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Studies on Permeation, Rejection, and Transport of Aqueous Poly(ethylene Glycol) Solutions using Ultrafiltration Membranes

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Abstract: The permeate flux and retention of aqueous solutions of poly(ethylene glycols) (PEG) with different molecular weights ranging from 4000 to 35,000 Da have been investigated using various compositions such as 100/0, 90/10, 80/20, and 70/30 wt% of cellulose acetate (CA)/sulfonated poly(etheretherketone) (SPEEK) ultrafiltration blend membranes. The factors affecting the rejection rate and permeate flux such as molecular weight of PEGs, concentration of the solute, composition of the membranes, and transmembrane pressures have been studied. It is seen that the increase in the concentration of PEG results in the decreased permeate flux and increased rejection for increasing CA content in the membranes. A similar observation in the flux and rejection was made for increasing the molecular weight of PEGs. Further, the mass transfer, diffusion, and true retention coefficients of the solute have been studied with different operating variables like molecular weight and concentration of PEGs. An increase in the molecular weight of PEGs results in the decrease of mass transfer and diffusion coefficients and increase of the true retention coefficient. A reverse trend is observed with increasing concentrations of PEG.

Keywords: Ultrafiltration, poly(ethylene glycols), permeate flux, rejection, mass transfer coefficient

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INTRODUCTION

Membrane separation processes have been considered as promising technology with significant technical and commercial impact. Membranes find many scientific and technological applications and they are used in many industries for the production of particle free solution. Ultrafiltration (UF) is a low-pressure filtration process with the ability to separate the molecules from the solution based on their size. UF is an established process being largely applied for macromolecular separations from aqueous streams and have been extensively used for the product recovery and pollution control in chemical, electronic, electrocoating as well as in food, pharmaceutical, and biotechnological industries (1). Cellulose acetate (CA) has been found to be a successful blend component with polysulfone (PS), sulfonated polysulfone (SPS), and epoxy resin in preparing UF membranes (2–4). The performance of CA may be improved by blending it with an appropriate polymer in view of the fact that polymer blends have provided an efficient way to fulfill new requirements for material properties. Sulfonated poly(etheretherketone) (SPEEK) is being used as a successful blend material due to its superior qualities (5). Recently, CA/SPEEK ultrafiltration membranes have been prepared, characterized, and studied for the rejection of proteins at various molecular weights (5). As the SPEEK is hydrophilic in nature, the blending of SPEEK with CA resulted in membranes with increased porosity as seen through various performance data and would also substantially reduce the fouling behavior of the resulted membranes. These membranes are chosen for the present studies due to their excellent film forming properties. The permeation and rejection characteristics depend on the process parameters and the device characteristics where the membrane is used. Poly(ethylene glycol) (PEG) is chosen to characterize the membranes 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 (6). The main objective of this work is to study the effects of CA/SPEEK blend compositions on the percentage rejection and product rate efficiencies of PEGs with various molecular weights from aqueous streams. The mass transfer coefficient, diffusion coefficient, and the true retention coefficient have been determined. From true retention coefficient, the radius of gyration of the PEG molecule has been calculated.

EXPERIMENTAL

Materials

PEGs of different molecular weights were obtained from E-Merck Ltd., Germany. Barium chloride (Laboratory Reagent) was procured from S.D.

Fine Chemicals Ltd., Mumbai, India. *N,N′*-dimethylformamide (DMF) and sodium lauryl sulfate (SLS) were obtained from Qualigens Fine Chemicals, Glaxo India Ltd., India, which were of analytical grade. Hydrochloric acid (Laboratory Reagent), potassium iodide (Analytical Reagent), and iodine (Hi-Pure) were purchased from Ranbaxy Fine Chemicals Ltd., New Delhi, India. These chemicals were used for spectrophotometric analysis. Deionized and distilled water was employed for the preparation of aqueous PEG solutions.

Methods

The UF experiments were carried out in a batch type, dead end cell (UF cell-S76-400-Model, Spectrum, USA) with a diameter of 76 mm fitted with a Teflon coated magnetic paddle. The rejection studies were carried out at room temperature using UF module of 450 ml capacity and holdup volume of 10 ml. The effective membrane surface area was 38.5 cm^2 and the applied pressure was 345 kPa. A constant agitation speed of 500 rpm was used throughout the study, in order to reduce the concentration polarization.

