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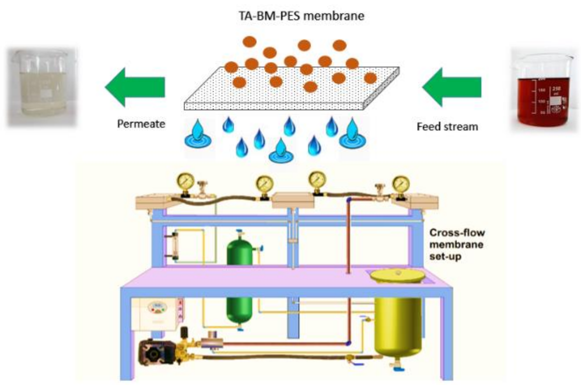
Influence of process and operating variables on the performance and fouling behavior of modified nanofiltration membranes treating licorice aqueous solution

Fariba Oulad[1], Sirus Zinadini*[,1], Ali Akbar Zinatizadeh[1,2], Ali Ashraf Derakhshan[1]

[1]Environmental Research Center, Department of Applied Chemistry, Razi University, Kermanshah, Iran.

[2]Department of Environmental Sciences, School of Agriculture and Environmental Sciences, University of South Africa, Florida, South Africa.

GRAPHICAL ABSTRACT



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ABSTRACT

The main purpose of this study was to investigate the effect of different operational parameters on performance and fouling trends of unfilled-polyethersulfone, 0.5 wt.% boehmite-polyethersulfone, tannic acid coated boehmite-polyethersulfone nanofiltration membranes during filtration of Licorice aqueous solution as model foulant. The impact of hydrodynamic conditions (such as transmembrane pressure and cross-flow velocity) and feed composition on permeation, fouling trends and rejection capability were evaluated using lab-scale cross-flow filtration set-up. The applied transmembrane pressure and cross-flow velocity were various in range of 6-12 bar and 0.5-2.5 cm/s, respectively. The results indicated that although, increasing of operational pressure and cross-flow velocity can enhance the permeability and rejection capability of NF membranes also incur appearance of the more severe fouling phenomenon. The least fouling for NF membranes was occurred at the lowest licorice concentration of 0.1 g/l. The rejection percentage of unfilled and embedded nanofiltration polyethersulfone membranes was more than 92 %.

1. Introduction

In the coming decade, the problem of clean water shortage is on the progress worldwide, owing to extended droughts, speedy growth in population and extension of industrial and agricultural activities (Fang et al. 2017; Kim et al. 2016). Therefore, many attempts have been examined to remove water pollution using various approaches. Biological treatment systems (Amin et al. 2013; Asadi et al. 2012; Pirsaheb et al. 2015; Pirsaheb et al. 2009; Zinatizadeh 2006), photocatalysis (Ghasemi et al. 2016a; Ghasemi et al. 2016b), chemical coagulation (Abbasi et al. 2020; Birjandi et al. 2013; Zinatizadeh et al. 2017), adsorption (Sharafi et al. 2015) and membrane filtration (Rahimi et al. 2016; Zinadini et al. 2017) are technologies which have been employed to treat different wastewaters. Moreover, natural treatment systems such as constructed wetland, pond, and anaerobic/aerobic lagoons have been used for wastewater treatment. But, use of the natural treatment systems is restricted because of considerable dead zone in some parts of the units, resulting in a low reactor volume yield relative to modern processes (Mansouri et al. 2012; Bonakdari and Zinatizadeh 2011). Hence, new advanced technologies are highly needed to provide a supply of safe water in an environmental-friendly approach (Hu et al. 2013). Among all the efficient strategies for production of fresh water, membrane separation technologies using polymeric

*Corresponding author E-mail: sirus.zeinaddini@gmail.com

membranes are regarded as one of the promising approaches and has been attracted interest recent years (Kim et al. 2015). The membrane filtration has practical advantages such as, easy scale-up, high efficiency, low operating temperature, low energy consumption, being eco-friendly, safe and cost-effective, (Wang et al. 2011; Zhou et al. 2016). However, irreversible precipitation of foulants on the polymeric membrane surface leads to enhancement of the resistance against water permeation and progress of membrane fouling, that degenerating the comprehensive membrane performance (Hadidi et al. 2014). Consequently, the treatment of polymeric membranes before practical application had to use to increase the filtration performance and antifouling capability (Yin et al. 2015). Various surface modification strategies were applied for improvement of membrane antifouling performance, that containing blending with hydrophilic nanoparticles (Li et al. 2014; Ma et al. 2016), surface coating (Ma et al. 2015; Moghimifar et al. 2014), radical polymerization (Hou et al. 2015; Zhang et al. 2010) and surface grafting (Hou et al. 2017; Peeva et al. 2012).

