

Characterization and retention of NF membranes using PEG, HS and polyelectrolytes

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Abstract

Nanofiltration (NF) membranes retention depends on charge repulsion and size exclusion, combining the properties of ultrafiltration and reverse osmosis membranes. The molecular weight cut-off of two NF membranes was determined using cross-flow filtration of different molecular weights of non-ionized polyethylene glycol (PEG). Their retention of humic acid (HA), fulvic acid (FA), diallyldimethylammonium chloride and copolymer of dimethyl aminoethyl acrylate was also determined. NF270 membrane's pure water flux was higher than NF90. NF270 also produced lower retention compared to NF90. Observed retention of PEG using NF270 was lower than NF90, but real retention of PEG was higher for NF270 than NF90. This unexpected result is due to the inconsistency of the concentration polarization model. According to the concentration polarization model, real retention determination is dependent on permeate flux (J_p) instead of taking account of membrane flux reduction. The two membranes are supposed to operate at similar flux to overcome this inconsistency. Incomplete retention of poly diallyldimethylammonium chloride (PDADMAC) and copolymer of dimethyl aminoethyl acrylate (CoAA) is due to the presence of their monomers. HA retention is higher than FA because of its higher molecular weight range. NF270 membrane retention of HA and FA is lower than NF90 because of its higher pore size and porosity. Solute retention is constant with increasing pressure due to the competition between concentration polarization and dilution effects at the studied range of trans-membrane pressure (TMP).

Keywords: Nanofiltration; Characterization; Humic substances; Polyelectrolytes; Cross-flow filtration

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1. Introduction

In the seventies, modified reverse osmosis (RO) membranes were developed. These membranes were distinguished from common RO membranes by their higher water flux production at relatively low pressures. Consequently, these membranes were named Nanofiltration (NF) membranes [1]. NF membranes first applications were reported in the second half of the eighties [2]. NF combines size exclusion removal of uncharged and charged solutes as known from UF membranes and charge effect removal experienced by RO membranes [3]. Comparison of retention performance between UF and NF membranes can be accomplished by filtering uncharged solutes. Membranes performance is compared by the membranes nominal molecular weight cut-off (MWCO), which is usually defined as the smallest molecular weight species for which the membrane has more than 90% rejection [4]. Filtration of uncharged solutes at low concentrations are usually used to determine the MWCO of membranes.

Non-ionized polyethylene glycols (PEGs) are used to characterize membranes in UF, NF and modified membranes characterization studies [5–9]. PEGs are used extensively in membrane characterization because they are water soluble and can be readily obtained with narrow molecular weight distributions. In addition, their adsorption is very low for almost every polymer surface [10]. Nanofiltration (NF) membranes MWCO is usually in the range of 0.3–1 kDa and operate at relatively low pressure in the range of 3–15 bars [11].

This paper studies the characterization and retention of NF membranes using PEG, HS and

polyelectrolytes, which is in conjunction with a previous paper that dealt with characterization of UF using PEG [12]. The purpose of this study is to determine the real and observed MWCO of two NF membranes using different molecular weights of non-ionic PEG solutions using flat sheet cross-flow configuration. HA, FA, poly diallyldimethylammonium chloride (PDADMAC) and copolymer of dimethyl aminoethyl acrylate (CoAA) permeability is also studied at varying trans-membrane pressure.

2. Material and methods

2.1. Material

The de-ionized water used for preparation the studied solutions was obtained through demineralization using ion exchange followed by reverse osmosis. The conductivity of the water was lower than 1 $\mu\text{S}/\text{cm}$. All the chemicals used in the experiments were reagent grade. All glasswares used in the experiments were rinsed with de-ionized water and dried at 70°C overnight.

NF membranes were kindly supplied by FILMTEC and manufactured by DOW chemical company (USA) (Table 1). These two membranes are made from polyamide. They were provided in flat sheet form and were cut to the desired dimensions in the laboratory. The membranes were immersed in de-ionized water for a minimum of 4 h before being used. The water was changed every hour during the first 4 h of immersing.

