

Influence of organic fouling and operating conditions on nanofiltration membranes to reduce phenol concentration in natural waters

N. García-Vaquero Marín and J. A. López-Ramírez

ABSTRACT

Phenol is a product widely used in manufacturing and as a disinfectant for industrial and medical applications. The treatment of aqueous effluents polluted with phenol and phenolic species has deserved much attention in the last years due to the toxicity and low biodegradability of those organic compounds. Nanofiltration (NF) has a great potential to remove these contaminants. The main objective of this work is to study the influence on the rejection of phenol by a NF90 2540 NF membrane of various operating conditions: pressure (0.3, 0.4 and 0.5×10^6 N/m²); phenol feed concentration (from 0.025 kg/m³ up to 0.150 kg/m³) and pH (from 5.9 to 11.5). The influence of organic fouling is also investigated. Results showed that when different phenol concentrations were compared at the same pressure, permeate fluxes decreased as phenol concentrations increased (from 20% for the highest phenol feed concentration tested to 10% for the lowest); when phenol concentration increased, phenol was probably adsorbed onto the membrane, which led to lower rejection values (from 78 to 59% rejection). Changes in the pH of the solution altered the phenol dissociation equilibrium and membrane rejection characteristics. The reduction in phenol rejection by organic fouling (maximum of 35%) was a consequence of the materials deposited onto the membrane, which led to the formation of a cake layer.

Key words | membranes, nanofiltration, natural waters, phenol, rejection

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INTRODUCTION

Membrane processes have been applied to water production for industrial applications and have more recently been applied to drinking water treatment. Membrane processes such as nanofiltration (NF) and reverse osmosis (RO) are being considered as an indispensable part of the treatment chain for high-quality water production from impaired sources including reclaimed wastewater and contaminated surface water (Bellona *et al.* 2004; Nghiem & Schäfer 2005; Yoon & Lueptow 2005; Xu & Drewes 2006; Bellona & Drewes 2007). The applicability of NF and RO membranes to the retention of low molecular weight organic compounds has been described in several publications (Verliefde *et al.* 2007; Comerton *et al.* 2009).

NF is an example of a pressure driven membrane filtration. It has been increasingly considered as a reliable and affordable technique for the production of high quality water from a variety of sources. In water treatment, NF membranes allow for the removal of small dissolved organic molecules (i.e., pesticides, endocrine disruptors, disinfection by-product precursors) and partial transmission of salts through the membranes. Despite the advantages of membrane processes, fouling remains a problematic and inevitable issue encountered in all membrane filtration systems. The term 'fouling' is used to describe the undesirable formation of deposits onto membrane surfaces. This occurs when rejected particles are not transported from the surface of the membrane back to the bulk solution.

Fouling is also described as flux decline, which can only be removed by chemical cleaning. Depending on the feed water, a significant flux decline can be observed. This flux decline has a number of components: (i) concentration polarisation, (ii) reversible fouling with chemical cleanings and (iii) irreversible fouling. Concentration polarisation is the accumulation of retained solutes in the membrane boundary layer. It creates a high solute concentration on the membrane surface compared to that in the bulk solution, which causes a flux reduction. This reduction is predominantly due to the increased osmotic pressure of the retained ions and the formation of gels by the retained organic molecules.

The presence of a fouling layer can drastically alter the surface characteristics of the membrane such as the surface charge and hydrophobicity. Several studies (Xu *et al.* 2006; Nghiem *et al.* 2008) demonstrate the complexity of the mechanisms that lead to the different effects of fouling on the retention of micro pollutants; elucidation of these mechanisms is greatly needed. It is noteworthy that little is known about the influence of membrane fouling on the retention of trace organic contaminants. In fact, membrane fouling is an inherent phenomenon of full-scale operation using NF/RO membranes due to the presence of substantial amounts of organic macromolecules and colloidal matter in the feed water. The fouling layer can considerably alter the separation behaviour of trace contaminants, resulting in either an increase or a decrease in retention as compared to a clean membrane (or no fouling) condition (Plakas *et al.* 2006; Xu *et al.* 2006; Agenson & Urase 2007). It has also been demonstrated that membrane pore size plays an important role in governing the effects of fouling on the rejection of hydrophilic pharmaceuticals (Nghiem & Hawkes 2007). Nevertheless, the impact of membrane fouling on other important rejection mechanisms, such as hydrophobic adsorption and electrostatic repulsion, remains poorly understood.

