Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.1003978

57 (2016) 5376–5384
March

Taylor & Francis
Taylor & Francis Group

Effects of various frequencies and powers of ultrasound on cleaning of flat sheet membrane during and after microfiltration

Abdolmajid Maskooki*, Masoud Hashemi Shahraki, Morteza Mohamadi

Department of Food Processing, Research Institute of Food Science & Technology (RIFST), Km12 Asian Highway, P.O. Box: 91735-147 Mashhad, Iran, Tel. +98 51 35003109; Fax: +98 51 35003150; email: a.maskooki@rifst.ac.ir (A. Maskooki), Tel. +98 9381321295; Fax: +98 51 35003150 (M.H. Shahraki), Tel. +98 9155149853; Fax: +98 51 35003150 (M. Mohamadi)

Received 27 June 2014; Accepted 26 December 2014

ABSTRACT

Membrane processing has found many applications in various industries as an advanced method of separating materials in liquid or gas. The greatest obstacle in using membrane filtration technology is fouling, particularly in porous membranes. The fouling phenomenon is the result of organic and inorganic materials depositing on the surface and pores of membrane. In this study, the effect of ultrasound frequency (37, 80 kHz, and tandem) and amplitude of sonication (30, 60, and 90%) were evaluated on flux recovery during and after microfiltration. Results showed that the 37 kHz and tandem frequencies significantly improved the permeate flux at earlier minutes, particularly when tandem frequency was applied. In addition, the permeate flux was increased as the sonication power increased. However, the interaction effects between frequency and sonication power showed that the ultrasound frequencies were more effective than sonication power on flux recovery. Furthermore, the calculation of fouling percentage showed that both low frequencies and high amplitude together significantly reduced the fouling agents during the cleaning process. However, there were no remarkable statistical effects among the same levels of sonication power during the cleaning process. The interaction effects of various frequencies and powers of ultrasound on cleaning membranes were evaluated and more cleaning efficiency was observed in comparison to sonication power when low frequency applied.

Keywords: Membrane separation; Ultrasound; Fouling; Membrane cleaning

1. Introduction

Membrane filtration technology has been applied to a wide range of various industries including water and wastewater treatment, Food (dairy, juice, and brewery) biotechnology (bioreactors), and medicine (hemodialysis). The most important techniques of membrane processing are microfiltration (MF), ultrafiltration (UF), Nanofiltration, and reverse osmosis [1]. These techniques are widely used in many separation procedures as well. The major disadvantage of membrane separation processing is fouling [2]. Fouling phenomenon is due to accumulation of feed constitutes such as colloids, proteins, macromolecules, and inorganic materials on the membrane surfaces and pores [3–5]. In this situation, the permeate flux significantly reduces while the maintenance and operating costs increases. Moreover, the concentration polarization assists to flux decline due to the formation of

---

*Corresponding author.

boundary layer in the vicinity of membrane surface or reducing the feed flux velocity during cross flow filtration [6]. Many procedures are being applied to reduce fouling and cleaning of the membrane by adopting hydrodynamic and back-flushing technique [7,8], and modifying the membrane material [9]. The conventional cleaning protocols recommended by membrane manufacturers, is included in a series of acid–alkaline or enzymes cleaning cycles depending on the feed and the membrane materials. The other techniques based on pretreatments such as coagulation, adsorption, and ozonation are usually utilized before membrane filtration to minimize fouling problems [10,11]. Although, pretreatments might have been employed to minimize fouling, most membrane cleaning techniques are still practically inadequate for membrane filtration systems. Typical methods of membrane cleaning that have been used in industrial applications are forward flushing (spiral wound and tubular) and backwashing (hollow fiber), which are useful with colloidal suspensions [12]. In the meantime, other techniques, such as hydraulic, chemical, mechanical, and electrical cleaning processes, were also utilized to mitigate fouling problems [2,10]. In spite of numerous cleaning procedures, it seems that the conventional techniques still appear to be inadequate for practical membrane filtration systems. Among new techniques, ultrasound has been a promising method in such membrane application. Power ultrasound has been well known as an effective method for cleaning materials because of the cavitation phenomenon [12]. Cavitation is occurred when acoustic waves transmitted in a liquid medium, such as water, because of the dispersion of ultrasonic waves in liquid medium; the medium is subjected to alternating rarefaction and compression cycles. If the distance of water molecules are longer than the distance of wander walls radius during the rarefaction cycle, bubble cavities are formed. This cavity may dissipate back into the liquid or grow to a resonant size and fluctuate about this size, and or grow to a size, at which the surface tension forces of the liquid cause it to collapse on itself. The latter phenomenon is termed cavitational collapse. The collapse of cavities results in extreme conditions producing light emission, shock waves, and localized high temperatures (up to 400˚C) and pressures (up to 100 Mpa) [13]. Cavitational effects also produce a number of phenomena that result in high velocity fluid movement and sufficient energy to overcome the adhesion between the foulant and the membrane, and remove the foulant from the surface of membrane. Therefore, cleaning effects of ultrasound is due to cavitation phenomenon. Furthermore, the creation of turbulence in feed and permeate in the vicinity of membrane can assist on flux recovery and reducing foulants [14]. Many investigations were carried out to use of power ultrasound for cleaning the polymeric membrane. However, the main parameters of sonic waves such as frequency, amplitude, combinational frequencies, and their interactions are still unknown. The aims of this investigation are to evaluate the majority effect of wavelength, amplitude, interaction of power, and intensity of sonication on cleaning of membrane during MF of milk as feed solution, as well as cleaning efficiency of ultrasound and deionized water in forward flushing cleaning method on fouled membranes.

