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Application of low-pressure reverse osmosis for effective recovery of bisphenol A from aqueous wastes

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ABSTRACT

In this study, bisphenol A (BPA) was removed from aqueous solutions using a low- pressure reverse osmosis system. The influence of various parameters such as feed pressure (136–544 kPa), feed flow rate (0.25–1.172 L/min), feed concentration (30–100 mg/L), and pH (8, 10, and 11) on BPA rejection was investigated. The results showed a maximum rejection of 87.34% for a 50 mg/L feed concentration at 408.1 kPa, pH 8, and 1.172 L/min feed flow rate. The effect of feed pressure on BPA rejection, showed a critical pressure at which the maximum rejection was observed. This critical pressure was measured to be in the range of 408–476 kPa. The most effective parameter on the BPA rejection was feed flow rate which showed a severe concentration polarization at the surface of the membrane. The effect of feed pH revealed a minimum rejection at pH 10.

Keywords: Bisphenol A; Reverse osmosis; Rejection; Membrane

1. Introduction

Phenol and phenolic compounds are common pollutants that exist in the effluents of many industries such as petrochemical units, plastics, dyes, and other industries [1,2]. Bisphenol A (BPA) is one of the phenolic compounds that is produced by condensation reaction of acetone and phenol [3] using an ion-exchange resin as catalyst [4]. BPA is used in manufacture of polycarbonate, epoxy resins, polysulfone, flame retardants, PVC, dental care, baby bottles, and other industrial goods [5–8]. BPA is in group of endocrine disrupting compounds (EDCS) [6] that these compounds have potential for adverse effects on human and wildlife [9]. Removal of EDCS attracted much attention because of estrogenic activity of these compounds [10]. Estrogenic activity of EDCS was first described by Krishnan's group in 1999, through release from polycarbonate flasks [3]. Among the effects of EDCS exposure on human health can point to increase in testicular, prostate, ovarian, and breast cancer [4]. It has been reported that BPA has estrogenic activity even at concentrations lower than 1 ng/L [4]. BPA also has adverse effect on animals and aquatic ecosystem [8,11] that disrupting effect of BPA on aquatic organisms even at concentration lower than 1 μg/L is reported [7]. Stability of EDCS has caused big problems in waste water treatment because these compounds are used in low levels and complete removal of them is hard [12].

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Removal of phenol compounds with estrogenic activity is one of the environmental problems [13]. BPA through release from industries that produced it and also wastewaters of industries that used it enters in the environment and polluted it [7]. Several methods exist that are used for the removal of this compound from aqueous solution, including Fenton's reagent, ultrasonic cavitation, photocatalysis, ozonation, and adsorption by activated carbon, biological and chemical procedures [10,13].

Membrane technologies are useful tools for water treatment because of many advantages such as low power consumption, produce water with high quality and low area required [14,15]. Among membrane processes for the removal of BPA it can be pointed to liquid membrane [8], ultrafiltration [6,16] and nanofiltration [17,18] and reverse osmosis (RO) [19]. Nanofiltration and RO are among membrane technologies that have the ability of removing organic contaminants and EDCS [17,20,21]. By using RO for wastewater treatment, the pollutions are concentrated into small volume compared to the total waste volume [22]. Other advantages of RO process are energy consumption reduction, simple design and easy to operate [23,24] in comparison with traditional processes. But in RO process existence of adverse phenomena including fouling, scaling, and concentration polarization reduce the efficiency of the process [25,26].

Ultra low-pressure reverse osmosis (ULPRO) membranes in comparison with RO membranes consume lower energy and have lower pressure requirement, while present good rejection. Also due to surface chemistry of the ULPRO membranes, water flux of these membranes is higher than RO membranes [27].

It have been reported that the efficiency as well as the mechanism of rejection of organic solutes by membranes are influenced by various parameters such as pressure, feed solution composition, flow rate, membrane-solute interactions and solute, and membrane properties [17,22,28].