While phenols are naturally occurring chemicals, the majority of phenols present in the environment have an anthropogenic origin. Phenols are organic compounds and are a major chemical intermediate in the manufacture of phenolic resins, synthetic fibres, plastics and related materials. Municipal and industrial wastewater treatment plants and untreated direct disposals are the main sources

for the presence of phenols in natural waters. Due to their toxicity and low biodegradability, these aromatic hydrocarbons are of environmental concern. Excessive exposure to phenol and its products may cause a variety of chemical-dependent health effects. Phenolic contamination of natural waters is a cause for growing concern in the fishing industry and in drinking water treatment.

Previous studies about the rejection of organic compounds by NF and RO membranes have reported that the removal efficiency depends on many parameters: operational conditions, steric hindrance, adsorption, hydrophobic and electrostatic interactions, solution effects on the membrane and the solute/membrane properties (Lin *et al.* 2002; Bellona *et al.* 2004; Nghiem *et al.* 2006; Van der Bruggen *et al.* 2006; Arsuaga *et al.* 2008; Bódalo *et al.* 2009; López-Muñoz *et al.* 2009; Verliefde *et al.* 2009). An understanding of how those factors affect the permeation of specific solutes in high-pressure membranes systems is essential for achieving their selective rejection. On this basis, the scope of this study is to investigate the effect of varying the operating conditions (pressure, phenol concentration and pH and) and the influence of organic fouling on the rejection of phenol by an NF membrane (NF-90).

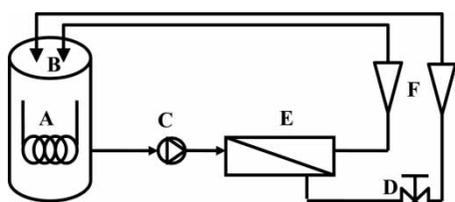
METHODS

The experiments were performed using an NF90-2540 NF membrane, manufactured by Film-Tec Dow Chemical Company (USA), in a bench-scale pilot plant. In Table 1, some of the main physical-chemical properties of the membrane are shown (provided by the manufacturer). This is a spiral wound membrane with an active area of 2.6 m².

The pilot plant consisted of a feed tank, a membrane module (2540), a high-pressure pump and two flow meters (one for the permeate and another for the concentrate). The storage tank had a capacity of 50 L. Permeate and concentrate were recycled back to the feed reservoir to work in a closed-loop circuit (Figure 1) The feed reservoir temperature was kept constant at 21 ± 1 °C by a heat exchanger (Refrigerated Recirculating Chillers V12 °C, VWR).

Table 1 | NF-90 membrane characteristics

Material	Polyamide
MWCO (Da)	200
NaCl retention (%)	85
Zeta potential at neutral pH (mV)	-17.5
Maximum temperature (°C)	45
Maximum pressure (bar)	41
pH range	4–11
Average pore diameter (nm)	0.68
Surface roughness (nm)	69.9
Contact angle (°)	42.5

**Figure 1** | Schematic Flow diagram of the NF test unit: (A) heat exchanger, (B) feed tank, (C) high pressure pump, (D) valve, (E) membrane module, (F) flow meters.

Effect of operating conditions

Several experiments were conducted to study membrane phenol rejection according to the different operating conditions of pressure, concentration and pH. At the beginning of each experiment, the feed solution, containing the predetermined concentration of phenol, was circulated through the pilot plant for 30 minutes to stabilise the membrane. The feed concentration varied from 0.025 to 0.150 kg/m³ of phenol. The synthesis grade phenol (Table 2) was purchased from Scharlab S.L. (Barcelona, Spain).

