Crossflow Filtration of Sodium Chloride Solution by A Polymeric Nanofilter: Minimization of Concentration Polarization by a Novel Backpulsing Method

Kambarani, Masoud
Membrane Laboratory, Academic Center for Education, Culture and Research (ACECR), Tehran, I.R. IRAN

Bahmanyar, Hosein*+
Surface phenomena and Liquid-liquid Extraction Research Laboratory, University of Tehran, Tehran, I.R. IRAN

Mousavian, Mohammad Ali
School of Chemical Engineering, College of Engineering, University of Tehran, Tehran, I.R. IRAN

Mousavi, Seyed Mahmoud
Department of Chemical Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad, I.R. IRAN

ABSTRACT: *In the present study, the production of low-salt water from salty water by nanofiltration as well as membrane fouling was investigated. Furthermore, a new method was proposed and tested experimentally for creating the backpulse in order to minimization of fouling and increase of the filtration efficiency. In the proposed method, the permeate was used instead of gas for creating the backpulse. To test the quality of this method, experiments were conducted using NaCl solution. In these experiments, the backpulse interval was changed and in constant backpulse duration, the effect of this parameter on the permeate flux was investigated. The results confirmed the effectiveness of the proposed system, especially when the concentration of the saline solution was increased.*

KEYWORDS: *Nanofiltration; Membrane; Fouling; Cross flow filtration; Backpulse duration; Backpulse interval.*

INTRODUCTION

Separation processes especially membrane filtration are one of the most used operations in chemical industry. There are many advantages for utilizing the membrane filtration in chemical industry. Nowadays, membrane technologies are more frequently used for separation of widely varying mixtures and can compete successfully

* *To whom correspondence should be addressed.*
+ *E-mail: hbahmany@ut.ac.ir*
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with traditional schemes [1]. The water recovery from wastewater and the production of fresh water from seawater are the most important application of membrane filtration. Usually reverse osmosis is used to produce fresh water but in recent years, nanofiltration is used for production of drinking water instead of or in addition to RO. Thus, the cost of produced fresh water has decreased [2, 3].

Despite the existence of many advantages for membrane filtration, the biggest problem of this process is the decrease of permeate flux affected by fouling of membrane. During membrane filtration, macromolecules and particles accumulate on the membrane surface. Moreover, particles enter into the membrane pores and reduce the membrane pore size. Decrease of the membrane pore size and the formation of a cake or gel layer on the membrane surface increase the resistance.

Depending on the type of membrane and feed, the different mechanisms have been considered for membrane fouling. *Sondhi & coworkers* [4, 5] introduced three mechanisms of pore blocking, pore constriction and cake formation for the fouling. The first two mechanisms are true for porous membrane and in these mechanisms, permeate flux drops sharply in the initial moment of filtration and reach next to zero. In cake formation mechanism, although the initial decline in permeate flux is high, but the permeate flux doesn't reach zero in a short time. The flux decline in these mechanisms was shown by different equations.

In industrial applications, there are some common methods to deal with the fouling problem:

1) Prevention: Feed pretreatment is a way to prevent the fouling. For this purpose, a filter with pore size larger than the membrane is used before the main membrane filtration. Accuracy in appropriate pretreatment such as special removal of solids, colloids and bacteria can have a significant impact on fouling reduction.

2) The removal of membrane fouling through the cleaning by chemicals (alkaline or acidic solution, organic solvents, etc.): In this method, the filtration operation is stopped and the washing operation is done by chemicals. This method reduces useful time of the filtration.

3) The reduction of the fouling during the filtration: It has been used in recent years and is being developed rapidly. Two major methods of ultrasonic and backpulsing are used for this purpose. The backpulsing method is described in the present paper.

Backpulsing is an effective technique for reducing the fouling phenomenon in membrane, improving the overall filtration rate and extending the cleaning intervals (the time between two consecutive membrane cleanings). In backpulsing method, periodically reversing TransMembrane Pressure (TMP) is used for membrane cleaning. Permeate liquid is forced back through the membrane to the feed side and dislodges the deposited foulants, which are carried out of the membrane module by flow of retentate [4]. Although the effective time of the filtration is decreased when the backpulsing is used, but if the backpulse is used at right time, the positive effect of the backpulsing on permeate flux will be quite significant. There are two important parameters of Backpulse Duration (BD) and Backpulse Interval (BI) associated with backpulsing. Backpulse duration is defined as the amount of time that filtration system is operating under negative TMP. Backpulse interval is the duration of time between two consecutive pulses. Nowadays the backpulsing method is quickly becoming more widely utilized for the fouling control and so must be improved for easier and more economical use.