High purity 0.3, 2, 4 and 6 kDa molecular weight PEG solid pellets and PDADMAC were purchased from Sigma Aldrich. The cationic

Table 1
Properties and characterization of NF membranes as specified by manufacturer

Membrane	Polymer	CaCl ₂ retention (%) (product water flow rate, m ³ /d)	MgSO ₄ retention (%) (product water flow rate, m ³ /d)
NF90	Polyamide	85–95 (28.4)	>97 (36.0)
NF270	Polyamide	40–60 (55.6)	>97 (47.3)

PDADMAC was supplied as viscous liquid of 20% dissolved in water and have an average MW of 200–350 kDa. The cationic PDADMAC was supplied as viscous liquid of 20% dissolved in water and is a monomer of diallyldimethylammonium chloride, which has a MW of 161 Da. It has an average MW of 200–350 kDa and 100% cationic charge density. The copolymer of CoAA was kindly supplied by Ciba Speciality Chemicals, UK. The cationic copolymer has the monomers of methyl chloride, adipic acid and dimethyl aminoethyl acrylate, which has a MW of 143 Da. It has an approximate average MW higher than 2000 kDa and has a cationic charge density of 85%. The average MW of CoAA was approximated using the intrinsic viscosity value supplied by the manufacturer.

2.2. Humic substance extraction

Humic acid (HA) and fulvic acid (FA) were extracted from soil using a modified procedure originally developed by Thurman and Malcolm (1981) [13–14]. First class soil with low fertilizer contents was purchased from Sainsbury's Homebase. 6.0 M HCl solution, anhydrous solid pellets NaOH, non-ionic polymethyl methacrylate (XAD-8) adsorption resin were all purchased from Sigma–Aldrich. 0.5 M NaOH was prepared from NaOH solid pellets and used through out the experimental work. An oven at 105°C was used to measure the concentration of solutes, i.e., Salt, HS and other solid contaminants, and a furnace at 600°C was used for salt concentration measurements. The purity of the extracted HA and FA are 89.5% and 74.5%, respectively.

2.3. Equipment

Shimadzu total organic carbon analyzer model TOC-5050A was used to measure the concentration of the solution. A user-friendly program was used in junction with the analyzer to provide automated concentration curves and readings.

The retention experiments were carried out in a laboratory-scale filtration cell. Schematic diagram of the experimental set-up is shown in Fig. 1. Five litres of the feed is pumped using a stainless steel gear pump into the filtration cell. A rectangular shaped membrane (3 cm × 10 cm × 0.5 cm) with an effective membrane area of 30 cm² was employed. The operating pressure and volumetric flow rate were adjusted using the concentrate (reject) outlet valve. The pressure was varied between 6–9 bars. The experiments were carried out at ambient temperature in total re-circulation mode, i.e., both the concentrate and the permeate streams are re-circulated into the feed tank, so that the feed concentration is kept approximately constant.

2.4. Procedure

Stock solutions preparation procedure is identical to the procedure of the previous UF membrane work [12]. Synthetic stock solutions of 1 g/L concentration were prepared for the tested chemicals by dissolving/diluting using de-ionized water. Due to the high viscosity of CoAA, the concentration of the stock solution used was 0.2 g/L. The stock solutions were diluted with de-ionized water to obtain 10 mg/L concentration and were used within a week of preparation. The pH of all solutions was regulated at ~7 before each experiment using NaOH. Membrane compaction was undertaken using 5 L of de-ionized water for 2 h at a pressure of 9 bars prior to any experiment. The pure water flux (J_0) against pressure curve

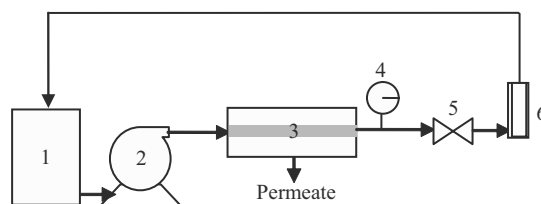


Fig. 1. Cross flow filtration model. 1: feed tank; 2: gear pump; 3: membrane cell; 4: pressure gauge; 5: valve; 6: rotameter.

was constructed using trans-membrane pressure range of 6.0–9.0 bars. Five litres of the feed were pumped using a stainless steel gear pump into the filtration cell channel. The pure water flux was then measured by measuring the time taken to collect 10 mL of permeate. The de-ionized water was then replaced by the filtrated solution. The system was run until an approximate constant permeate flux was observed. The solution volumetric flux (J_v) was measured and the concentration of permeate collected was analyzed using the TOC analyzer.