Table 2 | Properties of phenol

Formula	MW (g/mol)	Diameter (Å)	pKa	log P	Compound structure
C ₆ H ₅ OH	94.11	2.66	9.95	1.45	

pKa: acid dissociation constant.

log P: partition coefficient.

The feed solutions were made by dissolving the appropriate concentration of phenol in tap water (Table 3).

To study the influence of pressure, experiments for each phenol feed concentration were conducted at 0.3, 0.4 and 0.5 × 10⁶ N/m² (Pa = N/m²). These pressures were selected as results in previous membranes characterization work showed that the membranes worked successfully within this range of pressures with drinking water, and above it there was no significant difference between the results. The influence of increasing the concentration at a constant pressure was also studied.

With the objective of analysing the influence of pH on phenol rejection, several experiments were done in a pH range from 5.9 up to 11.5, while keeping the phenol concentration (0.050 kg/m³) and pressure (0.4 × 10⁶ N/m²) constant. The pH was fixed by the addition of analytical grade sodium hydroxide or hydrochloric acid aqueous solutions.

Effect of organic fouling

To study the influence of organic fouling on the phenol rejection membrane capacity, an organic model foulant, proteose peptone n.3, was used. This product includes fats, metals, salts, vitamins and many other biological compounds. This foulant was chosen due to its complete composition, which simulates the ubiquitous organic matter present in natural waters. A high flux was maintained for a fast membrane fouling.

Phenol concentrations were determined spectrophotometrically at 270 nm with a prior addition of NaOH 1M (Bódalo *et al.* 2009). Absorbance values were transformed to phenol concentration in the sample using a calibration curve (Abs 270 nm = 5.6698 [phenol] – 0.0083 R² = 0.999).

Table 3 | Feed water quality conditions (n = 2)

Feed solution	Conductivity (μS/cm)	DOC (mg/l)
Tap water	531 ± 1.4	2.57 ± 0.05
Tap water + 0.025 kg/m ³ phenol	533 ± 0.0	22.16 ± 0.77
Tap water + 0.050 kg/m ³ phenol	533 ± 0.0	41.38 ± 0.52
Tap water + 0.100 kg/m ³ phenol	533 ± 0.0	81.18 ± 0.89
Tap water + 0.150 kg/m ³ phenol	533 ± 0.7	118.55 ± 0.49

For dilute aqueous mixtures consisting of water and a solute, the selectivity of the membrane towards the mixture was expressed in terms of the solute rejection coefficient (Lin *et al.* 2002). The retention of a membrane, R , is defined as:

$$R = (1 - C_p/C_f) \times 100 \quad (1)$$

where C_p and C_f are permeate and feed phenol concentrations (kg/m^3), respectively.

The permeate flux (J_p) was calculated according to the following equation:

$$J_p = Q_p/A \quad (2)$$

where Q_p is the permeate flow (m^3/h) and A is the membrane active area (m^2).

RESULTS AND DISCUSSION

Influence of pressure and phenol feed concentration

Six tests were conducted (performed in duplicate) to observe how the pressure and the phenol feed concentration affect phenol membrane rejection. Three tests were done varying the pressure for a constant feed concentration, and, another three tests were done varying the phenol feed concentration at a constant pressure. Figures 2(a) and (b) respectively show the average permeate flux and rejection percentage with respect to pressure.