Key point and bottleneck to use this technique is backpulse device. So far different methods and devices have been used to produce the reverse flow. Some of them are introduced herein:

Sondhi et al. [4, 5] used a backpulse device including a cylinder and a piston. A nitrogen cylinder was used as the pressure source for the backpulse device. The backpulse unit sent a nitrogen pulse to the back of a piston that was attached on the permeate side of the membrane module. The piston expanded on the permeate side and caused some reversal through the membrane for a very short duration as a backpulse.

Mc Alexander et al. [6] used backpulse to control membrane fouling. A peristaltic pump was used to provide TransMembrane Pressure (TMP). During the recovery segment of the experiment, the pump supplied a vacuum to separate permeate. In backpulse period, the direction of the peristaltic pump rotation was changed. So a small amount of permeate returned to the permeate side of membrane. In this state, the TMP was zero and pressure of the peristaltic pump was enough to produce backpulse.

Ma et al. [7, 8] also tested backpulse to reduce the membrane fouling. A system with dual ability to create

Table 1: Conductivity vs. concentration for sodium chloride solutions at 25°C.

Concentration (wt. %)	0.01	0.03	0.1	0.3	1	3	5	10
Conductivity (μs/cm)	210	617	1990	5690	17600	48600	78300	140000

Table 2: Summary of the membrane characteristics

Membrane type	Thin-film composite
Membrane material	Polyamide (PA)
Max. operating pressure	600 psi (4.14 Mpa)
Operating pH range	2.0 – 11.0

backpulse (liquid backpulse and gas backpulse) was used. In liquid backpulse, pressurized N_2 was conducted to upper surface of liquid in a tank and then pressurized liquid was directed to the permeate side of membrane. In gas backpulse, pressurized N_2 or another inert gas came into the permeate side of membrane without an interposition.

E. Larsson [9] used microfiltration to eliminate the bacteria or fractionate the proteins during milk processing, using backpulse to reduce the fouling. The backpulse was created when piston blocked the permeate flow channel.

F. Meacle et al. [10] used a gas backpulsing device to reduce fouling. They used compressed N_2 as pressure source to create the backpulse.

L.C. Gramms et al. [11] presented a device containing a piston and cylinder assembly which was used for the backflushing and backpulsing of filters. Compressed air was used to cause the movement of the piston in the backpulse time.

The methods used to create backpulse in the aforementioned researches have the following characteristics:

1) Using gas as pressure transmitter has three problems, A: gas compressibility causes the delay in rapid pressure transmission to the membrane module, B: the return to the normal condition is slow and with fluctuation, C: the cost of compressed gas is imposed on the system.

2) A large part of the equipment suffers high pressure when the backpulse is created.

3) The actual time used to create the backpulse is always greater than the adjusted value for BD

4) Because the system has delay and fluctuation, thus a part of time is wasted.

To resolve these problems, in the present paper a new method is proposed and implemented to create backpulse. The main characteristic of the proposed method is the elimination of pressurized gas for creating backpulse. In this method, pressurized permeate is used to create backpulse. Therefore, speed of the backpulse is optimized and effective time of the filtration increases.

EXPERIMENTAL SECTION

Materials

To investigate the quality of nanofiltration and the backpulse system, NaCl solution with different concentrations was selected as feed for the filtration in all experiments. NaCl (Merck Millipore, 106404) and deionized water were used in the preparation of all experimental solutions. To compare feed concentrations, standard value of conductivity of NaCl solutions is shown in Table 1. A thin-film polymeric nanofilter (NE4040-90, CSM) was used in all experiments. Summary of membrane characteristics is shown in Table 2.