Boehmite (aluminium oxide hydroxide) nanoparticle with orthorhombic structure is extensively applied as a nanofiller (Vatanpour et al. 2012), a coating (Das et al. 2009; Mishra et al. 2000), a catalyst, an absorbent and etc. Because of the boehmite nanoparticles possess several benefits such as being cost-effective, resistant against oxidation/reduction reactions, highly abrasive and non-toxic. Vatanpour et al. (2012) reported preparation of antifouling mixed matrix nanofiltration membrane using incorporation of boehmite as a nanofiller. The embedded membranes depicted the improvement of permeability, antifouling capability and surface wettability, owing to existence of numerous hydroxyl groups on the membranes surface. The in situ embedment of hydrated and anhydrous aluminium oxide particles on the surface of PVDF membrane was studied by Wang et al. (2011). The results exhibited the enhancement of membrane hydrophilicity and diminution of membrane bio-fouling problem.

Tannins as cheap and available polyphenols extracted easily from plant tissues such as gallnuts, leaves, pods and seeds of certain plants and as well as, bark fruits (Bacelo et al. 2016; Wang et al. 2011). Tannic acid (TA) contained several advantages such as terrific chelating affinity towards macromolecules and metal ions, antioxidant, antimutagenic and anticarcinogenic properties, inexpensive and eco-friendly. Because of aforementioned properties, tannins have been used in resin production, food additives, water purification, pharmacy and absorbent of cationic dye (Luo et al. 2017; Ye et al. 2016). The iron-tannin-framework (ITF) complex was used by Fang et al. (2017) to increase fouling resistance capability and filtration performance of ultrafiltration polyethersulfone (PES) memb rane with utilization of non-solvent-induced phase separation method. Utilization of ITF leading to enhancement of membrane wettability and porosity as well as mitigation of pore size. The ITF/PES membrane indicated high water permeation flux and excellent antifouling property. The new nanocomposite membrane was prepared by Zhang et al. (2013) through interfacial polymerization using natural material trimesoyl chloride (TMC) and tannic acid (TA). The modified nanocomposite membrane was exhibited high flux recovery ratios and slight flux reduction ratios in the antifouling assessment experiments.

The objective of the present work was evaluation of the operational parameters effect on the performance of 0.5 wt. % boehmite (BM) and TA-functionalized-BM (TA-BM) embedded nanofiltration membranes in treating of licorice aqueous solutions. The performance of modified membranes was compared to bare nanofiltration membrane. The influence of variable operational parameters involving feed solution concentration and hydrodynamic effects (transmembrane pressure and cross-flow velocity) on water permeation flux, rejection and flux reduction ratio was evaluated.

2. Materials and methods

2.1. Materials

Polyethersulfone (PES Ultrason E 6020P, MW = 58000 g/mole Tg = 225℃) and N, N-dimethylacetamide (DMAc) were bought from BASF, Germany. Polyvinylpyrrolidone (PVP, MW= 25000 g/mole) was obtained from Mowiol, Germany. Distilled water was utilized in this work. Pure aluminum nitrate, sodium hydroxide and tannic acid (TA) were supplied from Merck, that used to nanofiller synthesize (BM, TA-BM). Licorice powder was purchased from Parzhak Co., Iran.

2.2. Fabrication of BM nanofiller

The following procedure was used for preparation of BM nanoparticles. Initially, the solution of 6.40g sodium hydroxide (NaOH) in 50ml deionized water and 20g Al $(NO_3)_3$. 9 H_2O in 30ml deionized water were prepared. Then, the obtained solution of NaOH was added gradually to the solution of Al $(NO_3)_3$. 9 H_2O during 17min on the vigorous stirring. The resulted milky solution was transferred in ultrasonic bath for 3h at room temperature. The obtained precipitates filtered, rinsed and dried at 220 °C (Rajabi et al. 2010). The photograph of prepared BM nanoparticles indicated in Fig.1a.