PEG was estimated by UV-visible spectrophotometry. Four milliliters of sample solution was added to 1 ml 5% (w/v) $BaCl_2$ in 1 N HCl. To this mixture, 1 ml of solution prepared by dissolving 1.27 g I_2 in 100 ml 2% KI (w/v) solution was added, which is further diluted 10 times. Color was allowed to develop for 15 min at room temperature, and absorption was read using a spectrophotometer at 535 nm against a reagent blank.

Preparation of Membranes

Various compositions of CA/SPEEK blend UF membranes such as 100/0, 90/10, 80/20, and 70/30 wt% were prepared and characterized as detailed earlier (5). Membranes were prepared using standard method of phase inversion technique. The polymer solution was first cast on a smooth glass plate with the help of a doctor blade. The thickness of the membrane was maintained at 0.22 ± 0.02 mm with the help of an oil sheet rolled at both the ends of the blade. The casting conditions are reported in our previous study (5). Prior to casting, a gelation bath of 2 liters consisting of 2.5% (v/v) *N,N′*-dimethylformamide and 0.2 wt% sodium lauryl sulfate in distilled water (non solvent) was prepared and the bath was ice-cooled to 18 ± 2°C. After 30 min of solvent evaporation in the casting chamber, the glass plate along with the polymer film was immersed in the gelation bath. After an hour of gelation, the membrane was removed from the gelation bath and thoroughly washed with distilled water to remove the residual solvent and surfactant from the membrane. The membrane sheet was subsequently stored in distilled water containing 0.1% of formalin solution to prevent microbial growth. Similar casting and gelation conditions were maintained for all the membranes.

Morphological Studies

CA/SPEEK membranes with composition 90/10, 80/20, and 70/30 wt% were cut into small pieces and mopped with filter paper. These pieces were immersed in liquid nitrogen for 20–30s and were frozen. The frozen pieces of membranes were further cut into bits and kept in a desiccator. These membrane samples were used for SEM studies. The membrane samples were mounted on studs and gold-sputtered to provide electrical conductivity to very thin layer of the polymeric membrane (7). The cross sections of the membranes were viewed using Jeol JSM-840A scanning electron microscope.

Rejection and Permeate Flux Studies

Effect of Membrane Composition

Aqueous solutions with various concentrations such as 10, 20, 30, 40, and 50 ppm were prepared for lower and higher molecular weight PEGs namely 4000 and 35,000 Da. The compacted CA/SPEEK membranes such as 100/0, 90/10, 80/20, and 70/30 wt% were mounted in the UF kit, and the prepared solutions were transferred and pressurized under nitrogen atmosphere at 345 kPa, which was maintained constantly throughout the operation. The permeate flux was calculated over measured time intervals using following equation (8).

$$J = \frac{Q}{A \cdot \Delta T} \tag{1}$$

where, J = Permeate flux, $1\,m^{-2}\,h^{-1}$; Q = Quantity of permeate, l; A = Membrane area, m^2; ΔT = Sampling time, h.

The PEG concentrations in the feed and permeate were analyzed by UV spectrophotometer at 535 nm against a reagent blank (9). From this the percentage rejection of PEGs was calculated using equation (2) (10).

$$\%SR = \left[1 - \left(\frac{C_p}{C_f}\right)\right] \times 100 \tag{2}$$

where, C_p and C_f are the concentrations of permeate and feed PEG solutions respectively.

Effect of Molecular Weight of PEGs

High concentration aqueous solutions (50 ppm) of PEGs with different molecular weights namely 4000, 10,000, 20,000, and 35,000 Da were prepared. The prepared CA/SPEEK membranes such as 100/0, 90/10, 80/20, and 70/30 wt% were mounted in the UF kit, and the prepared solutions were transferred and pressurized under nitrogen atmosphere at 345 kPa, which was maintained constantly throughout the operation. The permeate flux and percentage rejection were calculated as described above.