Equipment

A schematic of cross flow membrane filtration system for accomplishment of the experiments is shown in Fig. 1. As shown in this Fig., there is a system for cross flow filtration with backpulse equipment. The system has two pressure indicators PI1 and PI2 and also two pressure sensors PS1 and PS2. For normal filtration, the feed was stored in a five-liter tank (T1) and was sent to the Nanofilter Membrane Module (NFM) with an effective surface of 112 cm^2 by a high pressure rotary vane pump (P1)(Procon pump). Flow rate of the feed was measured by a rotameter (RO1). The valve V2 was used to adjust TMP. The permeate was stored in a two-liter tank (T2) and was pumped continuously into the loop of T2, P2 (backpulse pump)(Upricht, up 7000, diaphragm pump), permeate side of NFM, and T2. The permeate was recirculated to T1 to have a constant feed concentration. At this stage, there was low pressure in the stream, and two valves V4 and V5 were open. To create backpulse, the solenoid valve V5 was closed and the flow line of P2 was blocked. Then, the valve V4 was manually turned

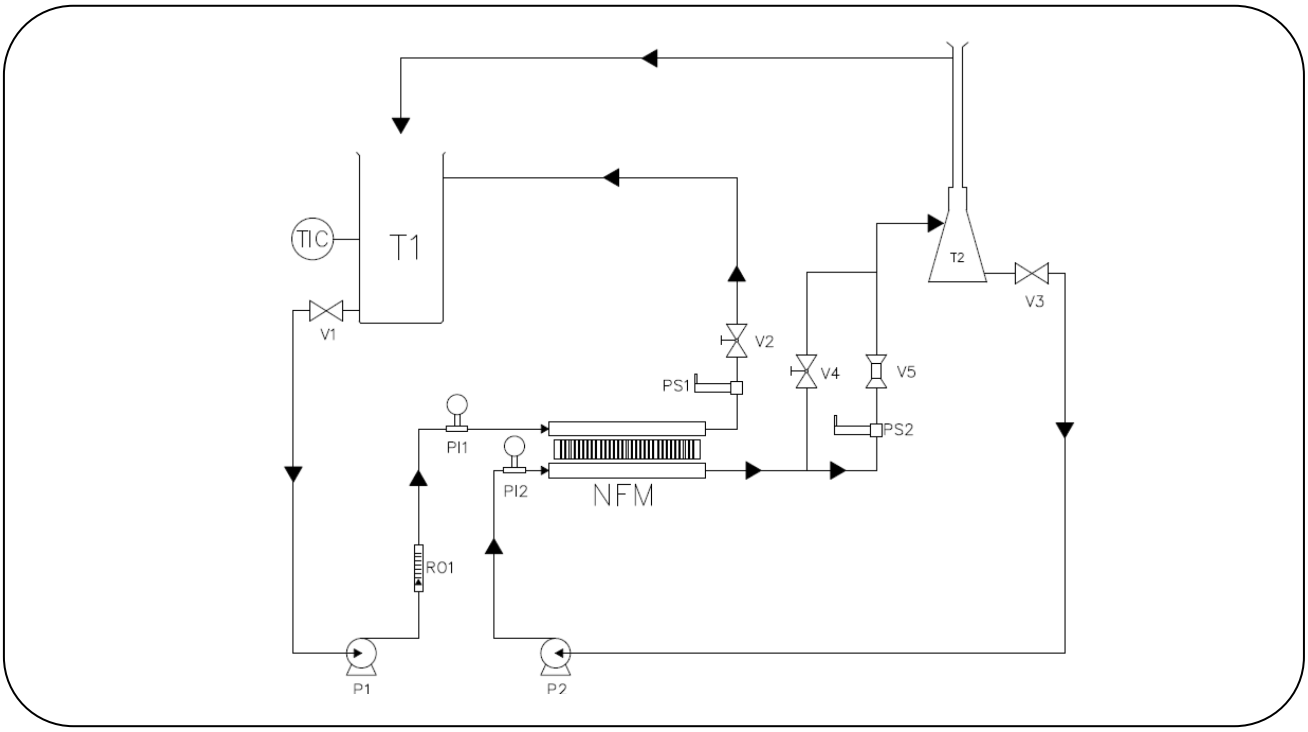

Fig. 1: Experimental set-up.

slowly to obtain the necessary pressure for backpulse. To control BD and BI, the solenoid valve V5 was controlled by a twin timer (DH48S-S, twin timer). The accuracy of this timer was one tenth of a second and it could control both the times. The feed and permeate pressures were measured by two high accuracy sensors (BT-210, Atek sensor technology AG) used in conjunction with data acquisition software (USB-4718, Advantech) and a personal computer. The saline solution concentration was measured by a conductometer.