2.5. Concentration polarization model

Membrane processes develop concentration gradient in the boundary layer at the surface of the membrane. A higher solute concentration is present at the membrane surface due to the gradual increase of the rejected solute near the membrane surface. Concentration polarization model predicts the effect of concentration polarization on solute retention and estimates the real retention of the solute (Fig. 2). Observed and real retention calculations are similar to the previous work in UF membranes characterization, with the exception of mass transfer coefficient estimation [12].

Observed retention equation is a simple equation to calculate the rejection characteristics of a membrane [10]:

$$R_{\text{obs}} = 1 - \frac{C_p}{C_b} \quad (1)$$

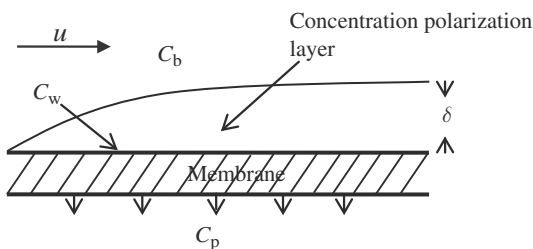


Fig. 2. Schematic of the film layer on membrane surface [12].

where C_p and C_b are the concentrations of permeate and bulk solutions, respectively. When concentration polarization is present, the concentration at the membrane surface is higher than the bulk concentration. Therefore, the real rejection of the membrane should be defined as [10]:

$$R_{\text{real}} = 1 - \frac{C_p}{C_w} \quad (2)$$

where C_w is the boundary layer (wall) solute concentration. C_w is not obtained directly. The concentration polarization model assuming concentration polarization occurring within the boundary layer of thickness (δ) is correlated as [15]:

$$J_v = k \ln \left(\frac{C_w - C_p}{C_b - C_p} \right) \quad (3)$$

where J_v is the flux ($\text{m}^3/\text{m}^2 \text{ s}$) and k is the mass transfer coefficient in the polarized boundary layer (m/s). The mass transfer coefficient for a rectangular flat sheet cross-flow filtration cell is given by the following expression [16]:

$$Sh = \frac{kd_h}{D} = 1.195 Re^{0.554} Sc^{0.371} \left(\frac{d_h}{L} \right)^{0.131} \quad (4)$$

Where Sh is the Sherwood number kd_h/D , d_h is the hydraulic diameter of the filtration channel (m) and D is the bulk diffusivity of the solute. Re is the Reynolds number (ud_h/ν), u is the linear velocity in the cross-flow filtration channel (m/s) and ν is the kinetic viscosity defined as η/ρ where η and ρ are the viscosity and the density of the fluid, respectively. The viscosity of the fluid was taken as the viscosity of water at 20°C (1.002 Pa s) and the density is equal to the density of water at 20°C (998.2 kg/m³). Sc is the Schmidt number (ν/D) and L is the length of the membrane channel (m). The diffusivity of the PEG solutes is calculated using [17]:

$$D = 2.74 \times 10^{-9} M^{-1/3} \quad (5)$$

where M is the molecular weight of the polymer (Da or k mol/kg).

The polyelectrolytes and humic substances do not have uniform molecular weight. Similar to previous work on UF membrane characterization [12], Eq. (3) is modified to determine the k value [10]:

$$\ln \frac{1 - R_{\text{obs}}}{R_{\text{obs}}} = \ln \frac{1 - R_{\text{real}}}{R_{\text{real}}} + \frac{J_v}{k} \quad (6)$$

A plot of $\ln(1 - R_{\text{obs}}/R_{\text{obs}})$ against J_v will produce a gradient equal to $1/k$.

3. Results and discussion

3.1. Pure water flux results

Pure water flux (J_0) of two NF membranes was determined. Fig. 3 presents the effect of trans-membrane pressure on J_0 of NF270 and NF90 membranes. J_0 is calculated using:

$$J_0 = \frac{V}{At} \quad (7)$$

where J_0 is the pure water flux ($\text{L}/\text{m}^2 \text{ h}$), V is the volume collected (L), A is the effective membrane area (m^2) and t is the time taken for 10 mL to filtrate (hour).

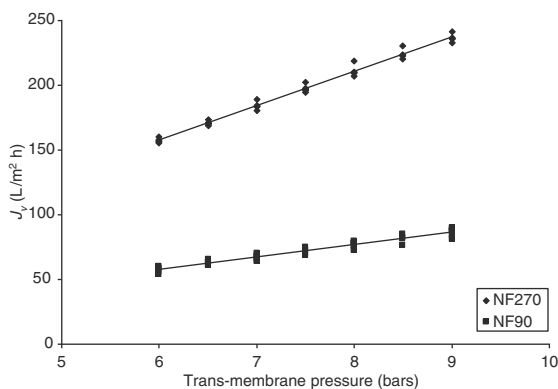


Fig. 3. Pure water flux (J_0) versus trans-membrane pressure using NF270 and NF90 membranes.