2.3. Fabrication of TA-functionalized BM (TA-BM) nanoparticles

The below experimental procedure was applied for fabrication of TA-BM nanoparticles. Initially, 1g TA was dispersed in 100ml of distilled water afterwards, 3g BM was added, sonicated for 15min and ultimately, resulted solution refluxed for 16 h. Lastly, the obtained precipitate was centrifuged, washed by distilled water and dried at 70 °C. The image of prepared TA-BM nanoparticles was exhibited in Fig.1b.

2.4. Preparation of 0.5 wt.% BM and TA-BM embedded NF PES membranes

The phase inversion method was utilized for fabrication of nanofiltration membranes. Firstly, 0.5 wt.% nanofiller (BM or TA-BM) was mixed into certain amount of DMAc and sonicated for 15min. Then, PES (20 wt.%) and PVP (1 wt.%) was added for preparation of casting solution. The obtained dope solution was homogenized by continuous stirring for 24 h to achieve homogenous solution. The dope solution was then, cast onto a smooth glass plate with 150 µm thickness by self-made casting knife and instantly dipped into a coagulation bath (distilled water, 15 °C). After immersion of prepared membranes in fresh distilled water for 24 h, the obtained membranes dried between two papers at room temperature.

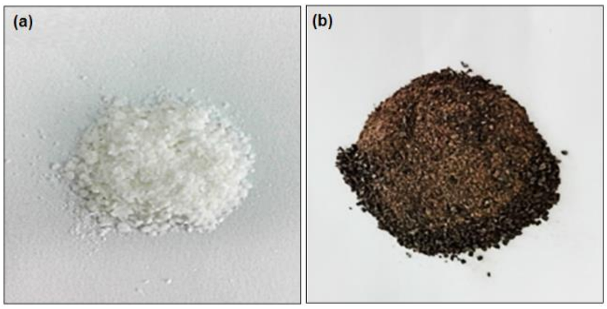

Fig.1. Digital photograph of BM (a) and TA-BM (b) nanoparticles.

2.5. Membrane characterization

The cross-sectional morphology of nanocomposite embedded membranes was characterized using Philips scanning electron microscopy XL30 (SEM, Camscan mv2300, Netherland).

2.6. Nanofiltration performance

The nanofiltration experiments of bare and embedded membranes in terms of water permeation flux, flux reduction ratio and rejection percentage were evaluated by lab-scale crass-flow filtration cell (effective area of 40 cm^2). The water permeation flux is calculated by (Zinadini et al. 2014).

$$J = \frac{M}{A \cdot \Delta t} \quad (1)$$

where, M(kg) is the collected permeates weight. A(m^2) is the effective area of membrane and Δt (h) is the time interval.

The rejection of licorice aqueous solution can be calculated by the following expression (Fang et al. 2017).

$$R\,(\%) = (1 - \frac{C_p}{C_f}) \times 100 \quad (2)$$

where, C_p and C_f are the concentration of licorice aqueous solution in permeate and feed respectively. The flux reduction percentage can be calculated by using the following Eq.

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$$J_{reduction}\,(\%) = \left(\frac{J_{Initial}-J_{Final}}{J_{Initial}}\right) \times 100 \quad (3)$$

where, $J_{Initial}$ and J_{Final} are permeation flux at beginning and ending filtration respectively.

3. Results and discussion

3.1. Membrane morphology

The unfilled-PES, 0.5 wt.% BM-PES and TA-BM-PES NF membranes were fabricated by phase inversion method. The cross-sectional morphology of nanocomposite membranes was exhibited in Fig. 2. As can be observed clearly, all of the prepared membranes demonstrate similar asymmetric morphologies with a dense skin top-layer and porous sub-layer. It is evidently shown that, the sub-layer macro-voids lengths were elongated by addition of 0.5 wt.% nanofiller (BM and TA-BM) in membrane matrix. Elongation of the macro-voids structures in the embedded membranes incur reduction of membrane resistance to permeation and consequently, promotion of water passage from the embedded NF membranes. The finger-like structure of the prepared mixed matrix membranes was not effected by adding BM or TA-BM as nanofiller. The same behavior was also reported by Rahimpour et al. 2008 and Razmjou et al. 2011.