Experimental procedure

The tests were classified in three parts. At first, the effect of the feed concentration on the permeate flux and the removal efficiency was evaluated. To this purpose, in different feed concentrations (0.1, 0.3, and 1 wt. % NaCl), the flux (J) and the rejection (R) were measured. Then, to show how to reduce the permeate flux during the filtration operation and also to fit a mathematical model, the variations of the permeate flux as a function of time was measured in a constant feed concentration. Finally, the most important objective of this research i.e. using backpulse to minimize the fouling and increase the filtration efficiency was examined. In this step, the amount of difference in the permeate flux with respect to the presence and absence of backpulse was measured. Furthermore, the effect of BI on the permeate flux was investigated. Thus, the experiments were done in three different feed concentrations (0.1, 0.3, and 1 wt. % NaCl) and several BIs (9, 59, 99, and 150 s).

RESULTS AND DISCUSSION

The effect of the feed concentration on the permeate flux and the removal efficiency

For NaCl solution, increase of the feed concentration results in increasing the concentration of Na^+ and Cl^- on the membrane surface. This condition causes two negative effects on the filtration operation i.e. reduction in both the permeate flux and the Apparent solute rejection [12-16]. The reduction of the permeate flux is because of the accumulation of Na+ and Cl- on the membrane surface which is called concentration polarization [13]. The permeate flux decline due to the concentration polarization or the deposition, can be described by resistance-in-series model corresponding with the following equation:

$$J = \frac{\Delta P}{\mu\left(R_m + R_f + R_p\right)} \quad (1)$$

Table 3: Results of the flux experiments.

Feed Concentration (wt. %) (approximately)	Feed conductivity (μs/cm) (Exactly)	Flux without backpulse (after 1 hr operation) (L/m^2h)	Flux with backpulse* (L/m^2h)	Change in permeate flux (%)
distilled water	0.8	32.8	32.8	0
0.1	1900	25. 25	25.7	1.8
0.3	6250	15.4	16.26	5.6
1	17400	3.96	5.45	37.6

BD: 0.9 s, BI: 150 s, TMP = 5 bar. Back TMP = 6 bar. Cross Flow velocity = 1.25 m/s

Table 4: The rejection of NaCl by the polymeric nanofilter at TMP = 5 bar.

Feed concentration (wt. %) (approximately)	0.1	0.3	1
Feed conductivity (μs/cm)	1990	5690	17600
Permeate conductivity (μs/cm)	205	635	2175
Rejection (%)	90	89	88

where ΔP is the TMP, μ is the permeate viscosity, and R_m, R_f, and R_p are the resistances of the membrane, fouling, and concentration polarization, respectively [5, 17, 18, 19]. To evaluate the effect of the feed concentration on the permeate flux; the experiments were done in three different feed concentrations. The results are shown in Table 3. As shown in this Table, the permeate flux was decreased by increasing the feed concentration.

Higher feed concentration results in increasing concentration polarization and decreasing rejection. The experimental results for the rejection are shown in Table 4, presenting a good rejection of NaCl by the polymeric NF. With respect to the Table data, the solute rejection was ranged from 88% to 90%. The rejection value was decreased from 90% to 88% with increasing the feed concentration from 0.1 to 1 wt. %.

Permeate flux modeling

To present a model for the permeate flux reduction in terms of time in the salty water system at the start-up of filtration operation, an experiment was done. The feed concentration in this test was considered 1 wt. %. The results are shown in Table 3. According to the following overall differential equation, the influence of fouling on the flux through the membrane can be demonstrated [20], the best fitted equation on the obtained experimental data could be described as Equation 2:

$$J = J_0\left[1+k\left(2-n\right)\left(A\cdot J_0\right)^{n-2}\cdot t\right]^{n-2} \quad (2)$$

$$J = 32.8\times\left(1+0.04t\right)^{-0.6}$$

As shown in Fig. 2, the proposed model is fitted to the experimental results very well.

