 are presented the permeability characteristics of the membrane with different aqueous solutions, where membrane surface turbulence is not promoted. The higher the concentration of NaCl, the faster the permeability decline is observed. The inorganic constituent in the water clearly affects the permeability decline of the membrane. With time their concentration increases, consequently a boundary of higher surface concentration is created. On the other hand, the mixing and creating surface turbulence improves the permeability characteristics of the membrane.

Table 3. Membrane permeability within time using different aqueous solutions.

Time [min]	Membrane permeability with sanitary water	Membrane permeability with 10% NaCl	Membrane permeability with 20% NaCl
0	277	278	277
10	272.2682	269.253	167.1975
20	268.7102	238.8535	155.2548
30	260.5675	229.2994	143.3121
40	257.2268	229.2994	13.3121

In general, differences in membrane composition are related to differences in filtration characteristics (Boerlage et al. 2003;). In the research made by Roorda (2004) the tested membrane permeability with pre-treated wastewater treatment plant effluent was in range of 170-250 l/m^2 h bar and the maximum flux for stable performance showed large differences. As the applied membrane systems were comparable, the major determining factor for the membrane fouling problems should be related to the feedwater composition (Roorda, 2004). In the last experiment, the effluent from an SBR that is treating wastewater from an airport treatment plant was collected.

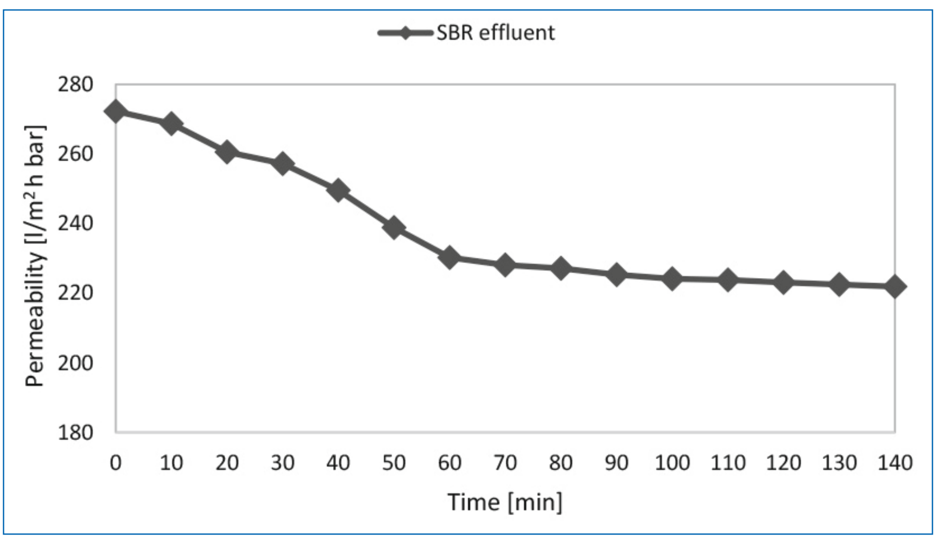

Figure 9. Dead-end filtration using SBR effluent

The SBR is designed for operating in 4 cycles per day with effluent sand filtration unit. It is treating 75 m^3/day wastewater originating from the Airport passenger terminal in Skopje. The collected effluent was used as a feed solution to the dead-end membrane filtration module, shown in Figure 9.

The decline in permeability is alike to the membrane filtration with tap water characteristics, as the concentration polarization is having a similar effect. The difference in the membrane permeability originates from the concentration of organic constituents in the SBR effluent. These results indicate that the components retained on the membrane surface predominantly determine the filtration characteristics.

CONCLUSION

The membrane reactors are state of the art systems. They are used for water and wastewater treatment. The membrane filtration always leads to increase membrane resistance to the permeate flow. The transmembrane pressure (TMP) is one of the key parameters when operating with membrane filtration and shows the tendency of membrane fouling. To keep the permeate flow constant, the applied membrane pressure should be increased, thus resulting in higher energy consumption. Another option is to install higher membrane area which would increase the investment costs.

The stability of the process is important for any MBR. The membrane permeability is important factor when designing membrane reactors, especially membrane bioreactors (MBR). In this investigation, the result showed us that the membrane flux can be seriously affected by the aqueous solutions constituents. The concentration polarization on the membrane surface can have a negative impact on the membrane flux. In the design approach of the MBR, detailed characterization of the solution must be made. In the feed water, the organic and inorganic foulants can be pre-determined. Only then we can minimize the effect of the concentration polarization and the membrane fouling in the MBR, and with the right membrane area applied, we can have a stable permeate flux and lower energy consumptions. The findings of this study would provide theoretical supports for the control and design of the MBR in treatment of urban wastewater. The results show as that this type of membrane with adding turbulence can be used in construction and design of MBR whit ultrafiltration properties where the water composition plays an important role in membrane permeability reduction.

REFERENCES

Abdessemed, D., Nezzal, G. & Ben Aim, R. (2000). Coagulation-adsorption-ultrafiltration for wastewater treatment and reuse. Desalination, 131 (1), 307-314.

Aliyu, U.M., Rathilal, S. & Isa, Y.M. (2018). Membrane desalination technologies in water treatment: A review. Water Practice and Technology, 13, 738–752.

Boerlage, S.F.E., Kennedy, M.D., Dickson, M.R., El-Hodali, D.E.Y. & Schippers, J.C. (2002). The modified fouling index using ultrafiltration membranes (MFI-UF): characterisation, filtration mechanisms and proposed reference membrane. Journal of Membrane Science, 197, 1- 21.