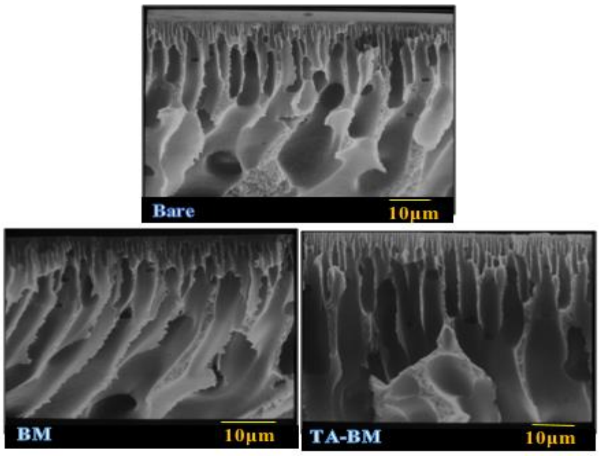

Fig. 2. The SEM images of nanocomposite membranes.

3.2. Influence of operational pressure

The influence of transmembrane pressure on the licorice foulants purification by unfilled, 0.5 wt.% BM and TA-BM nanofiltration PES membranes was analyzed under an operational pressure range of 6-12 bar. Fig. 3 presents flux permeation results of unfilled and embedded NF PES membranes at various operational pressure levels. Enhancement of operational pressure from 6 to 12 bar for unfilled NF PES membrane cause increment of permeated flux from 3.77 to 10.56 kg/m^2. h. For 0.5 wt.% BM/PES embedded membrane by increasing the transmembrane pressure from 6 to 12 bar, permeated flux was improved from 67.123 to 84.3 kg/m^2. h. For 0.5 wt.% TA-BM/PES blended membrane as the transmembrane pressure was elevated from 6 to 12 bar, permeated flux was enhanced from 143.6 to 280.43 kg/m^2. h. As observed above, the embedded nanofiltration membranes possessed higher permeated flux levels than unfilled NF PES membrane. It can be related to presence of numerous hydrophilic groups induced by nanofiller (BM or TA-BM) that improved membrane hydrophilicity and permeability. Licorice aqueous solution with concentration of 0.3 g/L at different pressure levels (6 and 12 bar) was induced to permeate through the membranes for 2h to evaluate the influence of operational pressure on the fouling behavior in term of flux reduction of unfilled and embedded membranes. Fig. 4 exhibited the permeated flux profile versus time of unfilled and embedded nanofiltration membranes. As can be seen clearly from the Fig. 4, 0.5 wt.% TA-BM/PES embedded membrane indicated almost steady permeated flux during licorice foulants filtration in comparison of bare-PES and 0.5 wt.% BM/PES membranes. The obtained results of flux reduction ratios of NF membranes during treatment of licorice aqueous solution were shown in Fig. 5 that confirmed mentioned content above. For unfilled membrane by raising the operational pressure from 6 to 12 bar, the flux reduction ratio was elevated from 11.37 % to 15.18 %. The 0.5 wt. % BM/PES blend membrane exhibited flux reduction ratio of 20.18 % (at 6 bar) and 29.44 % (at 12 bar). Increment of transmembrane pressure from 6 to 12 bar for 0.5 wt. % TA-BM/PES blend membrane leads to enhancement of flux reduction ratio from 4.71 % to 8.4 %. According the results, although enhancing the operational pressure can enhance the permeated flux level, the higher permeated flux level at greater transmembrane pressure (12 bar) can be ascribed to the more fouling trend (i.e., more flux reduction) (Wang et al. 2016). It can be associated with increasing operational pressure which results in the provision of favourable conditions for fouling (Oulad et al. 2018). The same results were obtained by other researcher (She et al. 2013; Zou et al. 2011).