The effect of backpulse on the permeate flux

In this paper, one way to prevent the membrane fouling was evaluated. When the concentration of the salt in the feed was increased, the permeate flux decreased rapidly and use of backpulse had a good effect on the permeate flux. In order to have a more complete understanding of the backpulse effect, three different concentrations of the salt were considered and the amount of change in the permeate flux was calculated. The experimental results are shown in Table 3. As shown in this table, the backpulse effect strongly depended on the salt concentration and a significant recovery of the permeate flux (more than 37% improvement) was clearly observed in the maximum feed concentration (1 wt. %).

The effect of BI on the permeate flux

To evaluate the effect of BI on the permeate flux, the experiments were done in three feed concentrations of 0.1, 0.3, and 1 wt.% and four BIs of 9, 59, 99, and 150 s. In all experiments, BD, TMP and backpulse TMP were 0.9 s, 5 bar, and 6 bar, respectively.

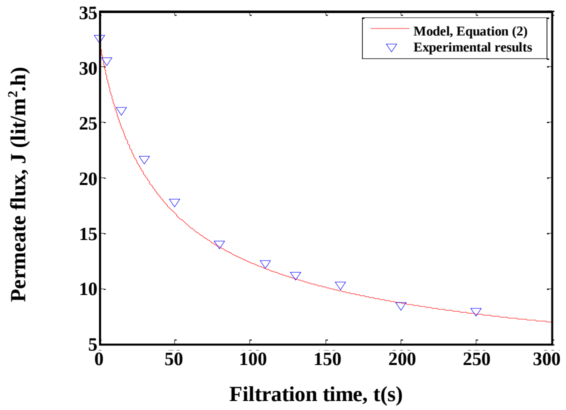

Fig. 2: Permeate flux reduction at the start-up of filtration operation.

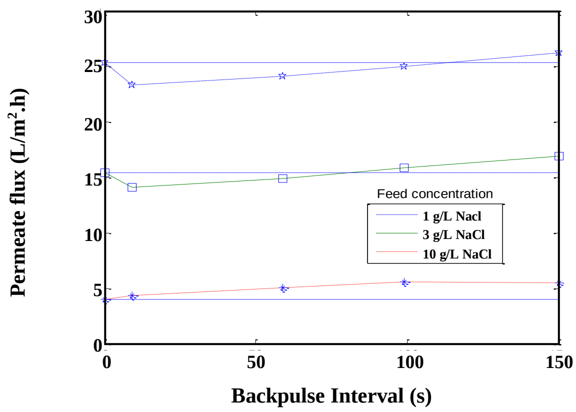

Fig. 3: The effect of BI on the permeate flux.

For comparison, the clean membrane water flux is 32.8 lit/m^2.hr. The results are shown in Fig. 3. These experiments led to several significant results:

1) When a feed with very low concentration of solute was used, the resistance caused by the accumulation of the solute molecules deposited on the membrane surface was increased very slowly, and the use of short BI, not only did not have a positive effect on removing the resistance layer and increasing the permeate flux, but it also caused the permeate flux to be reduced. As can be seen in Fig. 3, when the feed concentration was 0.1 wt. %, the positive effect of backpulse was observed for the BI in more than 115 s. For the feed concentration of 0.3 wt. %, this BI was about 85 s. In the BI over these values, the amount of the permeate flux was greater than the initial value.

2) When a feed concentration of 1 wt. % was used, a maximum point in the permeate flux curve was observed. The maximum flux took place when the BI was about 99 s. So this time was the optimum BI for that condition. This means if the BI was greater than 99 s, the resistance layer was formed and it had a negative effect on the permeate flux.

CONCLUSIONS

In the present study, the ability of NF to remove the NaCl from saline water was investigated, and also an appropriate model for the permeate flux reduction in terms of time was introduced. Moreover, a new method for backpulse creation was designed and tested. With respect to the results, the backpulse effect depended on the feed concentration and a noticeable recovery of the permeate flux was obtained in the upper feed concentration. In backpulsing method, the effect of BI on the flux recovery was depended on the feed concentration. Generally, this **novel** method does not have some of the problems of the previous methods that were used for backpulse creation.