J_0 is directly proportional to trans-membrane pressure. Increasing TMP increases J_0 . J_0 of NF270 is 157.4, 184.2, 211.1 and 236.7 at TMP of 6, 7, 8 and 9 bars, respectively. NF90 produced lower J_0 compared to NF270 at the same TMP. J_0 of NF90 is 57.4, 67.2, 76.9 and 86.4 at TMP of 6, 7, 8 and 9 bars, respectively. These results are consistent with Hilal et al. [18] AFM study using NF270 and NF90 membranes. According to Hilal et al. [18] NF90 membrane has a relatively smaller pore size and lower porosity compared to NF270 membrane, therefore, the higher water flux of NF270 is expected.

3.2. PEG retention results

NF270 and NF90 membranes were operated at 8 bars TMP and 10 mg/L PEG during membrane characterization experiments. Fig. 4 presents the $\text{MWCO}_{\text{Real}}$ of NF270 and NF90 membranes. NF270 membrane $\text{MWCO}_{\text{Real}}$ with respect to real retention is lower than $\text{MWCO}_{\text{Real}}$ of NF90, which is unexpected. NF270 $\text{MWCO}_{\text{Real}}$ is ~ 0.18 kDa compared to ~ 0.38 kDa for NF90 (Fig. 4a). The higher NF270 $\text{MWCO}_{\text{Real}}$ is due to the inconsistency in real retention equation, Eq. (3). Real retention determination is dependent on permeate flux (J_v) instead of taking account of the reduction of flux of membrane. The NF270 membrane was suppose to operate at lower pressure to obtain a permeate flux similar to that of NF90 to compensate the inconsistency of the equation. Unfortunately, both of these membranes act strangely at low pressures, according to previous experimental results not included in this paper.

The MWCO of the two membranes with respect to observed retention ($\text{MWCO}_{\text{Observed}}$) agree with the AFM results by Hilal et al. [18]. NF90 membrane produced lower $\text{MWCO}_{\text{Observed}}$ compared to NF270 membrane (Fig. 4b). $\text{MWCO}_{\text{Observed}}$ of NF270 is ~ 1.11 kDa compared to ~ 0.74 kDa for NF90. The lower $\text{MWCO}_{\text{Observed}}$ of NF90 is due to its relatively smaller pore size and lower porosity compared to NF270.

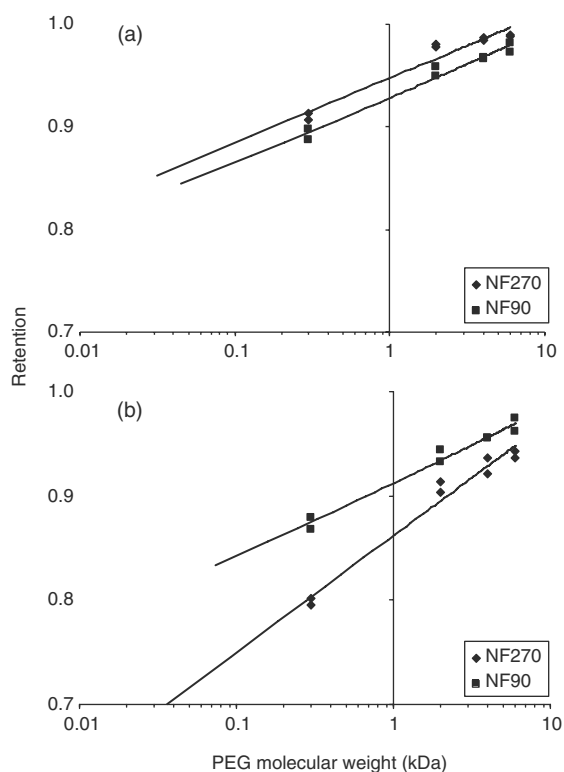


Fig. 4. Molecular weight cut-off of NF270 and NF90 membranes at 8 bars TMP using PEG; (a) real retention and (b) observed retention.