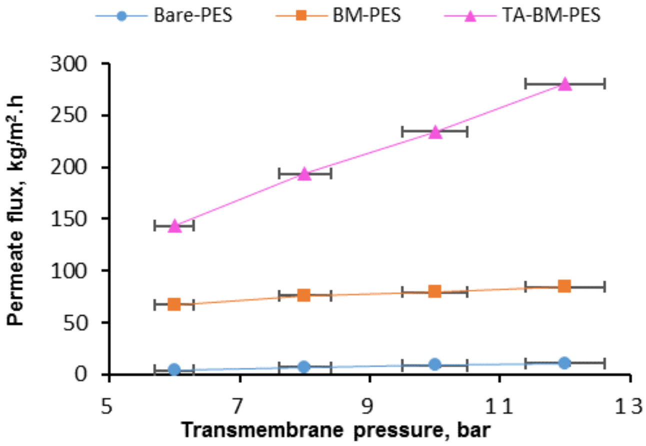

Fig. 3. Influence of operational pressure on permeation flux of NF membranes (C=0.3 g/L, time=30 min, V=1 cm/s, pH = 5.3).

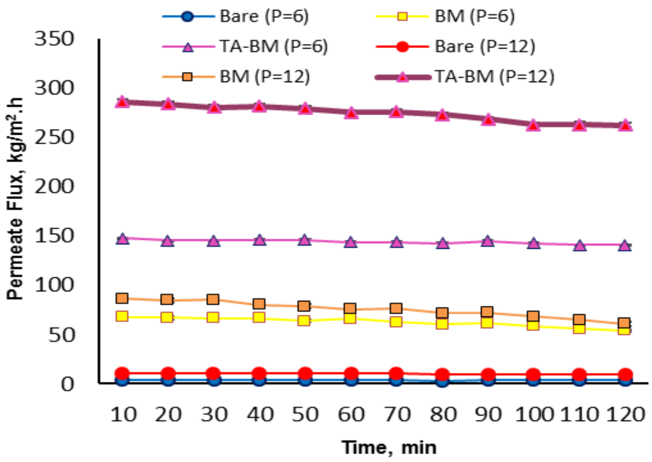

Fig. 4. The permeated flux profile versus filtration time of nanofiltration membranes (C=0.3 g/l, time=120 min, V=1 cm/s).

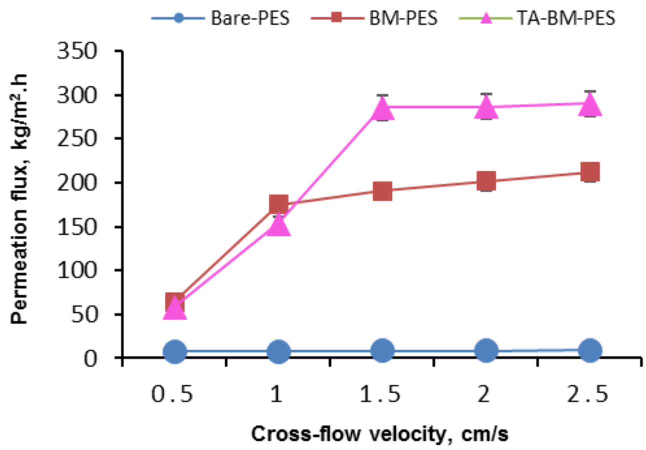

Fig. 5. The results of flux reduction ratios of NF membranes at various operational pressure.

The influence of operational pressure on rejection percentage of unfilled-PES BM-PES and TA-BM-PES membranes was shown in Fig. 6. The rejection percentage under variable operational pressure (6-12 bar) was different between range of 92.2-94.2 % for bare-PES membrane, between range of 94-98.2 % for BM/PES membrane and between range of 96.2-99% for TA-BM/PES membrane. From the rejection results in Fig. 6, it was found that the rejection percentage of nanofiltration membranes was enhanced by increasing the applied transmembrane pressure. It was associated with deposition of licorice foulants on the surface of NF membranes that causes formation of secondary obstacle and prevents the transit of other foulants molecules through the membrane.

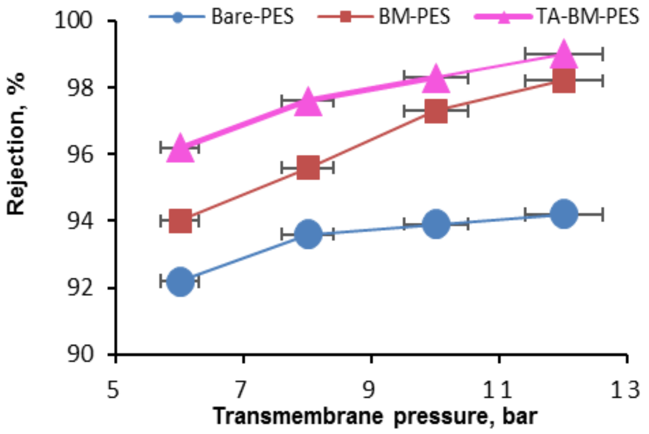

Fig. 6. Influence of operational pressure on rejection of nanofiltration membranes (C=0.3 g/L, time=30 min, V=1 cm/s).