2. Materials and methods

2.1. Materials

Skimmed milk powder was purchased from the local market and used as feed with 1 wt% solid content concentration during MF processing and provide the fouled membrane. The Physicochemical properties of feed samples were shown in Table 1.

Flat-sheet polyvinylidene fluoride (FDA-Approvable PVDF–MFB Sheet Membrane) Sepro Company USA MF membrane with 0.2 μ pore size and effective membrane area 112 cm^2 was used in Minitan S (Millipore Inc.).

The membrane (15 × 11 cm) was placed between two large perforated silicon rubbers in order to create a series of linear crossflow channels. Two acrylic manifolds of thickness 2.3 cm were placed in upper and lower sides of membranes, which were covered by two stainless steel plates of 1.1 cm thickness in the upper side.

2.2. Methods

2.2.1. Separation

The experimental setup used for this investigation is shown in Fig. 1. The system consists of a feed tank that connected to an N_2 gas cylinder for supply sufficient and constant pressure. The inlet of feed solution and outlet of retentate pressure were measured using two pressure gauges.

The difference between feed and retentate flow was measured by flow meter and considered as the flow of permeate. In order to achieve more accuracy, the volume of permeate was also measured by the permeate weight as a unit of time on an electronic balance A&D Co.0.01 mg. The permeate flux and hydrodynamic resistance of membranes were expressed as volumetric flux ($m^3/m^2/s$) and resistance of membrane, respectively. During the fouling process,

Table 1
Physicochemical properties of skimmed milk as feed sample

Ash (g/100 g)	Lactose (g/100 g)	Protein (g/100 g)	Density (g/100 ml)	Viscosity (cp)	Conductivity (µs/cm)	Brix (%)	TDS (ppm)	pH	Particle size of powder range (µm)
0.00721	0.04657	0.03035	1.032	1.47	910	1.11	460	6.93	20–250

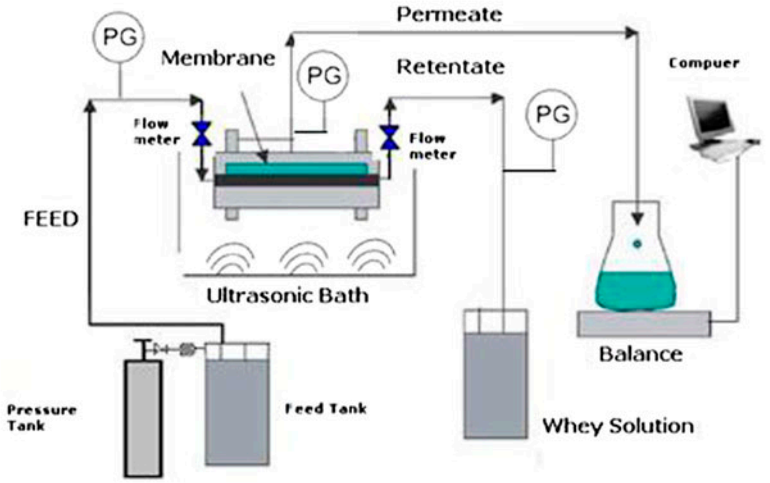

Fig. 1. Experimental setup of cross flow MF under sonication (PG represented of pressure gage).

permeate and retentate were recycled to the feed tank to maintain the feed concentration.