In Figure 2(a), the direct relationship between permeate flux and pressure is displayed. It is observed that, in all cases, the flux increases when pressure increases. This observation was predicted by the solution-diffusion model (Wijmans & Baker 1995; Wijmans 2003), which is used to estimate membrane performance characteristics. According to Equation (3) derived from this model, when the phenol concentration is constant, the water flux through the NF membrane depends exclusively on the applied pressure because the osmotic pressure remains constant:

$$J_i = A(\Delta p - \Delta \pi) \quad (3)$$

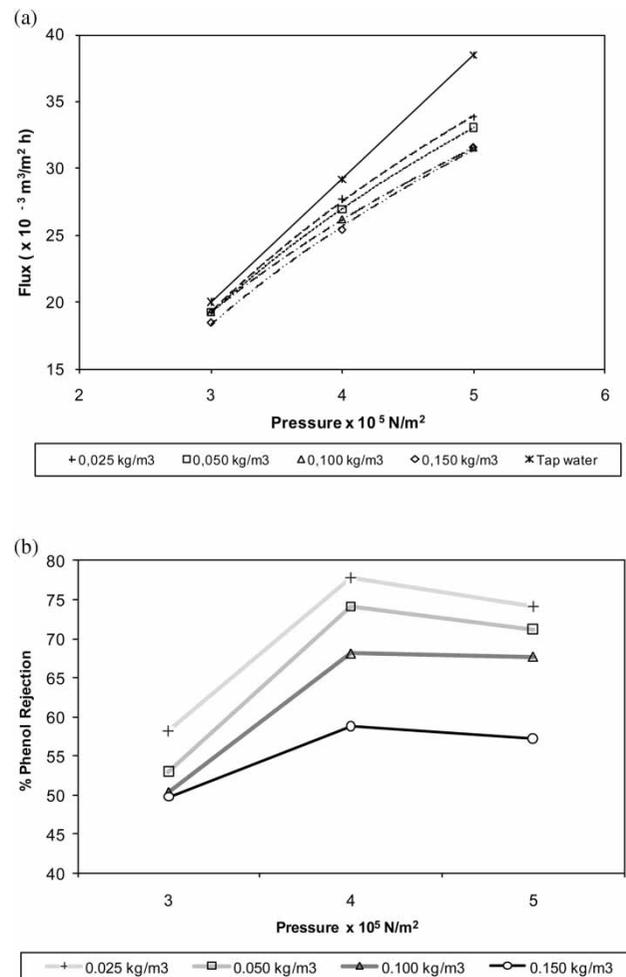


Figure 2 | (a) Effects of pressure ($\text{Pa} = \text{N}/\text{m}^2$) on permeate flux for different phenol concentrations ($n = 2$). (b) Effects of pressure on phenol rejection for different phenol concentrations ($n = 2$).

where J_i is the water flux, A is the water permeability constant, Δp is the difference in hydrostatic pressure across the membrane and $\Delta \pi$ is the difference in osmotic pressure across the semi-permeable membrane.

Figure 2(a) shows that, when different phenol concentrations are compared at the same pressure, flux decreases as concentration increases. For example, at $0.5 \times 10^6 \text{ N}/\text{m}^2$ the permeate flux declined by nearly 20% from the tap water at the highest phenol feed concentration tested; while it just decreased by 10% at the lowest one. At the same time that phenol concentration increases, osmotic pressure also increases, and, according to the solution-diffusion model, flux decreases slightly. In the case of tap water, the osmotic pressure does not vary at any time

because tap water concentration remains constant, so the tap water flux is always higher than that in any experiment with phenol.

Flux decrease at higher phenol concentrations may also happen as a result of organic solute adsorption on the membrane surface. Adsorption of organic molecules from an aqueous solution onto the membrane surface or inside the membrane pores has a large influence on the water flux. The more the given component is adsorbed on the membrane, the larger the decline in flux. Molecules with a molecular weight around 100 Da (in this case phenol) have the highest tendency to adsorb onto the membrane and, thus, have a higher fouling tendency (Van der Bruggen *et al.* 2002).