Effect of Transmembrane Pressure

An aqueous solution with high concentration, namely 50 ppm, was prepared for higher molecular weight PEG namely 35,000 Da. The CA/SPEEK membranes such as 100/0, 90/10, 80/20, and 70/30 wt% were mounted in the UF kit and the prepared solution was transferred. The transmembrane pressure was varied as 69, 138, 207, 276, and 345 kPa under nitrogen atmosphere. The permeate flux and percentage rejection were calculated as described above.

Determination of Mass Transfer Coefficient, Diffusion Coefficient, and Radius of Gyration

The concentration of the solute at the membrane surface is greater than that of the bulk resulting from concentration polarization. This can be studied using the film layer model (11) that assumes a zone where the concentration decreases from the membrane to the surface at a distance δ inside the retentate phase (12–15) and this leads to

$$J_v = K_m \ln\left[\frac{C_m - C_p}{C_f - C_p}\right] \tag{3}$$

where, J_v = volume flow per unit area and time through the membrane, m/s; K_m = mass transfer coefficient, m/s; C_m = membrane concentration in contact with the high pressure interface, mol/m^3; C_p = permeate concentration, mol/m^3; C_f = feed concentration, mol/m^3. Incorporating the effects of concentration polarization, the observed retention can be compared to the true retention of a membrane system by the following equation (16, 17).

$$\ln\left[\frac{1 - R_0}{R_0}\right] = \ln\left[\frac{1 - R}{R}\right] + \left[\frac{J_v}{K_m}\right] \tag{4}$$

where R_0 is the observed retention coefficient and R is the true retention coefficient (6). ln $[(1 - R_0)/R_0]$ was plotted against J_v for experiments with various molecular weight of PEGs at 50 ppm concentration as well as high molecular weight PEG at all the concentration levels. The plots showed a linear fit. From the corresponding line equation, the slope $(1/K_m)$ and intercept ln $[(1 - R)/R]$ were obtained. From which, the mass transfer coefficient, K_m, and true retention coefficient, R, were determined.

The relationship between the mass-transfer coefficient and the diffusion coefficient for liquid–solid interfaces in the case of the membrane is given as (18),

$$K_m = \frac{D_m}{\delta} \tag{5}$$

where, K_m is membrane mass-transfer coefficient, D_m is diffusion coefficient, and δ is membrane thickness. D_m is calculated from Eq. (5). The thickness of the membrane is taken as the average constant value.

According to Flory's theory (19), the radius of gyration, r_g, of PEG molecule was derived from its molecular weight as follows.

$$r_g = \left[\frac{\langle r \rangle^2}{6}\right] \tag{6}$$

where $\langle r \rangle$ is the distance from the chain ends of a PEG molecule, m.

For aqueous solutions of PEGs at 298 K $\langle r \rangle$ is given by,

$$\langle r \rangle = (775 \pm 30) \times 10^{-13} M_w^{0.5} \tag{7}$$

where M_w is the molecular weight of PEG.

RESULTS AND DISCUSSION

The blended membranes with different CA/SPEEK compositions such as 100/0, 90/10, 80/20, and 70/30 wt% have been used for the rejection of various molecular weight PEG solutions. The above membranes were previously characterized for its compaction, pure water flux, membrane hydraulic resistance and water content (5). The effect of various parameters like the molecular weight of the PEG, concentration of the solute, composition of the membranes and transmembrane pressure on the performances of these membranes have been studied.

Morphological Studies

Morphology of the blend membranes with different polymer compositions were analyzed by SEM and the results are depicted in Fig. 1. At 100 wt% of CA, the membrane exhibits smaller pores distributed evenly (5). It is evident from Fig. 1 that as the SPEEK composition increased in the casting solution, the pore size increased proportionally. The cross section of the membrane confirms the asymmetry in the ultra structure of the membrane. The addition of SPEEK to CA greatly increased the formation of macrovoids. Thus it is logical to expect a substantial increase in the membrane permeability upon increasing SPEEK component in the CA/SPEEK blend membranes.