2.2.2. Sonication

The Minitan S. device consists of flat sheet membrane which was directly placed in ultrasonic cleaning bath. The dimensions of ultrasonic cleaning bath were 200 × 300 × 505 mm (Elmasonic, Germany) that was connected to temperature control circulator. The membrane unit was kept 10 cm above the bottom and 7 cm far from the transducers. Generally, the ultrasound power with low frequencies and high intensities are used in the range of 20–100 kHz, and up to 1,500 W, respectively [15]. The sonicator generated 37 and 80 kHz frequencies and output power 1,250 W. The ultrasonic experiments were performed in 37, 80 kHz, and tandem (switch on 37 and 80 kHz alternatively for each 1 min) in various sonic power 30% (375 W), 60% (750 W), and 90% (1,125 W).

All of experimental tests were carried out for 30 min and in each test, a new membrane was used. We experimentally found that this time duration is enough to obtain the constant value. However, the MF processing by milk solution was continued when the flow rate of permeate reached below of 1 ml/min to produce fouled membrane.

Evaluating the cleaning effects of ultrasound on fouled membranes was carried out by a forward flushing method using deionized water under same conditions of ultrasound for 30 min. During cleaning, the filtration was continued with deionized water as feed under different ultrasonic treatments. The fluxes and hydrodynamic resistances of new and cleaned membranes were separately measured by passing deionized water through membranes in the same condition in each experiment. The measured values were used as reference to membrane permeability. The viscosity of permeate was measured by Brookfield viscometer Tokimec model BL.

2.2.3. Parameters calculation

The permeate flux (J) was measured by using the following Eq. (1):

$$J = (W_{\text{ti}} - W_{\text{ti}} - 1) \times (d \times \Delta t) - 1 \tag{1}$$

where W_{ti} is permeate weight in time i; $W_{ti}-1$ is permeate weight in time $i-1$; d is density of permeate; and Δt is time interval.

The hydrodynamic resistance was calculated by Eq. (2):

$$R = \Delta P/\mu \times J \tag{2}$$

where ΔP is the steady-state system pressure; μ the viscosity; and J the permeate flux.

ΔP could consider as crossflow pressure and was defined as Eq. (3):

$$\Delta P = (P_F - P_R/2) - P_P \tag{3}$$

where P_F is feed pressure; P_R retentate pressure; and P_P is permeate pressure.

For quantify of the effect of ultrasonic waves on permeation flux, flux enhancement factor was defined as Eq. (4):

$$EF\% = (J_{US} - J) \cdot J^{-1} \times 100 \tag{4}$$

where J_{US} is the permeate flux under US treatment; and J is the control permeate flux.

In order to evaluate the effect of US membrane cleaning, cleaning efficiency was defined as Eq. (5):

$$CE\% = (R_F - R_C)/(R_F - R_N) \times 100 \tag{5}$$

where R_F is the fouled membrane resistance; R_C is the cleaned membrane resistance; and R_N is the new membrane resistance.

The differences between deionized flux before and after membrane fouling per unit of cleaning time were represented as fouling percent and calculated using Eq. (6):

$$\text{Fouling } (\%) = (1 - J_{wp}/J_w) \times 100 \tag{6}$$

where J_{wp} and J_w are flux of membrane after and before fouling, respectively.

2.2.4. Statistical analysis

Each treatment was performed at least three times. The acquired raw data were statistically analyzed using multifactor design in ANOVA table. The least significant differences calculated and the obtained means evaluated by Duncan's multiple range test. Statistical analysis was performed using SigmaStat 3.1 and Microsoft EXCEL software.