3.3. Influence of cross-flow velocity

The influence of cross-flow velocity on the purification of licorice foulants by bare PES, 0.5 wt.% BM/PES and TA-BM/PES NF membranes was evaluated under a cross-flow velocity range of 0.5-2.5 cm/s. The results of permeated flux of unfilled and embedded membranes at different cross-flow velocity levels was shown in Fig. 7. As can be seen clearly, the permeated flux was improved significantly by increasing the cross-flow velocity. From the Fig. 7, it was found that by enhancing cross-flow velocity from 0.5 to 2.5 cm/s the permeated flux was raised from 7.76 to 9.27 kg/m^2.h for bare-PES NF membrane, from 63.93 to 212.21 kg/m^2.h for 0.5 wt.% BM/PES NF blended membrane and from 58 to 290.2 kg/m^2.h for 0.5 wt.% TA-BM/PES NF embedded membrane. The cross-flow velocity had a major impact on concentration polarization. Production of higher permeated flux at high cross-flow velocity can be related to reduction of concentration polarization and appeared turbulence (Xu et al. 2005). The least flux reduction ratio was observed under the lowest licorice concentration of 0.1 g/l (5.66, 13.0242 and 3.04 % for bare, BM/PES and TA-BM/PES membranes respectively). From the Fig. 12, it was found that by enhancing the licorice foulants concentration from 0.1 to 0.3 g/l, the flux reduction ratios were enhanced from 5.66 to 11.37 % for bare membrane, from 13.02 to 20.18 % for BM/PES membrane and from 3.04 to 4.71% TA-BM/PES membranes. More substantial flux reduction occurred when the licorice foulants concentration in the feed solution elevated from 0.3 to 0.5 g/L. Because of increment of licorice concentration (from 0.3 to 0.5 g/L) might be accelerated the fouling rate (Liu and Mi. 2012). It can be justified by greater amount of foulants that was deposited on the membranes surface at the higher licorice concentration. Fig.13 indicated the photos of unfilled-PES, 0.5 wt.% BM-PES and TA-BM-PES NF membranes after licorice fouling and cleaning process with distilled water. The relatively negligible foulant film was formed on the TA-BM-PES membrane surface compared to bare-PES and BM-PES NF membranes due to the TA-BM-PES membrane had significantly antifouling capability and flux recovery ability. To analyse the influence of cross-flow velocity on the fouling trends of nanofiltration membranes, licorice aqueous solution at various cross-flow velocity range of 0.5-2.5 cm/s was induced to permeate via the membranes for 2h. The permeation flux profile against time of bare and blended membranes depicted in Fig. 8. In order to clarify fouling trends of membranes, the flux reduction ratios were calculated and exhibited in Fig.9. As can be observed, the flux reduction ratio was augmented from 10.26% to 20.62% for bare-PES membrane, from 12.37 % to 24.38% for BM/PES membrane and from 3.43 % to 16.96 % for TA-BM/PES membrane with enhancement of cross-flow velocity from 0.5 to 2.5 cm/s. The results might have recommended that, the enhanced cross-flow velocity caused an increase in the fouling tendency during filtration of foulants. The influence of cross-flow velocity on rejection percentage of unfilled and blended nanofiltration membranes was indicated in Fig. 10. As can be observed, with increment of cross-flow velocity from 0.5 to 2.5 cm/s, the rejection percentage of nanofiltration membranes was improved. The rejection percentage of NF membranes was various between 92%-99 %. Improvement of membrane rejection percentage induced by enhancing cross-flow velocity may be attributed to concentration polarization mitigation. As regards, the lower concentration polarization in the membrane surface vicinity incur lower passage via the membrane or in the other words, higher rejection.