Boerlage, S.F.E., Kennedy, M.D., Aniye, M.P., Abogrean, E., Tarawneh, Z.S. & Schippers, J.C. (2003). The MFI-UF as a water quality test and monitor. Journal of Membrane Science, 211, 271-289.

Bolton, G., LaCasse, D. & Kuriyel, R. (2006). Combined models of membrane fouling: Development and application to microfiltration and ultrafiltration of biological fluids. Journal of Membrane Science, 277, 75–84.

Brauns, E., Faes, K., Van Hoof, E., Doyen, W., Dotremont, C.& Leysen, R. (2002). The measurement and presentation of the fouling potential method with a new method. In Proceedings of the 5th Conference “Membranes in Drinking and Industrial Water Production”,37, 381-388. Mulheim/Ruhr, Germany.

Cetinkaya, A.Y. & Bilgili, L. (2019). Life cycle comparison of membrane capacitive deionization and reverse osmosis membrane for textile wastewater treatment. Water, Air & Soil Pollution, 230, 149.

Debien, I.C.D.N., Gomes, M.T.D.S., Ongaratto, R.S. & Viotto,L.A. (2013). Ultrafiltration performance of PVDF, PES, and cellulose membranes for the treatment of coconut water (Cocos Nucifera L.), Food science and technology, 33 (4)

Fane, A.G., Tang, C.Y. & Wang, R. (2010). Membrane technology for water: Microfiltration, ultrafiltration, nanofiltration, and reverse osmosis. Treatise on Water Science, 4, 301–335.

Gao, W., Liang, H., Ma, J., Han, M., Chen, Z.L., Han, Z.S. & Li, G.B. (2011). Membrane fouling control in ultrafiltration technology for drinking water production: A review. Desalination, 272, 1–8.

Galinha, C.F., Sanches, S. & Crespo, J.G. (2018). Chapter 6-Membrane bioreactors. In Fundamental Modelling of Membrane Systems; Luis, P., Ed.; Elsevier: Amsterdam, The Netherlands.

Jin, Y.X., Ju, Y.G., Lee, H. & Hong, S. (2015). Fouling potential evaluation by cake fouling index: Theoretical development, measurements, and its implications for fouling mechanisms. Journal of Membrane Science, 490, 57–64.

Lee, S.H., Fane, A.G., Amal, R. & Waite, T.D. (2003). The effect of floc size and structure on specific cake resistance and compressibility in dead-end microfiltration, Separation Science and Technology, 38, 869 - 887

Li, R., Gao, B., Wang, W., Yue, Q. & Wang, Y. (2020). Floc properties and membrane fouling in coagulation/ultrafiltration process for the treatment of Xiaoqing River: The role of polymeric aluminum-polymer dual-coagulants. Chemosphere, 243.

Li, K., Li, S., Huang, T., Dong, C., Li, J., Zhao, B. & Zhang.S. (2019). Chemical cleaning of ultrafiltration membrane fouled by humic substances: comparison between hydrogen peroxide and sodium hypochlorite, International journal of environmental research and public health, MDPI

Lo, C.H., McAdam, E. & Judd, S. (2015). The cost of a small membrane bioreactor. Water Science & Technology, 72, 1739–1746.

Mallada, R. & Menéndez, M. (2008). Inorganic Membranes: Synthesis, Characterization and Applications; Elsevier: Amsterdam, The Netherlands.

Park, W., Jeong, S.H., Im, S.J. & Jang, A. (2020). High turbidity water treatment by ceramic microfiltration membrane: Fouling identification and process optimization. Environmental Technology & Innovation, 17, 1–10.

Poele, S. T. & van der Graaf, J.H.J.M. (2002). Physical and chemical conditioning of effluent for decreasing membrane fouling during ultrafiltration In Proceedings of the 5th Conference “Membranes in Drinking and Industrial Water Production”, 37, 765-773. Mulheim/Ruhr, Germany.

Roorda, J.H. (2004). Filtration characteristics in dead-end ultrafiltration of wwtp-effluent, J.H.Roorda, printed in Delft, Netherlands.

Rushton, A., Ward, A.S. & Holdich, R.G. (1995). Introduction to Solid-liquid Filtration and Separation Technology; Wiley-VCH Verlag GmbH: Weinheim, Germany; printer in New York, NY, USA.

Judd, S. (2010). The MBR book: Principles and Application of Membrane Bioreactor in Water and Wastewater Treatment, 2nd ed.; Elsevier.

Judd, S. (2016). The status of industrial and municipal effluent treatment with membrane bioreactor technology. Chemical Engineering Journal, 305, 37–45.

Singh, R. & Hankins, N. (2016). Emerging Membrane Technology for Sustainable Water Treatment; Elsevier.

Sousa, M., Lora-García, J., López-Pérez, M. & Heran, M. (2020). Polyethersulfone membranes used to remove colloids and dissolved matter from paper mill treated effluent, Water 2020 12 (2),365;MDPI.

Wu, J.L., Chen, F.T., Huang, X., Geng, W.Y. & Wen, X.H. (2006). Using inorganic coagulants to control membrane fouling in a submerged membrane bioreactor. Desalination, 197 (1–3),124–136.

Wang, S., Chew, J.W. & Liu, Y. (2020). Development of an integrated aerobic granular sludge MBR and reverse osmosis processs for municipal wastewater reclamation, Science of the total environment,748

Yang, W., Cicek, N. & Ilg, J. (2006). State-of-the-art of membrane bioreactors: Worldwide research and commercial applications in North America. Journal of Membrane Science, 270, 201–211.

Recived: May 17, 2021
Accepted: June 16, 2021