Some authors studied BPA removal from aqueous solution by membrane separation. Dong et al. and Wu et al. have used ultrafiltration membrane for BPA removal at very low concentrations [6,16]. In another work, Dong et al. have studied the BPA removal with a hollow fiber MF membrane. They have investigated the effect of different factors such as initial BPA concentration, feed pH, and found that the BPA removal was intensified by adsorption mechanism [29]. It should be note that the number of the articles that studied the removal of BPA with nanofiltration/RO membranes is few, especially with RO membrane. But effects of operating conditions such as pressure, feed concentration, pH, and feed flow rate have not been completely investigated. For example, in a study that conducted by Zhang et al. BPA removal have been studied with a nanofiltration membrane and effect of trans-membrane pressure was investigated. In their study, maximum rejection of 90% has been reported [17]. In another study that conducted by Dudziak and Bodzek, removal of BPA using two NF and one RO membranes without investigation of the effect of operating conditions was examined. The maximum rejection for these membranes was reported as 61, 69, and 67%, respectively [25].

In this work, we focused on the effect of operating parameters on BPA removal by a low-pressure RO membrane. The effects of various parameters such as feed pressure, concentration, pH, and flow rate on BPA removal were investigated and optimum conditions were reported.

2. Materials and methods

2.1. Materials

BPA with 97% purity was purchased from Merck. The main properties of BPA were reported in Table 1. Sodium hydroxide (97% purity), ammonium hydroxide (25% purity) and potassium ferricyanide (99.5% purity), were purchased from Merck. Chloroform and 4-aminoantipyrine were supplied by Dr Mojallali Laboratory Chemicals Co. (99% purity) and Alfa Aesar (97% purity), respectively.

2.2. Methods

2.2.1. Experimental setup

Fig. 1 presents the schematic of the experimental setup. This setup consists of a feed reservoir, diaphragm pump, membrane, membrane module, flow meter, pressure gage, two needle valves, and one diaphragm valve. Cross-flow filtration was used in this work. Feed tank was a glass vessel with capacity of 2 dcm^3. The membrane was a low-pressure polyamide thin film composite RO membrane (TW30-1812-100) that manufactured by Dow Filmtec Company. The specifications of the membrane were shown in Table 2.

Feed flow rate measurement and adjustment were performed by a flow meter (Bailey Fischer & Porter) incorporated with needle valve (AISI 316L manufactured by Fujikin) on the feed line. A second needle valve was used for pressure adjustment. Pressure gauge (MARSH, 0-100 Psig) was mounted after flow meter for monitoring the inlet feed pressure. The pump that was used in this work was a diaphragm pump (HEADON model HF-8367) with maximum

Table 1
The physico-chemical properties of bisphenol-A

Property	Value	Unit	Reference
Formula	$C_{15}H_{16}O_2$	–	[18]
Molecular weight	228	g/mol	[18]
Molar volume	199.50	cm^3/mol	[30]
Melting point @ 25℃	155	℃	[31]
Boiling point @101.3 kPa	398	℃	[32]
Water solubility	120–300	mg/L	[17]
pKa	9.6–10.2	–	[18]
log K_{ow}	3.4	–	[18]
Dipole moment	2.13	Debye	[33]

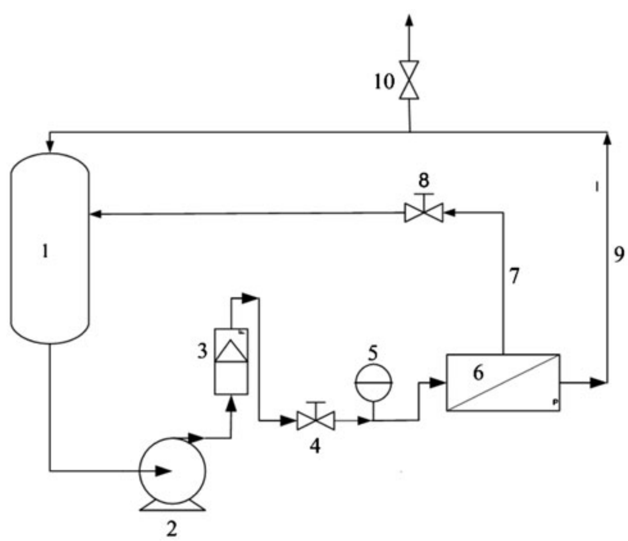

Fig. 1. Schematic diagram of the reverse osmosis set-up. 1-Feed tank, 2-pump, 3-flow meter, 4-flow regulating needle valve, 5-pressure gauge, 6-membrane module, 7-reject line, 8-pressure regulating valve, 9-permeate line and 10-sampling valve.