3. Results and discussion

3.1. Permeate flux

Changing in US frequency had a significant effect on permeate flux. By decreasing in US frequency, the permeate flux was increased. As expected, the 37 kHz of US frequency and tandem mode significantly enhanced the permeate flux in comparison with 80 kHz and control, although, there was no significant difference between 37 kHz and tandem mode of US input frequencies. The effects of frequency changing on permeate flux (Tandem mode) and their comparisons with control (no US treatment) were shown in Fig. 2. The highest permeate flux was obtained in the

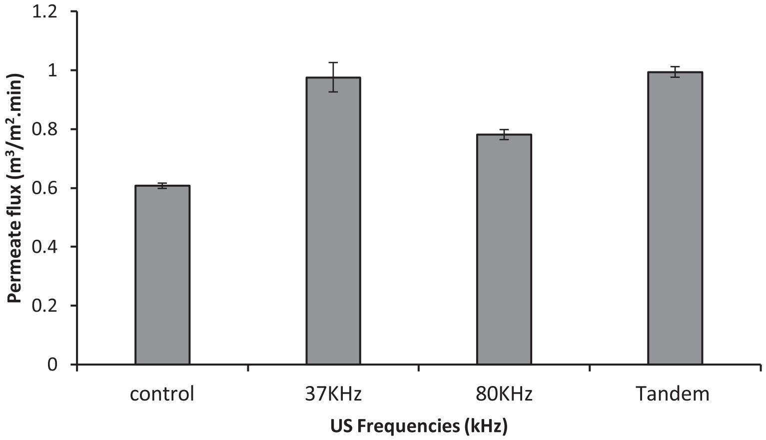

Fig. 2. Changing in permeate flux with US frequencies during MF and their comparison with control.

tandem mode of US frequencies, in other words, changing in US input frequency from 37 to 80 kHz every one min, led to increase the turbulence near the active zone of membrane and resulted in concentration polarization reduction. In addition, because of the bubbles created by cavitation in 37 kHz led to mechanical removal of sedimentations and prevention of membrane fouling. The bubbles created in 80 kHz of US frequency are smaller than 37 kHz and they are able to penetrate to the pores of the membrane and remove the sedimentations inside pores. In the initial stages of the filtration process, no filtration cake was formed and it can be the reason that the US treatment more effective than the initial stages of the process. Maskooki et al. showed the US input frequency changing by intensifying effect, which can lead to decrease in the fouling of polymeric membrane. These results were approved by Avila et al. and Shahraki et al. [16,17].

The results showed that the 37 kHz and tandem frequencies significantly improved the permeate flux at earlier minutes, particularly when tandem frequency was applied. The reason could be that during the MFprocess, the gel layer thickness was increased and led to reduce the effects of ultrasound treatment as much [18].

3.2. Hydrodynamic resistance

Hydrodynamic resistance of membrane shows inhibition across the membrane against total flux. This resistance is included in intrinsic resistance, concentration polarization resistance and resistance of electrically charged particles, Van der Waals, and other factors.

Because the new membrane resistance is constant and the other system parameters such as feed flow rate and type of particles are fixed in this process. Thus, membrane resistance was affected by factors such as pressure changes caused by the deposition of fouling material and the concentration polarization. Therefore, to calculate the hydrodynamic resistance, viscosity of permeates and the difference between input and output pressure are important. Highest hydrodynamic resistance was obtained for control with any US treatment and it had a considerable difference with 80 kHz of US treatment. Although the difference between 37 kHz and tandem mode of US treatments was not significant, 37 kHz and tandem mode of US treatments had a lower resistance in comparison with 80 kHz and control treatments. In general, reducing in the membrane hydrodynamic resistance increases the permeation flux. Same result has been reported by Maskooki et al. [14].

3.3. Effect of US frequency on flux enhancement during filtration

The effect of high intensity of US frequencies was investigated on MF. During the first 10 min, a high decreasing was observed. Since the flux numbers in the first 10 min were much greater than after 10 min, the first 10 min was removed and after 10 min are shown in Fig. 3.

As expected, the 37 kHz of US frequency and tandem mode significantly increased enhancement of the permeate flux in comparison with 80 kHz and control, although, there was no significant difference between 37 kHz and tandem mode of US input frequencies.