Although according to the solution-diffusion model the retention of organic compounds should steadily increase with applied pressure, all the present results show that the major rejection occurs at $0.4 \times 10^6 \text{ N/m}^2$ ($\text{Pa} = \text{N/m}^2$) (Figure 2(b)). This is the case, for example, of the phenol rejection percentages for the lowest concentration: 58% at $0.3 \times 10^6 \text{ N/m}^2$, 78% at $0.4 \times 10^6 \text{ N/m}^2$ and 74% at $0.5 \times 10^6 \text{ N/m}^2$. The trend in phenol rejection can be attributed to the molecular characteristics of phenol such as acidity and solubility. These two characteristics can lead to the deformation of the active pore layer when phenol is adsorbed onto the membrane surface (Bódalo *et al.* 2009). As the phenol concentration increases, so does the adsorption onto the membrane, which results in a decline of the rejection (78–74–68–59% in ascending order of phenol concentration) (Figure 3).

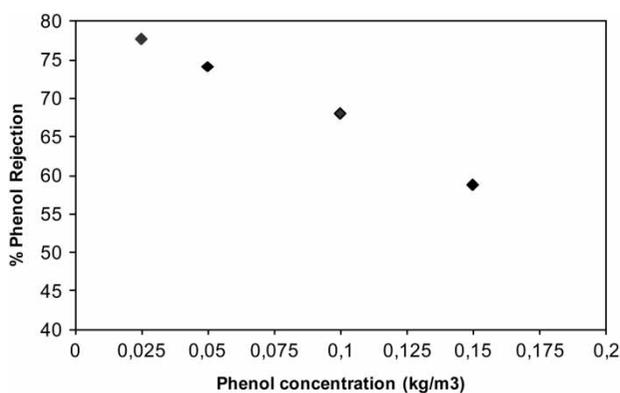


Figure 3 | Effect of phenol concentration on rejection at a constant pressure of $0.4 \times 10^6 \text{ N/m}^2$.

Influence of feed pH

To determine how feed pH affects the removal of phenol, several experiments were conducted (performed in duplicate) while maintaining constant pressure and phenol concentration. The only variable was the feed pH, which rose from 5.9 to 11.5. During the experiments, the pH was measured at the beginning, during and at the end of them to make sure it was kept constant. The pressure was fixed at $0.4 \times 10^6 \text{ N/m}^2$ because the best results for phenol removal were obtained at this pressure.

Changes in the feed pH alter the dissociation equilibrium of the acidic organic solutes and membrane functional groups. As a result, both the electrostatic repulsion between phenol and the membrane and the steric interaction inside pore walls were altered.

Figure 4(a) shows that higher rejections are reached under alkaline conditions, so as the pH increases, so does the rejection of phenol. When the feed pH becomes more