Table 2
The specification of TW30-1812-100 RO membrane [34]

Specification	Value	Unit
Area	4.8	ft^2
Diameter	1.75	in
Length	10	in
Max. working pressure	300	psi
Max flow rate	7.6	L/min
Max. feed water temperature	45	℃
pH range	2–11	–

pressure 125 psi and 1.2 LPM flow rate. A stainless steel diaphragm valve (Nupro, SS-4DAL) was used for samplings.

2.2.2. Procedure

For feed solution preparation, a stock solution was prepared by dissolving the required amount of BPA in distilled water at pH 8. Because of low solubility of BPA in water in acidic and neutral mediums, the solution was prepared at pH 8. For investigating the influence of feed concentration, four BPA aqueous solutions with concentrations of 30, 50, 70 and 100 mg/L were prepared by diluting the stock solution. Also for investigating the effect of feed pH, solutions with pH of 8, 10 and 11 were prepared by adding sodium hydroxide solution to the solutions. Because, dissolved BPA precipitates at acidic solutions; we choose the alkaline range for pH. All of the experiments were done at room temperature 25 ± 2℃.

The feed solution (with adjusted pH and concentration) was pumped into the membrane module with the desired pressure and flow rate. The rejected and permeated streams were recycled into the feed tank. Sampling from permeate and rejected streams was done until establishing the steady state condition. We found that a maximum one hour recirculation was needed for attaining steady state condition (equilibrium time). For each experiment, the membrane was washed with distillated water for two hours and the streams were analyzed for BPA content. The effect of feed flow rate was investigated at 0.25, 0.44, 0.56, and 1.172 L/min flow rates and effect of feed pressure was investigated at 136–544 kPa.

Solute rejection was calculated as:

$$R = \left(1 - \frac{C_P}{C_F}\right) \times 100 \tag{1}$$

where C_P is the permeate concentration and C_F is the feed concentration [21].

Permeate flux also can be calculated from Eq. (2) as follow:

$$J_p = \frac{Q_p}{S} \tag{2}$$

where Q_p is volumetric permeate flux (m^3/s) and S is the effective area of the membrane (m^2).

According to well-known "Spiegler–Kedem–Katchalsky" model for RO membrane the flux of solvent is as follow:

$$J_w = L_p\left(\frac{dP}{dx} - \sigma\frac{d\Pi}{dx}\right) \tag{3}$$

and for solute flux:

$$J_s = P_s \frac{dC_s}{dx} + (1 - \sigma)C_s J_w \quad (4)$$

where P_s and σ are the solute permeability and reflection coefficient, respectively.

The simplified form of the Spiegler–Kedem–Katchalsky model expresses water and solute fluxes as follow:

$$J_w = L_p(\Delta P - \sigma \Delta \Pi) \quad (5)$$

$$J_s = P_s(C_m - C_p) + (1 - \sigma)C_s J_w \quad (6)$$

in which, ΔP and $\Delta \Pi$ are pressure difference and the osmotic pressure difference across the membrane, respectively. L_p is pure water permeability, C_s is the logarithmic averaged of solute concentration between feed and permeate sides, C_m is the solute concentration at the membrane surface, and C_p is the solute concentration in permeate side [35].

The rejection in this model is as follow:

$$R = \frac{\sigma(1 - F)}{1 - \sigma F} \quad (7)$$

where F can be expressed as follow [35]:

$$F = \exp\left(-\frac{1 - \sigma}{P_s} J_w\right) \quad (8)$$

2.3. Analytical method

The BPA concentration in the feed, permeate, and reject streams was determined by "sensitive 4-amino-antipyrine method" [36]. A visible range spectrophotometer (Cecil, CE1010) at 460 nm was used for determination of the samples concentration.