At low frequencies, the compression (and rarefaction) cycles are long enough to grow the bubble to a size sufficient to cause disruption of the liquid. As a result, lower ultrasound frequencies had higher cleaning efficiencies than higher frequencies [2,19]. The increase in permeate flux was because of an increase in turbulence and resulted in a decrease in concentration polarization that can be used in the enhancement

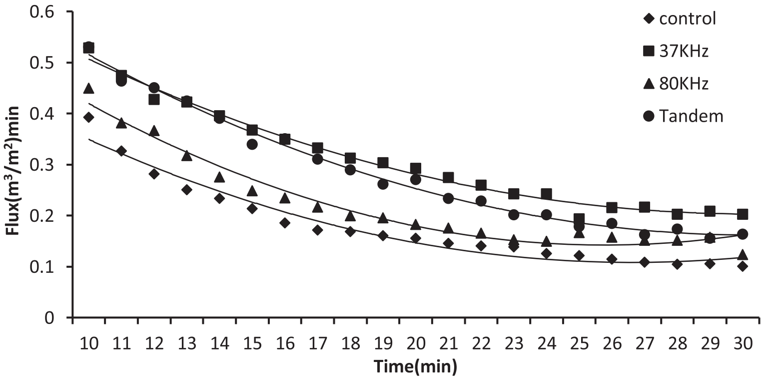

Fig. 3. Permeation of flux under different frequencies after 10 min during MF.

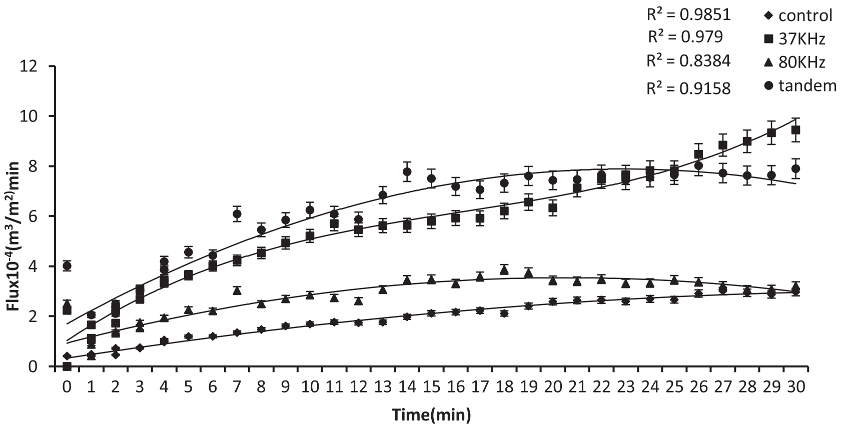

Fig. 4. Effect of different frequencies on membrane cleaning during MF and comparison with control.

of flux in UF and MF. It seems that regularly changing of input waves or tandem mode of US has led to regularly resize of the cavitation bubble and a more desirable condition can be created. The graph of tandem mode is not smooth and showed distortions. The reason of this distortion can be input frequency changing during filtration that led to more turbulence near the active zone of separation.

3.4. *Effect of US frequencies on flux recovery during cleaning*

The fouled flux recovery during 30 min with deionized water under different frequencies is shown in Fig. 4. By increasing the ultrasonic frequencies, the production and intensity of cavitation in liquids decreases, which resulted in a decreased membrane cleaning.

The tandem mode of US frequencies had a higher effect on flux recovery. As demonstrated, the tandem mode of irradiation was too effective to be caused by a change in the size and differences in the collapse power during the cavitation process [6]. Higher frequencies have more cavitation bubbles with time; they are smaller in size and collapse less energetically. They may not be capable of detaching particles from the cake layer as readily as lower frequencies, but they may have more penetration [18].

Results showed that ultrasonic waves were very effective on cleaning efficiency. Best EC (96.2%) was obtained at tandem mode. Other cleaning efficiency under different condition is shown in Table 2.

Table 2
Cleaning efficiencies of fouled membrane under various frequencies and power of ultrasound

Treatment	Cleaning efficiency (%)
Cleaning (without US)	20.374
Cleaning under 37 kHz	91.302
Cleaning under 80 kHz	77.613
Cleaning under Tandem mode	93.833
Cleaning under power 30%	65.601
Cleaning under power 60%	71.832
Cleaning under power 90%	74.909
Cleaning under 37 kHz and power 30%	88.596
Cleaning under 37 kHz and power 60%	90.388
Cleaning under 37 kHz and power 90%	94.922
Cleaning under 80 kHz and power 30%	61.627
Cleaning under 80 kHz and power 60%	83.049
Cleaning under 80 kHz and power 90%	88.164
Cleaning under tandem mode and power 30%	91.806
Cleaning under tandem mode and power 60%	93.517
Cleaning under tandem mode and power 90%	96.177

3.5. *Effect of US power intensity on flux during filtration and cleaning*

The results of US power intensity is shown in Fig. 5, indicating that the permeate flux increased linearly with ultrasonic power, confirming previous results [2,20,21]. Also, there is an indication that further increases in ultrasonic power could lead to higher cleaning efficiency. The same results were obtained on the effect of power intensity in cleaning and filtering. Although in the first seconds of cleaning process, the effects of all power intensity were same, but during cleaning, this difference increased [22,23].