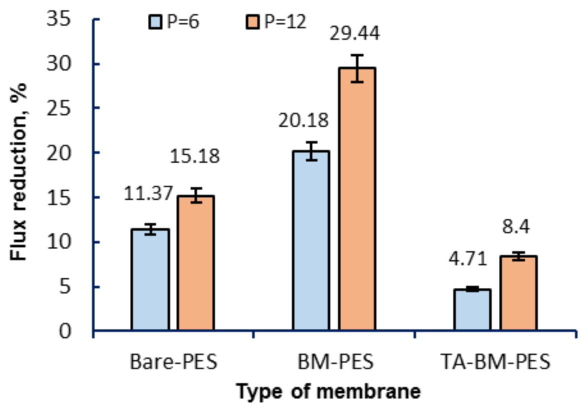

Fig. 7. Influence of cross-flow velocity on permeation flux of nanofiltration membranes (P=6 bar, C=0.3 g/L, time=30 min).

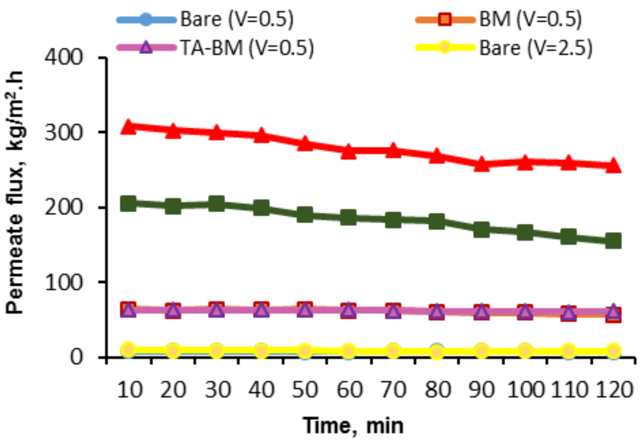

Fig. 8. The permeated flux profile against filtration time of nanofiltration membranes (C=0.3 g/l, time=120 min, P=6 bar).

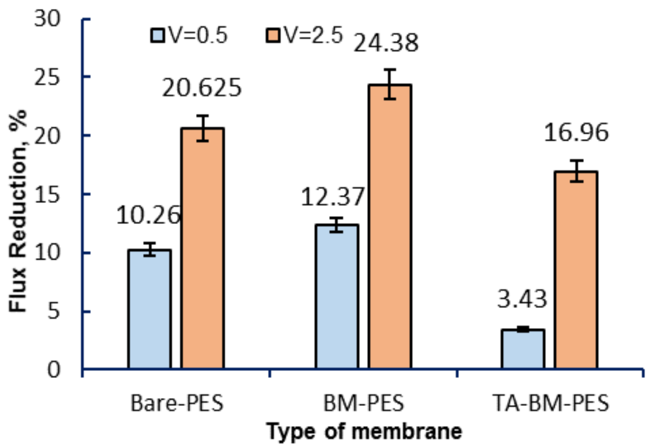

Fig. 9. The results of flux reduction ratios of NF membranes at diverse cross-flow velocity.

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3.4. The influence of feed composition

The influence of foulants concentration on the purification of licorice aqueous solution by unfilled and embedded NF membranes was verified by varying the concentration of licorice foulants. For this purpose, licorice aqueous solution at various concentration values (0.1, 0.3, 0.5 g/l) was induced to permeate through the unfilled and embedded NF membranes for 2h. The permeated flux profile against time of NF membranes at various concentration values was shown in Fig. 11. The results of flux reduction ratios of NF membranes duration of licorice solution filtration was indicated in Fig.12.

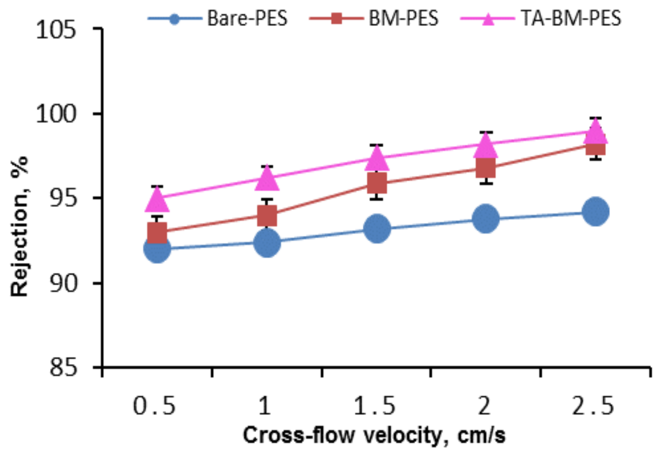

Fig. 10. Influence of cross-flow velocity on rejection of nanofiltration membranes (P=6 bar, C=0.3 g/l, time=30 min, PH = 5.3).