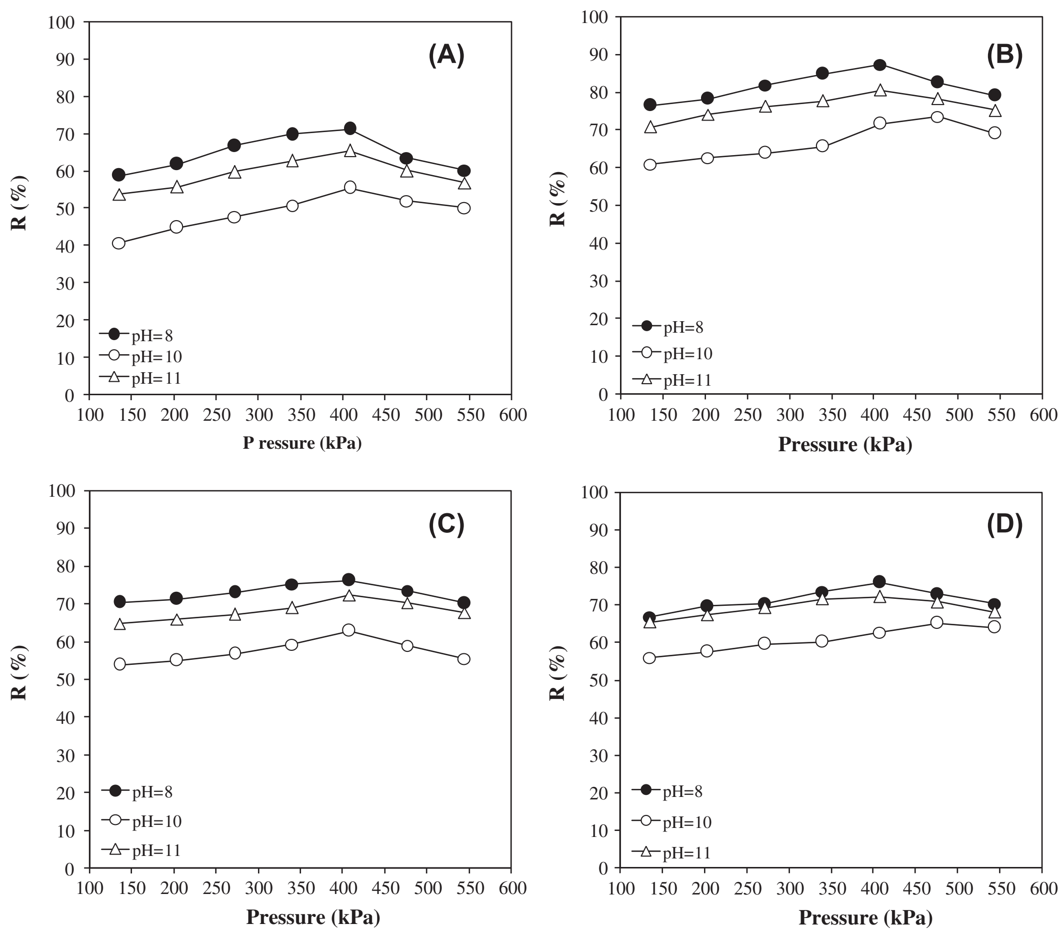

Fig. 2. Effect of feed pressure and pH on BPA rejection percent at 1.172 L/min feed flow rate for different feed concentrations: (A) 30 mg/L, (B) 50 mg/L, (C) 70 mg/L, and (D) 100 mg/L.