Effect of PEG Concentrations on Membrane Compositions

The membranes with different compositions were analyzed for permeate flux and rejection by varying the PEG concentrations as 10, 20, 30, 40, and 50 ppm for molecular weights 4000 and 35,000 Da. As the composition of the CA increases from 70 to 100 wt%, the permeate flux decreases and the rejection increases for all the concentrations of PEG with molecular weight 4000 Da, as shown in Figs. 2 and 3 respectively. The increase in the permeate flux and the

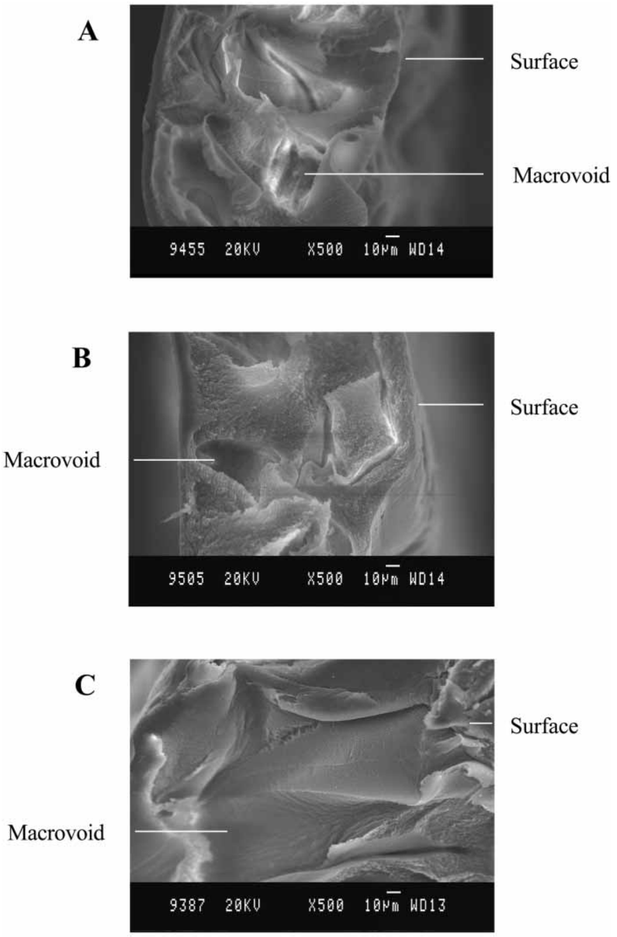

Figure 1. SEM micrographs of cross section of CA/SPEEK membranes with different blend compositions: A) 90/10; B) 80/20; C) 70/30 wt% of CA/SPEEK blend membranes (original magnification, 500X).

decrease in rejection upon increase in SPEEK composition may be due to the formation of pores with bigger pore sizes. CA/SPEEK membrane of 70/30 wt% composition offered a higher permeate flux of 87 $l/(m^2h)$ while 100 wt% CA membrane offered a higher rejection of 85% for a PEG (M_W 4000 Da) concentration of 10 ppm. Increase in the concentration of PEG solutions resulted in decrease in permeate flux as well as rejection for all the membrane compositions as expected. A similar trend can be seen for PEG with molecular weight of 35000 Da. However, the values of the permeate flux and the rejection differs to that of PEG with a molecular weight of 4000. The CA/SPEEK membrane of 70/30 wt% composition provided the highest permeate flux of 56 $l/(m^2h)$

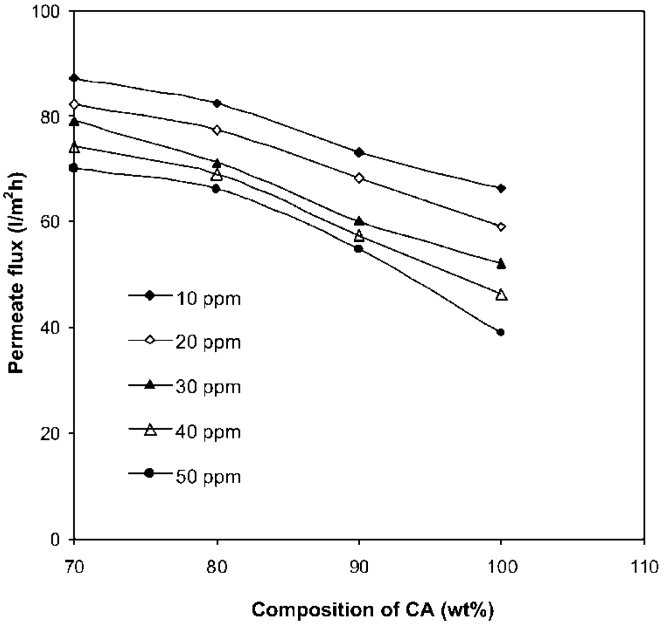

Figure 2. Effect of CA/SPEEK membrane compositions on permeate flux for various PEG concentrations ($M_w = 4000$).