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REFERENCES

[1] Ravanchi M.T., Kaghazchi T., Kargari A., Application of Membrane Separation Processes in Petrochemical Industry:a Review, *Desalination*, **235**: 199-244 (2009).

[2] Alzahrani S., Mohammad A.W., Hilal N., Abdullah P., Jaafar O., Comparative Study of NF and RO Membranes in the Treatment of Produced Water—Part I: Assessing Water Quality, *Desalination*, **315**: 18-26 (2013).

[3] Khalik A., Praptowidodo V.S., Nanofiltration for Drinking Water Production from Deep Well Water, *Desalination*, **132**: 287-292 (2000).

[4] Sondhi R., Bhave R., Role of Backpulsing in Fouling Minimization in Crossflow Filtration with Ceramic Membranes, *Journal of Membrane Science*, **186**: 41-52 (2001).

[5] Sondhi R., Lin Y.S., Alvarez F., Crossflow Filtration of Chromium Hydroxide Suspension by Ceramic Membranes: Fouling and Its Minimization by Backpulsing, *Journal of Membrane Science*, **174**: 111-122 (2000).

[6] McAlexander B. L., Johnson D.W., Backpulsing Fouling Control with Membrane Recovery of Light Non-Aqueous Phase Liquids, *Journal of Membrane Science*, **227**: 137-158 (2003).

[7] Ma H., Hakim L.F., Bowman C.N., Davis R.H., Factors Affecting Membrane Fouling Reduction by Surface Modification and Backpulsing, *Journal of Membrane Science*, **189**: 255-270(2001).

[8] Ma H., Bowman C.N., Davis R.H., Membrane Fouling Reduction by Backpulsing and Surface Modification, J*ournal of Membrane Science*, **173**: 191-200 (2000).

[9] Emma Larsson, "Microfiltration of Milk with Backpulsing", Tetra Pak Processing Systems AB, Lund, Sweden, February (2011).

[10] Francis Meacle, Anne Aunins, Rachel Thornton and Ann Lee, Optimization of the Membrane Purification of A Polysaccharide-Protein Conjugate Vaccine Using Backpulsing, *Journal of Membrane Science*, **161**: 171-184 (1999).

[11] Gramms L.C., Bowman R.W., Craycraft R.R., Backpulse Piston Assembly for Crossflow Filters, *USP 5512167*, (1996).

[12] Baker R.W., "Membrane Technology and Application"s, Wiley, , ISBN 0470854456 , p. 165 (2004).

[13] Prisciandaro M., Mazziotti di Celso G., Back-Flush Effects on Superficial Water Ultrafiltration, *Desalination*, **256**: 22–26 (2010).

[14] Bohdziewicz J., Bodzek M., Wqsik E., The Application of Reverse Osmosis and Nanofiltration to the Removal of Nitrates from Groundwater, *Desalination*, **121**: 139-147 (1999).

[15] Ku Y., Lee P., Wang W., Removal of Acidic Dyestuffs in Aqueous Solution by Nanofiltration, *Journal of Membrane Science*, **250**: 159–165(2005).

[16] Amini M., Arami M., Mahmoodi N.M., Akbari A., Dye Removal from Colored Textile Wastewater Using Acrylic Grafted Nanomembrane, *Desalination*, **212** (2010).

[17] Choi J.S., A Systematic Approach to Determine the Fouling Index for a RO/NF Membrane Process, *Desalination*, **238**: 117-127 (2009).

[18] El Rayess Y., Albasi C., Bacchin P., Taillandier P., Raynal J., Mietton-Peuchot M., Devatine A., Cross-Flow Microfiltration Applied to Oenology: A Review, *Journal of Membrane Science*, **382**: 1-19 (2011).

[19] Robert Field, "Fundamentals of Fouling, Membranes for Water Treatment": Volume **4**, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim (2010).

[20] Cui Z.F., Muralidhara H.S., "Membrane Technology: A Practical Guide to Membrane Technology and Applications in Food and Bioprocessing", Utterworth-Heinemann, (2010).