In tandem mode, the flux enhancement factor during filtration, increased from 110.7 to 197.6% by increasing in power intensity of ultrasonic waves from 30 to 90%. Increasing in amplitude of ultrasonic

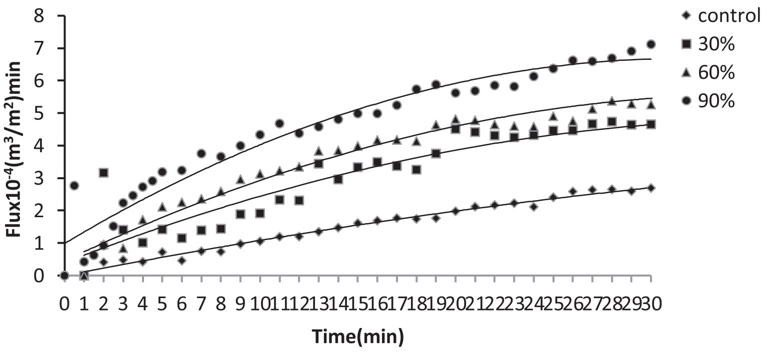

Fig. 5. Effect of power intensity of ultrasonic waves on flux during MF.

waves led to an increased number of cavitation. It could lead to more turbulence in fluid medium, improving detachment and movement of foulant particles from the membrane holes [21].

Changing in flux recovery by increasing the frequency of ultrasound in fixed power intensity, was more than by increasing the power intensity in fixed frequency, so the selected ultrasound frequencies were more effective than selected sonication power on flux recovery.

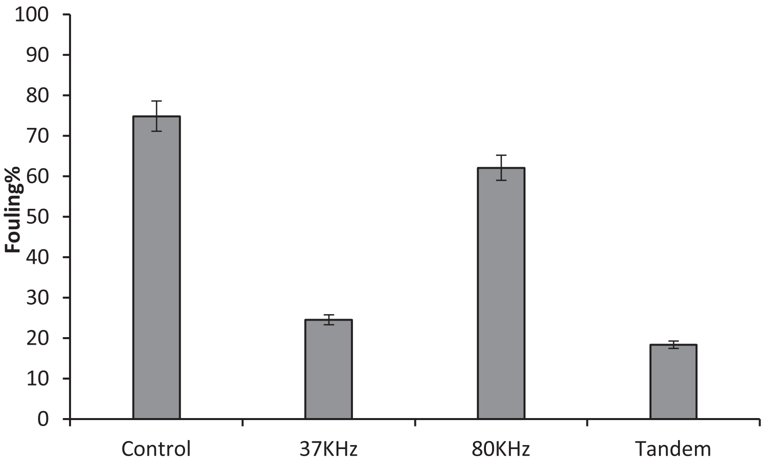

Fig. 6. Fouling percentage of membrane under different US frequencies.

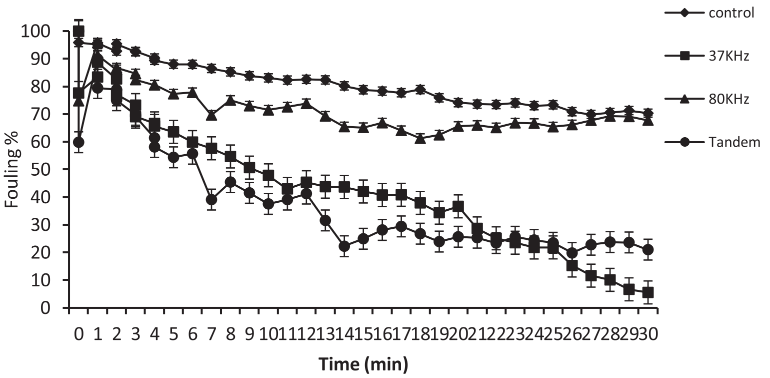

Fig. 7. Fouling percentage rate of membrane under different frequencies.