3. Results and discussion

3.1. Effect of feed pressure

The effect of feed pressure on BPA rejection in the range of 136–544 kPa was examined. Fig. 2(A–D) show the effect of feed pressure on BPA rejections for various pH and four feed concentrations (30, 50, 70 and 100 mg/L). As shown in this figures, BPA rejection was increased with pressure, but after a critical pressure, the rejection was decreased. This critical pressure was observed to be 408 kPa for the most of experiments expect for two that was 476 kPa. According to the Spiegler–Kedem–Katchalsky model, pressure is the driving force for solvent transport and concentration is the driving force for solute transport. According to this model, the solute flux is lower pressure dependent than water flux [35]. Thus, it is reasonable that the water flux (J_w) is increased directly with pressure. But the solute flux is the result of two terms: solute flux due to concentration difference and solute flux due to water flux. As the water flux is increased by pressure, a concentration polarization layer forms at the membrane-feed interface which increases the real concentration at the membrane surface respect to the feed bulk concentration. By increase in the water flux, the BPA molecules accumulate at the membrane surface (concentration polarization). Concentration polarization increases the osmotic pressure of the system [37]. At low feed pressures, this concentration is not too important and could be neglected. Higher feed pressures intensify the polarization effect and consequently the solute concentration at the membrane–feed interface is increased so that the effect of increasing osmotic pressure cannot be neglected. This point shows the critical pressure in which the competition between solvent flux and solute concentration difference for solute transport is changed from pressure to concentration effect. It was observed that at working pressures less than critical pressure, the concentration polarization was not considerable and the feed flow rate was sufficient to overcome the concentration polarization effects. At feed

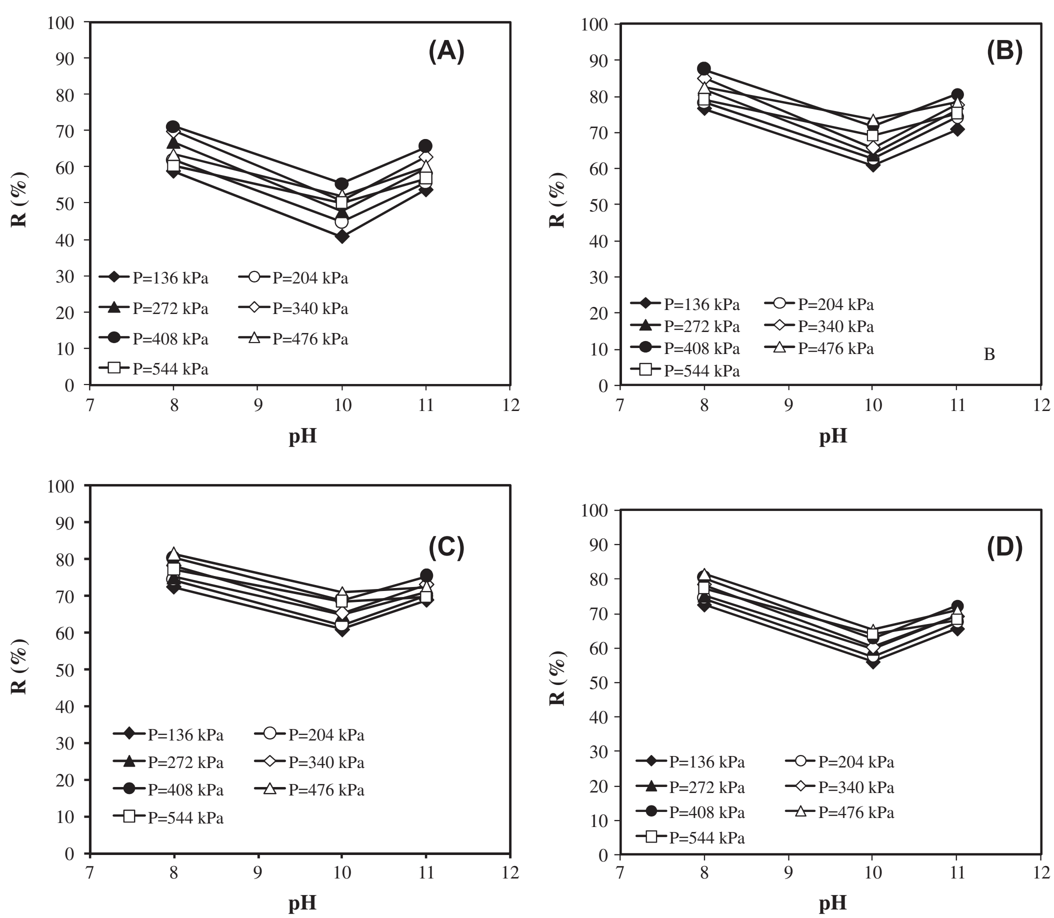

Fig. 3. Effect of feed pH and pressure on BPA rejection percent at 1.172 L/min feed flow rate for different feed concentrations: (A) 30 mg/L, (B) 50 mg/L, (C) 70 mg/L, and (D) 100 mg/L.

pressures higher than this critical pressure, the osmotic pressure reduces the effective pressure driving force for solvent transport. Then the solvent transport decreases while the solute passage increases through the membrane. But at critical, pressure the concentration polarization was developed so that the feed flow rate was not sufficient to remove the polarization layer and then the rejection was decreased. According to Eq. (8) at constant feed pressure, by decrease in water flux across the membrane which is the resulted of sever concentration polarization, *F* value is increased and according to Eq. (7), this reduces the rejection coefficient.