3.3. Polyelectrolytes retention results

The retention of the coagulating polyelectrolytes, i.e., CoAA and PDADMAC were studied at different TMPs (7–9 bars) and 10 mg/L polyelectrolyte concentration. Figs. 5 and 6 present the observed (R_{obs}) and real retention (R_{real}) of PDADMAC and CoAA against TMP. No change in R_{obs} and R_{real} of both polyelectrolytes was experienced when TMP was increased using both membranes. For example, R_{obs} of PDADMAC using NF270 was 0.94, 0.95 and 0.95 at TMP of 7, 8 and 9 bars, respectively, and R_{real} of PDADMAC using NF270 was 0.96, 0.98 and 0.98 at TMP of 7, 8 and 9 bars, respectively. Two competing phenomena determine solutes retention with respect to TMP. These competing factors are the concentration polarization and the dilution effects. Solute retention is the sum of these opposing effects. Increasing TMP increases concentration polarization, thus increasing the amount of solute settling at the surface of the membrane and reducing retention. The dilution effect counteracts the concentration polarization effect. The dilution effect increases when the TMP is increased. When the TMP is increased, the permeate water flux through the membrane increases, while the ion flux remains virtually unchanged, resulting in lower solute concentration at the permeate [19].

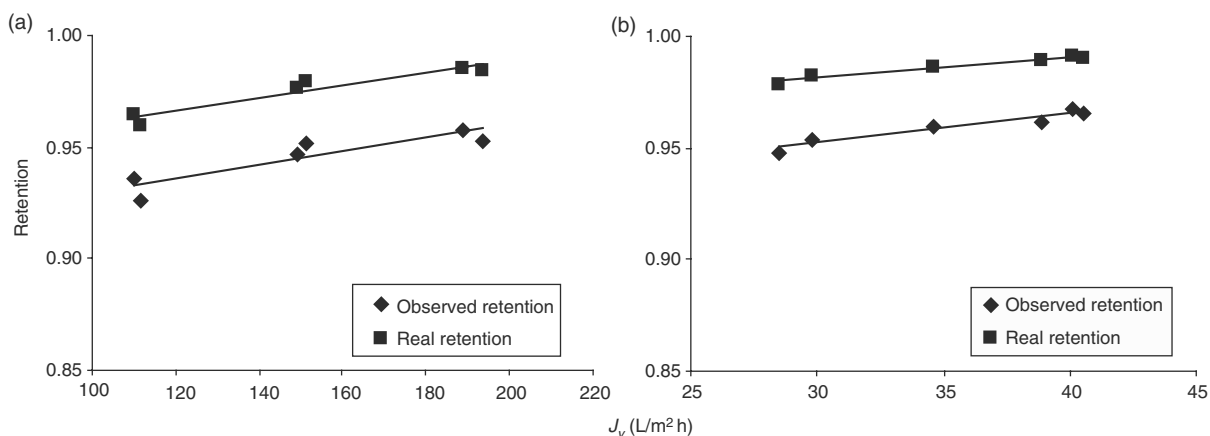


Fig. 5. Real and observed PDADMAC retention of two membranes; (a) NF270 membrane and (b) NF90 membrane.

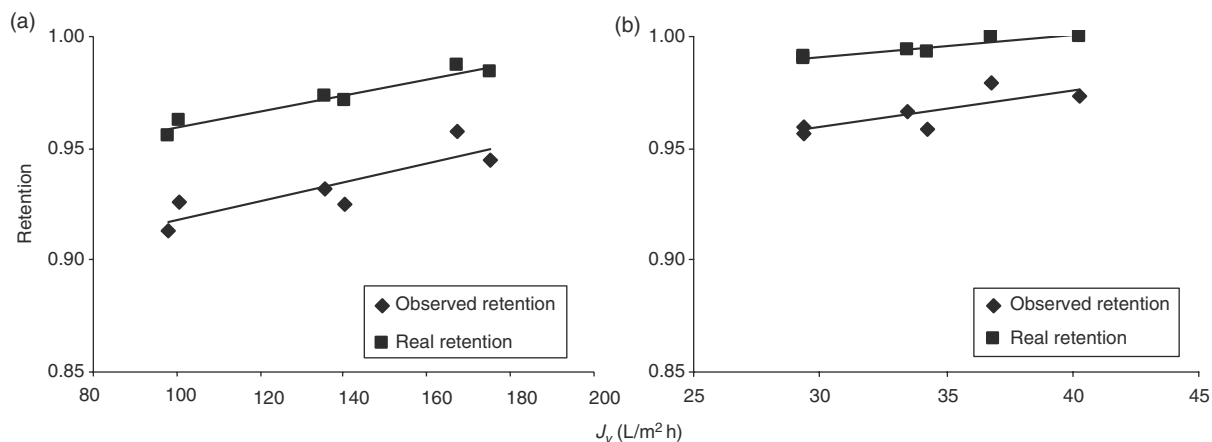


Fig. 6. Real and observed CoAA retention of two membranes; (a) NF270 membrane and (b) NF90 membrane.