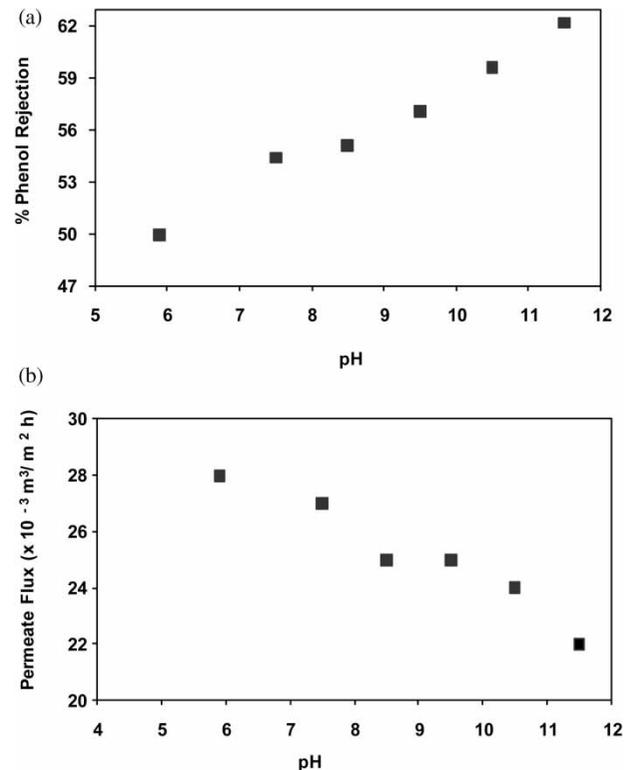


Figure 4 | (a) Effect of feed solution pH on phenol rejection at a constant pressure of $0.4 \times 10^6 \text{ N/m}^2$ and a phenol concentration of 0.050 kg/m^3 ($n = 2$). (b) Effect of feed solution pH on permeate flux at a constant pressure of $0.4 \times 10^6 \text{ N/m}^2$ and a phenol concentration of 0.050 kg/m^3 ($n = 2$).

alkaline, the amount of dissociated phenolic species increases. As inferred from its zeta potential, the NF90 membrane has an isoelectric point at a pH of 3.5 (López-Muñoz *et al.* 2009). Thus, the membrane becomes more negative when pH increases until 6.0; however, the changes are less pronounced in the pH range of 6 to 12 (Boussu *et al.* 2006; Park & Jaeweon 2008). As a consequence of both the increase of dissociated phenolic species in the feed solution and the negatively charged surface membrane, the electrostatic repulsion between the membrane and the negatively charged solutes is strengthened (Boussu *et al.* 2006), thus, enhancing the rejection of phenol. At pH higher than ~ 9.5 , the rejection of phenol is even higher due to its deprotonation ($pK_a = 9.95$), which leads to the formation of the negatively charged phenolate.

Figure 4(b) shows the results of the analysis of the influence of feed pH on the permeate flux at a constant pressure. Throughout the experiments, the flux decreased when the pH increased. As López-Muñoz *et al.* (2009) stated, this might be due to the negative zeta potential that, when increased, promotes a stronger electrostatic interaction between dissociated functional groups of the membrane material and causes pore constriction.

These changes in the structure of the membrane are in agreement with the observed variations in phenol rejection within this pH range. Nevertheless, the high phenol rejection for $pH \geq 10$ should be attributed to the electrostatic repulsion between negative charged membrane and dissociated solute species (phenolate).

Influence of organic fouling on phenol removal

Organics interact with membranes in several ways, which makes it difficult to isolate individual interaction mechanisms. These interactions are strongly dependent on the type of organic, its chemical characteristics and its affinity for the membrane material. Adsorption plays an important role in the fouling of NF membranes by organic compounds. Adsorption is an equilibrium-driven process between the boundary layer concentration, which is determined by the concentration polarisation, and the adsorbed organics (Nghiem & Schäfer 2005).

To cause a fast membrane fouling, peptone was developed at an initial permeate flux of $0.036 \text{ m}^3/\text{h m}^2$. The flux

decline rate slowed down considerably and a quasi-steady-state fouling layer was fully developed after approximately 20 h of filtration (Figure 5).

Then, the permeate flux was readjusted to a lower value of $0.023 \text{ m}^3/\text{h m}^2$ and the phenol solution was added to the feed tank. Feed and permeate samples were taken for analysis at specified time intervals.

For the model organic foulant used in this study, initial foulant–membrane interactions appear to have a much greater effect on flux decline than foulant–foulant interactions at a later stage of the fouling development process. These two distinct fouling stages were consistent with previous studies (Yuan & Zydney 2000; Lee & Elimelech 2006; Li *et al.* 2007; Nghiem & Coleman 2008). Figure 5 shows how a rapid initial flux decline occurred immediately after the peptone was introduced to the feed solution followed by a more gradual flux decrease. Due to the initial adsorption of organic foulants onto the membrane, rapid flux decline was possibly caused by pore blocking or pore constriction of the membrane surface. In the second fouling stage, the flux decline rate slowed down considerably and a quasi-steady-state fouling layer was fully developed. Flux decline in this stage can be attributed to the compaction and thickening of the fouling cake layer, which induce an additional cake resistance.