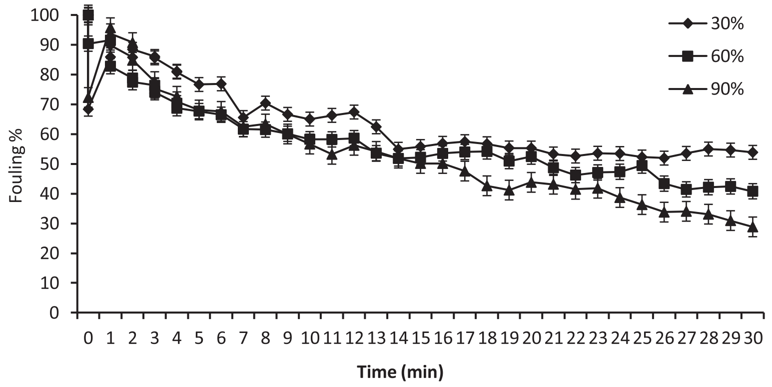

Fig. 8. Fouling percentage rate of membrane under different US power.

3.6. Fouling percentage of membrane under various ultrasound frequencies

Fouling percentage indicates the performance of membrane cleaning under different factors as US treatment and deionized water. The fouling percentage for membrane cleaning under tandem mode and 37 kHz of US frequencies treatment were obtained 20 and 25%, respectively, demonstrating the ability of high-frequency ultrasonic cleaning (Fig. 6). The fouling percentage for membrane under cleaning with 80 kHz of US frequency was more than 60% and showed no good cleaning performance. But, regardless of the overall process time and power, It is observed that ultrasound sonication could reduce the large number of membrane fouling and be used instead of chemical detergents. It should be noted that the control samples without sonication cleaning had very low capability for cleaning. The same results were obtained by Mutakumaran et al. [20] and Hashemi Shahraki et al. [17]. They investigated cleaning capability instead of fouling percentage and also, they have reported that the cleaning capability increased as the US intensity increased.

In Fig. 7, the rate of membrane fouling during the washing process is observed. In all treatments, especially in high frequency 37 kHz, the membrane fouling is reduced during process. The highest fouling percentage was related to the cleaning by using pure water without ultrasonic that was about 30 percent at the end of 30 min, while fouling percentage was over 90% for 37 kHz of US frequency treatment.

3.7. Fouling percentage of membrane under various ultrasound powers

It is observed in Fig. 8 that fouling percent of membrane decreases by an increase in US power intensity. The highest reduction in fouling was obtained at 90% of US power after 30 min of sonication and the lowest cleaning percentage was obtained at 30%.

Acknowledgments

The author acknowledges Iran National Science Foundation (INSF) and Research Institute of Food Science and Technology (RIFST) for financial support. Our special thanks are dedicated Dr Rassoul Kadkhodaee director of RIFST for technical supports and scientific advises.

Symbols

J — permeate flux
W_{ti} — permeate weight in time i
d — density of permeate
Δt — time interval
R — hydrodynamic resistance
ΔP — steady–state system pressure
μ — viscosity
P_F — feed pressure
P_R — retentate pressure
P_P — permeate pressure
J_{US} — permeate flux under US treatment
R_F — fouled membrane resistance
R_C — cleaned membrane resistance
R_N — new membrane resistance
J_{wp} — flux of membrane after fouling
J_w — flux of membrane before fouling

References

[1] R.W. Baker, Overview of membrane science and technology, in: Membrane Technology and Applications, Wiley, West Sussex, 2004, pp. 1–14.

[2] M.O. Lamminen, H.W. Walker, L.K. Weavers, Mechanisms and factors influencing the ultrasonic cleaning of particle-fouled ceramic membranes, J. Membr. Sci. 237 (2004) 213–223.

[3] N. Lee, G. Amy, J.-P. Croue, H. Buisson, Identification and understanding of fouling in low-pressure membrane (MF/UF) filtration by natural organic matter (NOM), Water Res. 38 (2004) 4511–4523.