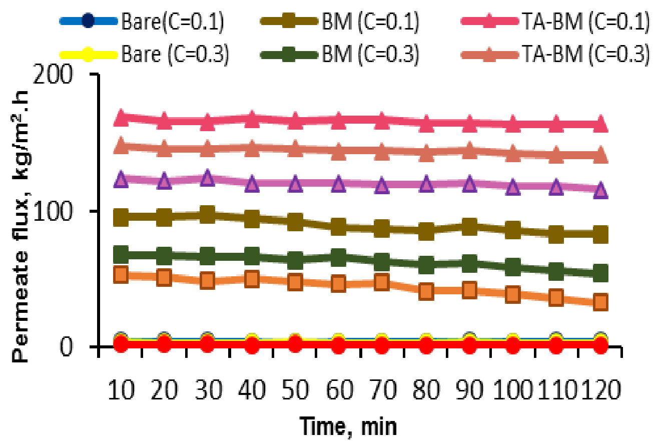

Fig. 11. The permeated flux profile against filtration time of nanofiltration membranes (P=6 bar, time=120 min, V=1 cm/s).

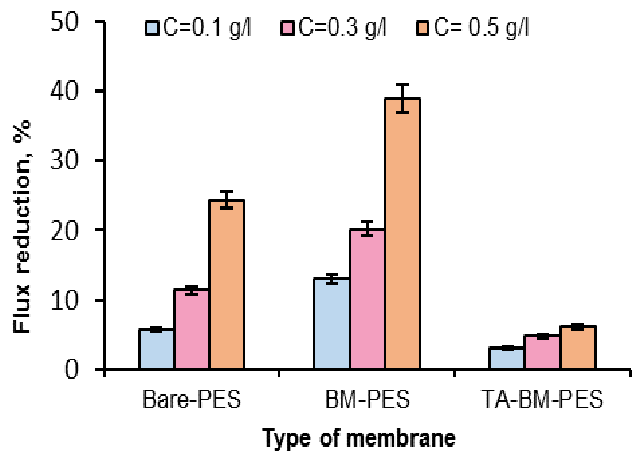

Fig. 12. The results of flux reduction ratios of NF membranes at various feed composition.

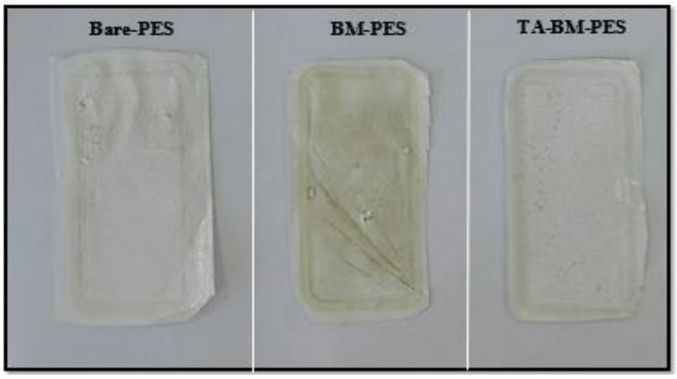

Fig. 13. The photo of nanocomposite membrane after licorice fouling.

4. Conclusions

In this paper, the unfilled and 0.5 wt.% BM/PES and TA-BM/PES blended membranes were applied for purification of licorice aqueous solutions as model foulant. The effect of variable operational parameters on the performance of NF membranes was studied to further clarify the effective physical and chemical foulants on the improvement of membrane permeability and rejection capability. Experimental results revealed that enhancement of transmembrane pressure and cross-flow velocity causes increment of permeated flux and rejection percentage levels of NF membranes. On the other hand, enhancement of hydrodynamic conditions has been substantial effect on the progress of fouling phenomenon. The assessment of foulant feed composition for NF membranes indicated that most fouling was occurred at the highest licorice concentration of 0.5 g/l. It may be related to deposition of greater amount of licorice on the membrane surface at high foulant concentration.

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