3.2. Effect of feed pH

The effect of feed pH on BPA rejection was shown in Fig. 3(A–D). In these figures, BPA rejection has been reported at different feed pressures and concentrations. The effect of feed pH at range of 8–11 was investigated. The results showed the maximum rejection at pH 8 and minimum rejection at pH 10. This behavior was observed at all of feed concentrations. Polyamide membranes at alkaline solutions is hydrolyzed from free carboxylic acid groups, so the membrane ionization is occurred at pH values above 7 and makes the membrane surface negatively charged [35]. At pH 8, BPA molecule is ionized to bisphenolate with one negative charge. The existence of OH group in one side of the non-ionized molecules and O^- in the ionized molecules resulted an attraction between O^- of one molecule and OH of the another molecule. This attraction decreases the effective negative charge of the ionized molecules which results lower rejections for pH higher than 8. Also, interaction between ionized membrane and bisphenolate was associated to increase of the BPA rejection. By increasing the feed pH higher than 10, the second OH group of BPA is dissociated and bisphenolate ion with two negative charges is formed which increases both the repulsion between the ionized BPA molecules and membrane surface which consequently increases the rejection by RO membrane. Such a high pH values (higher than 10) for operation with these RO membranes was not

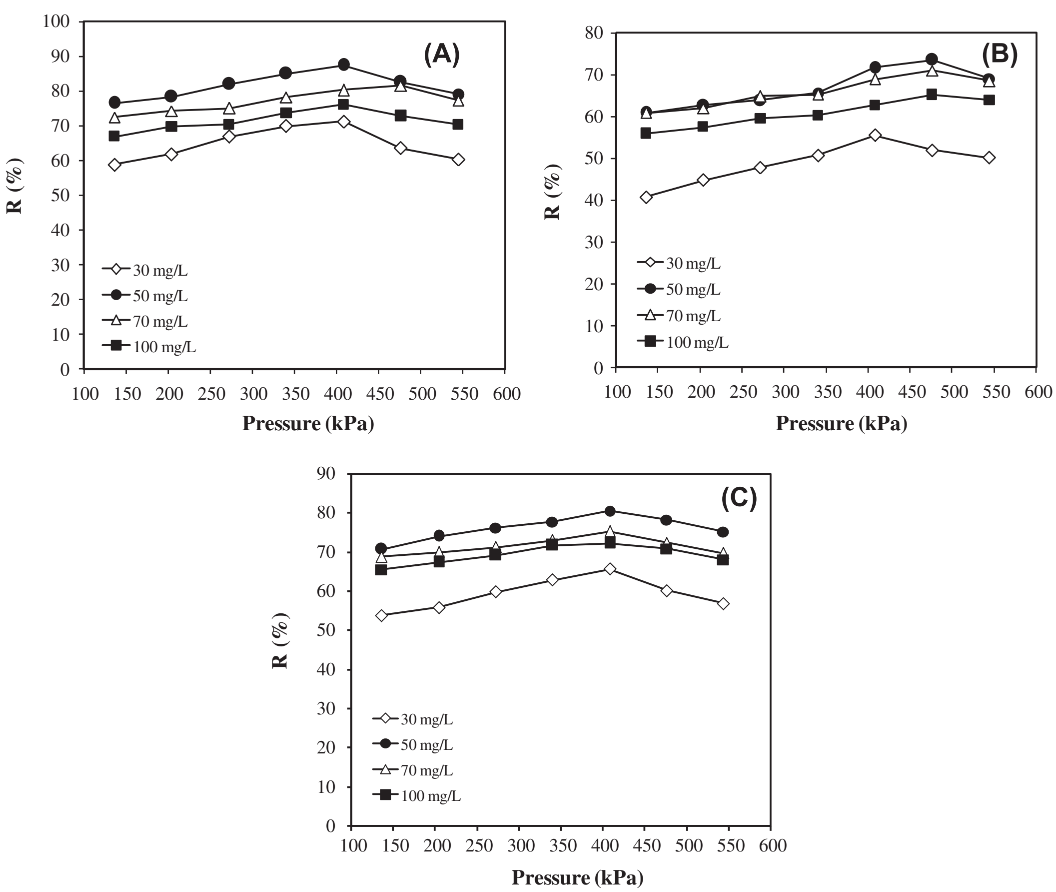

Fig. 4. Effect of feed pressure and concentration on BPA rejection percent at 1.172 L/min feed flow rate for different feed pH value: (A) pH 8, (B) pH 10, and (C) pH 11.

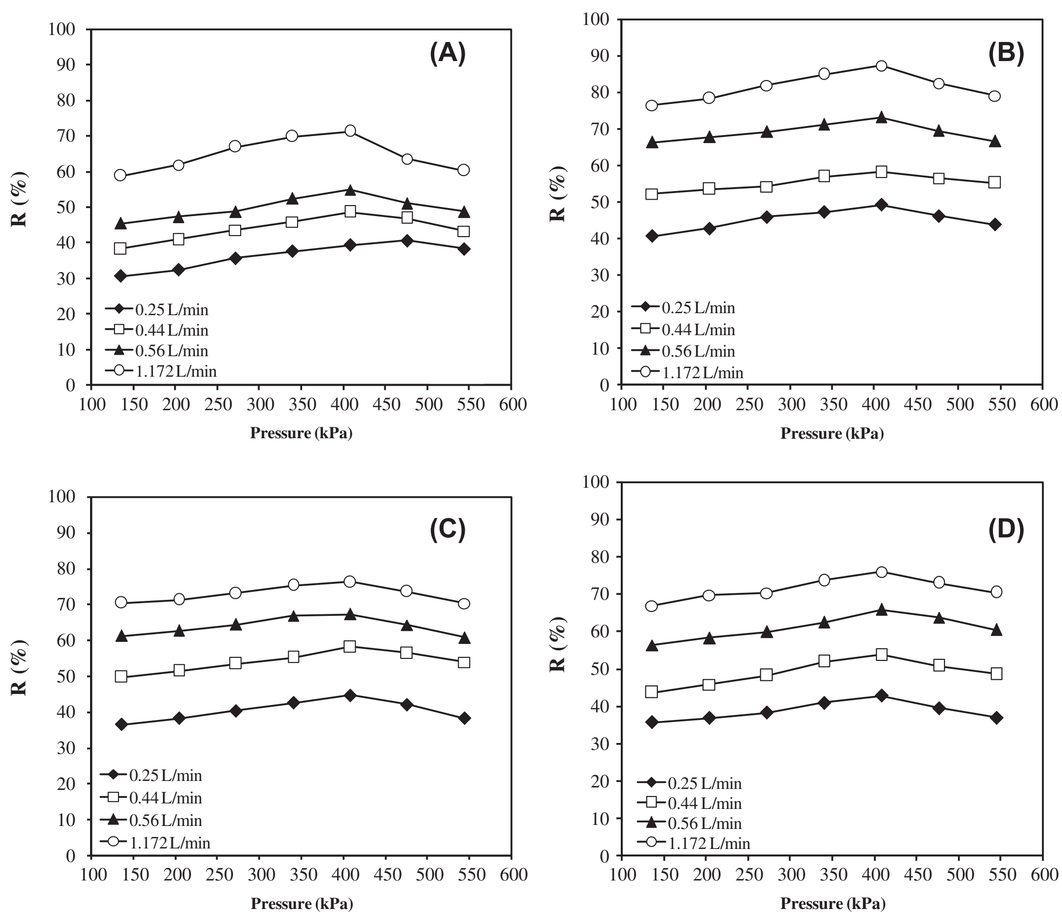

Fig. 5. Effect of feed pressure and flow rate on BPA rejection percent at pH 8 for different feed concentrations: (A) 30 mg/L, (B) 50 mg/L, (C) 70 mg/L, and (D) 100 mg/L.

recommended because of hydrolysis of the polymeric membrane.