[4] I.S. Kim, N. Jang, The effect of calcium on the membrane biofouling in the membrane bioreactor (MBR), Water Res. 40 (2006) 2756–2764.

[5] F. Meng, S.-R. Chae, A. Drews, M. Kraume, H.-S. Shin, F. Yang, Recent advances in membrane bioreactors (MBRs): Membrane fouling and membrane material, Water Res. 43 (2009) 1489–1512.

[6] A. Maskooki, T. Kobayashi, S.A. Mortazavi, A. Maskooki, Effect of low frequencies and mixed wave of ultrasound and EDTA on flux recovery and cleaning of microfiltration membranes, Sep. Purif. Technol. 59 (2008) 67–73.

[7] I. Wenten, Mechanisms and control of fouling in crossflow microfiltration, Filtr. Sep. 32 (1995) 252–253.

[8] H. Ma, L.F. Hakim, C.N. Bowman, R.H. Davis, Factors affecting membrane fouling reduction by surface modification and backpulsing, J. Membr. Sci. 189 (2001) 255–270.

[9] S. Ebrahim, H. El-Dessouky, Evaluation of commercial cleaning agents for sea-water reverse osmosis membranes, Desalination 99 (1994) 169–188.

[10] Y. Chen, B. Dong, N. Gao, J. Fan, Effect of coagulation pretreatment on fouling of an ultrafiltration membrane, Desalination 204 (2007) 181–188.

[11] T. Liu, Z.-L. Chen, W.-Z. Yu, S.-J. You, Characterization of organic membrane foulants in a submerged membrane bioreactor with pre-ozonation using three-dimensional excitation–emission matrix fluorescence spectroscopy, Water Res. 45 (2011) 2111–2121.

[12] J. Li, R.D. Sanderson, E.P. Jacobs, Ultrasonic cleaning of nylon microfiltration membranes fouled by Kraft paper mill effluent, J. Membr. Sci. 205 (2002) 247–257.

[13] M.O. Lamminen, H.W. Walker, L.K. Weavers, Cleaning of particle-fouled membranes during cross-flow filtration using an embedded ultrasonic transducer system, J. Membr. Sci. 283 (2006) 225–232.

[14] A. Maskooki, S.A. Mortazavi, A. Maskooki, Cleaning of spiralwound ultrafiltration membranes using ultrasound and alkaline solution of EDTA, Desalination 264 (2010) 63–69.

[15] Y. Hakata, F. Roddick, L. Fan, Impact of ultrasonic pretreatment on the microfiltration of a biologically treated municipal effluent, Desalination 283 (2011) 75–79.

[16] S.R.G. Avila, F. Prabowo, A. Kumar, C.-D. Ohl, Improved ultrasonic cleaning of membranes with tandem frequency excitation, J. Membr. Sci. 415 (2012) 776–783.

[17] M.H. Shahraki, A. Maskooki, A. Faezian, Effect of various sonication modes on permeation flux in cross flow ultrafiltration membrane, J. Environ. Chem. Eng. 2 (2014) 2289–2294.

[18] H. Kyllönen, P. Pirkonen, M. Nyström, Membrane filtration enhanced by ultrasound: A review, Desalination 181 (2005) 319–335.

[19] T. Kobayashi, T. Kobayashi, Y. Hosaka, N. Fujii, Ultrasound-enhanced membrane-cleaning processes applied water treatments: Influence of sonic frequency on filtration treatments, Ultrasonics 41 (2003) 185–190.

[20] S. Muthukumaran, S. Kentish, S. Lalchandani, M. Ashokkumar, R. Mawson, G.W. Stevens, F. Grieser, The optimisation of ultrasonic cleaning procedures for dairy fouled ultrafiltration membranes, Ultrason. Sonochem. 12 (2005) 29–35.

[21] A. Mirzaie, T. Mohammadi, Effect of ultrasonic waves on flux enhancement in microfiltration of milk, J. Food Eng. 108(1) (2012) 77–86.

[22] J.P. Chen, S. Kim, Y. Ting, Optimization of membrane physical and chemical cleaning by a statistically designed approach, J. Membr. Sci. 219 (2003) 27–45.

[23] S. Muthukumaran, S.E. Kentish, G.W. Stevens, M. Ashokkumar, Application of ultrasound in membrane separation processes: A review, Chem. Eng. 22 (2006) 155–194.