Complete retention of PDADMAC and CoAA was expected using both membranes, due to their high molecular weights. Figs. 5 and 6 present PDADMAC and CoAA retention using NF270 and NF90 membranes. Both polyelectrolytes were not completely rejected. Real retention of PDADMAC is 0.98 for both membranes, while CoAA real retention is 0.96 and 0.99 using NF270 and NF90, respectively, at 7 bars TMP. The incomplete rejection is due to polyelectrolyte manufacturing process. Polyelectrolyte formulations contain contaminants from the manufacturing process such as residual monomers, other reactants and reaction by-products [20]. PDADMAC and CoAA monomers have molecular weights of 161 and 143 Da, respectively. These monomers have a molecular weight lower than the MWCO of the membranes; ~ 0.74 and ~ 1.11 for NF90 and NF270, respectively, and may pass through the membranes pores.

3.4. Humic and fulvic acid retention results

The retention of HA and FA using NF270 and NF90 membranes was studied at different TMPs (7–9 bars) and 10 mg/L HS concentration. Figs. 7 and 8 present the observed (R_{obs}) and real

retention (R_{real}) of HA and FA against TMP. No change in R_{obs} and R_{real} of both HS was experienced when TMP was increased using both membranes. For example, R_{obs} of HA using NF270 was 0.93, 0.94 and 0.94 at TMP of 7, 8 and 9 bars, respectively, and R_{real} of PDADMAC using NF270 was 0.96, 0.97 and 0.98 at TMP of 7, 8 and 9 bars, respectively. These results show the same trend as polyelectrolyte retention experiments (section 3.3). HS retention with respect to TMP experiences a competition between concentration polarization and dilution effect. The observed retention is the sum of these opposing effects.

Real and observed retention of HA and FA are both slightly higher using NF90 compared to NF270. Real retention of HA is 0.96 and 0.98 using NF270 and NF90, respectively, while FA real retention is 0.94 and 0.97 using NF270 and NF90, respectively, at 7 bars TMP. As explained previously, the higher retention of NF90 membrane is due to its relatively smaller pore size and lower porosity compared to NF270. FA real and observed retention was also lower than HA retention using both membranes. This is expected since FA has a lower molecular weight range compared to the molecular weight of HA.

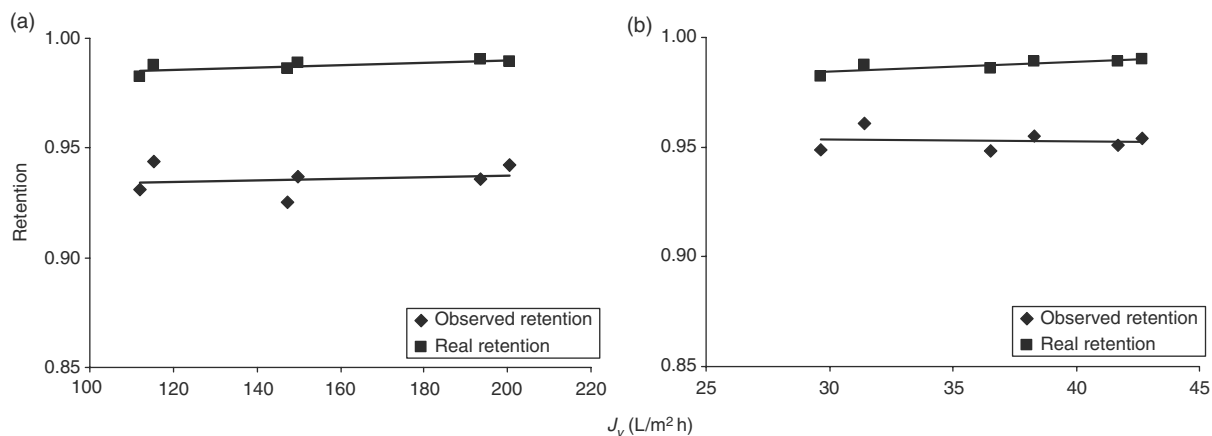


Fig. 7. Real and observed HA retention of two membranes; (a) NF270 membrane and (b) NF90 membrane.