3.3. Effect of feed concentration

The effect of feed concentration on BPA rejection was shown in Fig. 4(A–C). At maximum feed flow rate, room temperature, and different pH, this effect was shown. By increase in BPA concentration, the rejection was decreased which is due to higher concentration polarization. In RO system, increase of feed concentration increases the osmotic pressure and thus decreases the driving force that can decrease the water flux and also according to Eq. (5) with increase of osmotic pressure, water flux decreases. Increase in feed concentration increases the solute concentration at the membrane surface and refer to Eq. (6), the solute flux increases along with decrease in water permeation flux, decreases the rejection percent for BPA. By increase of feed concentration, the solute accumulation and concentration polarization are increased that can decrease the solute rejection. The results show that at 50 and 30 mg/L maximum and minimum BPA rejection, respectively, were obtained. In low concentrations, osmotic pressure difference is low, so according to Eq. (5), water flux is high. Also at low concentrations, solute concentration on the membrane surface is low and solute flux is also low but at concentration of 30 mg/L, the water flux is so high that can carry the dissolved BPA in the membrane surface toward the permeate side. So at 30 mg/L the rejection was low.

3.4. Effect of feed flow rate

The effect of feed flow rate on BPA rejection at pH 8 was shown in Fig. 5(A–D). As its clear, by increasing the flow rate, the rejection is increased. At 1.172 L/min feed flow rate (maximum feed flow rate); the maximum rejection was obtained for all of the concentrations. This effect is due to concentration polarization effect. By increasing the feed flow rate, the concentration polarization layer thickness decreases and therefore the effective osmotic pressure decreases. By decrease of osmotic pressure difference according to Eq. (5), water flux increases and according to Eqs. (7) and (8) as was discussed earlier, the rejection of BPA is increased. As it is clear, at low feed flow rates, concentration polarization is high and rejection is reduced considerably. The maximum rejection was observed at 1.172 L/min feed flow rate and 50 mg/L feed concentration.

4. Conclusion

An effective method for BPA separation from aqueous solution by a low-pressure RO membrane was introduced. The effect of various parameters such as feed flow rate, feed pH, concentration, and pressure was investigated. The results showed that:

(1) There is a critical pressure for rejection. In most cases, this critical pressure was observed at 408 kPa and in a few cases it was 476 kPa. The rejection for pressures lower than this critical pressure increased by pressure and after that, it was decreased.
(2) By increasing pH from 8 to 10, BPA rejection was decreased, because at pH 8 most of the BPA molecules ionized and interaction between them caused higher rejection. But from pH 10 to 11, rejection was increased.
(3) By increasing feed concentration from 30 to 50 mg/L, BPA rejection was increased and for higher concentration the rejection was decreased. This observation showed the effect of concentration polarization on reduction of BPA rejection. Because at higher feed concentrations, the effect of concentration polarization was more considerable and thus permeation and consequently rejection was decreased.
(4) Increase in feed flow rate increases rejection percent due to lowering the adverse effect of concentration polarization.
(5) A maximum rejection of 87% was obtained at 50 mg/L feed concentration.
(6) This low-pressure RO system could be considered as an effective method for BPA removal from waste waters.

Symbols

BPA — bisphenol A
C_F — solute concentration in feed solution (kg/m^3)
C_m — solute concentration at the membrane surface (kg/m^3)
C_P — solute concentration in permeate stream (kg/m^3)
C_s — the logarithmic averaged of solute concentration between feed and permeate sides (kg/m^3)
J_w — water flux (kg/m^{2}s)
J_p — permeate flux (m/s)
J_s — solute flux (kg/m^{2}s)
L_p — pure water permeability (m/s)
P_s — solute permeability (m/s)
Q_p — volumetric permeate flux (m^3/s)
R — rejection (dimensionless)
S — effective area of the membrane (m^2)
ΔP — pressure difference across the membrane (Pa)
$\Delta\Pi$ — osmotic pressure difference of the solute across the membrane (Pa)
σ — reflection coefficient of the solute from membrane surface.

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