Figure 6(a) illustrates the results of phenol rejection by both a fouled and a clean membrane. Reduction in the phenol rejection was observed for all experiments with the fouled membrane. This is probably due to the small pore size of this membrane that is speculated to be free of pore blocking, in which the fouling is limited to foulant–membrane surface interaction (Nghiem *et al.* 2008). As

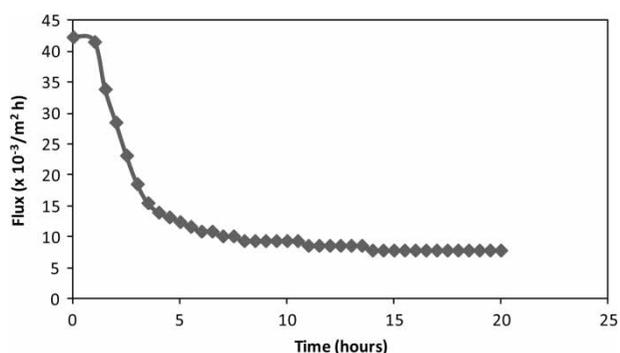


Figure 5 | Permeate flux decline while fouling takes place.

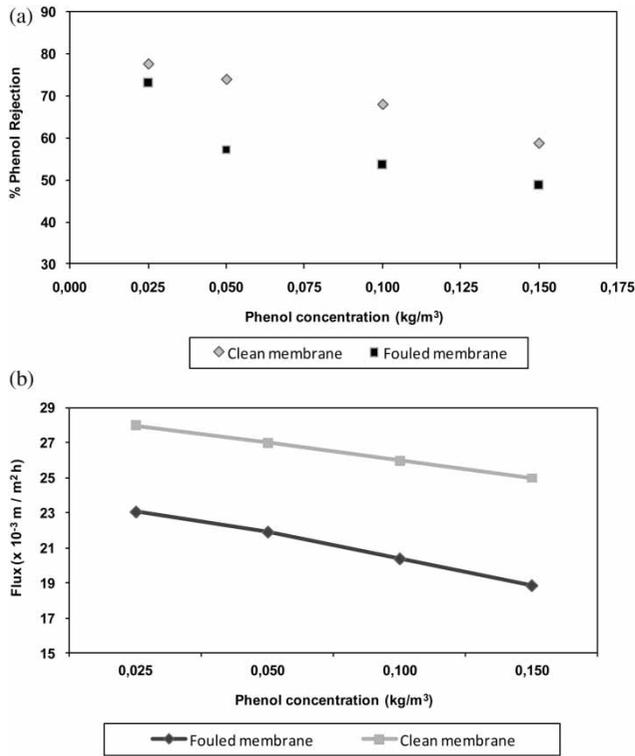


Figure 6 | (a) Effect of organic fouling on phenol removal ($n = 2$). (b) Flux decline in a clean membrane and in an organic fouled membrane ($n = 2$).

peptone has both hydrophilic and hydrophobic components, the phenol concentration is spread among the permeate, the membrane surface and peptone. Although a repulsion force between the membrane and peptone components should be developed to avoid fouling, a higher convective force towards the membrane allows the permeate to penetrate the membrane and leave the solute back on the membrane. As a result, both the concentration polarisation and the accumulation of components onto the membrane increase. As Figure 6(a) shows, these phenomena lead to the deposition of materials on the membrane in a cake layer that reduces the rejection of phenol, and also reduce the permeate flux (Figure 6(b)).

CONCLUSIONS

In this study the following conclusions can be drawn:

1. In all cases permeate flux was raised by increasing the pressure, which confirmed the solution-diffusion model.

2. When the phenol concentration in feed solution increased, the osmotic pressure also increased; according to the solution-diffusion model, this would slightly decrease the flux.
3. Changes in the pH of the solution altered the dissociation equilibrium of phenol and the membrane characteristics. An increase of phenol rejection is observed when the solution pH is increased.
4. Phenol rejection by the fouled membrane was lower due to the cake layer of peptone materials on the membrane.

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