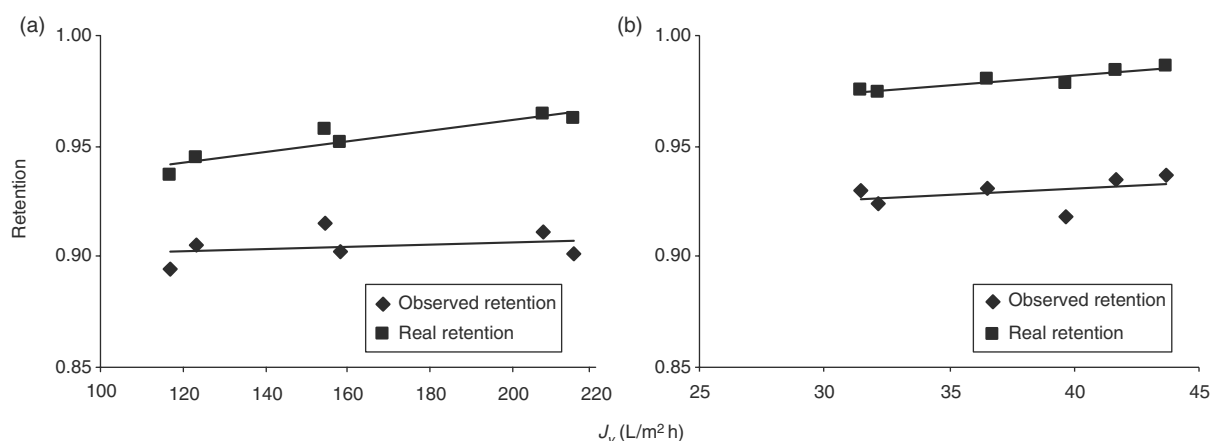


Fig. 8. Real and observed FA retention of two membranes; (a) NF270 membrane and (b) NF90 membrane.

4. Conclusions

- Pure water flux of NF270 membrane is higher than NF90 membrane because of its relatively high pore size and porosity.
- Observed MWCO of NF90 (~0.74) is lower than NF270 (~1.11), which is expected. The unexpected higher real MWCO of NF90 (~0.38) compared to NF270 (~0.18) is due to the inconsistency of the concentration polarization model.
- Incomplete retention of PDADMAC and CoAA is due to the presence of their monomers.
- HA retention is higher than FA because of its higher molecular weight range. NF270 membrane retention of HA and FA is lower than NF90 because of its higher pore size and porosity.
- Solute retention is constant with increasing pressure due to the competition between concentration polarization and dilution effects at the studied range of TMP.

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Nomenclature

R_{obs}	Observed retention
R_{real}	Real retention
C_p	Permeate concentration (mg/L)
C_b	Bulk concentration (mg/L)
C_w	Wall concentration (mg/L)
J_0	Pure water flux (L/m ² h) or (m ³ /m ² /s)
J_v	Solution volumetric flux (L/m ² h) or (m ³ /m ² /s)
k	Mass transfer coefficient (L/m ² h) or (m ³ /m ² /s)
D	Bulk diffusivity of solute (m ² /s)
δ	Thickness of film layer (m)
ν	Kinetic viscosity
η	Viscosity of solution (Pa s)
ρ	Density of solution (kg/m ³)
M	Weight-average molecular weight (kg/kmol or Da)
$\text{MWCO}_{\text{Real}}$	Membrane molecular weight cut-off with respect to real retention
$\text{MWCO}_{\text{Observed}}$	Membrane molecular weight cut-off with respect to observed retention
Sh	Sherwood number (kd_h/D)
Re	Reynolds number (ud_h/ν)
Sc	Schmidt number (ν/D)
d_h	Hydraulic diameter of the filtration channel (m)
D	Bulk diffusivity of solute
u	Linear velocity in the cross-flow filtration channel (m/s)
ν	Kinetic viscosity (η/ρ)
η	Viscosity of water at 20°C (1.002 Pa s)

ρ	Density of water at 20°C (998.2 kg/m ³)
L	Length of the membrane channel (m).

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