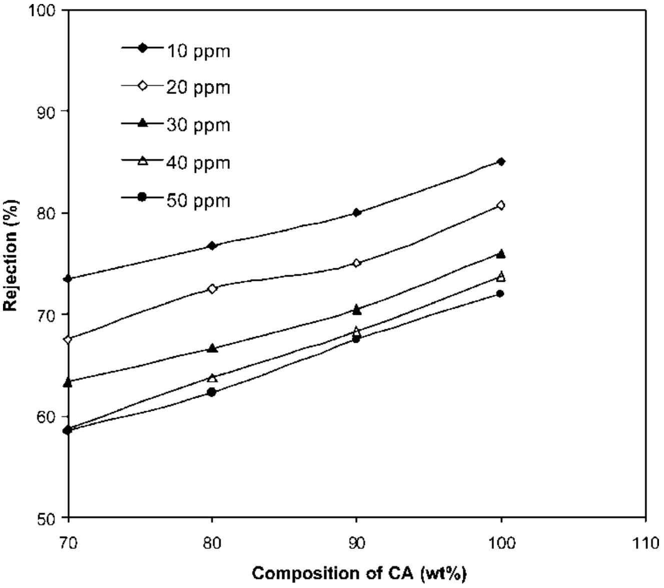

Figure 3. Effect of CA/SPEEK membrane compositions on rejection (%) for various PEG concentrations ($M_w = 4000$).

while the CA membrane offered the highest rejection of 98% for a PEG (M_W 35,000 Da) concentration of 10 ppm. The higher rejection and lower permeate flux observed in the 100 wt% CA membrane could be due to the smaller pore size in relation to a decreasing segmental gap between the polymeric chains during the membrane formation (20). These results are in agreement with the morphological studies.

Effect of Molecular Weight of PEG on Membrane Performance

The membranes with different compositions were analyzed for permeate flux and rejection by varying the molecular weights of PEG's as 4000, 10,000, 20,000, and 35,000 Da at a concentration of 50 ppm. As the molecular weight of PEG increases permeate flux decreases while the rejection increases for all the membranes as shown in Figs. 4 and 5 respectively. As the molecular weight of PEG increases, the viscosity of the solution increases and this leads to the decrease in permeate flux and increase in the rejection (21–24). Highest permeate flux is achieved with 70/30 wt% CA/SPEEK membrane for PEG with molecular weight of 4000 Da, which is 70 l/(m^2h). On the other hand, higher rejection is accomplished with 100 wt% CA membrane for PEG with molecular weight of 35,000 Da, which is 89%.

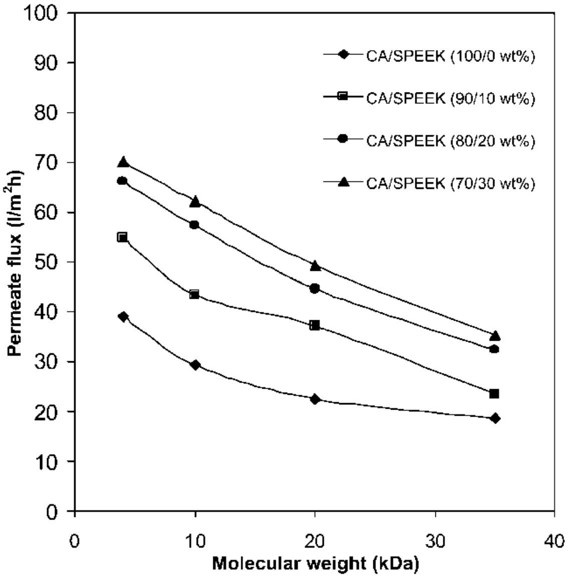

Figure 4. Effect of molecular weight of PEGs on permeate flux for different CA/SPEEK membrane compositions.

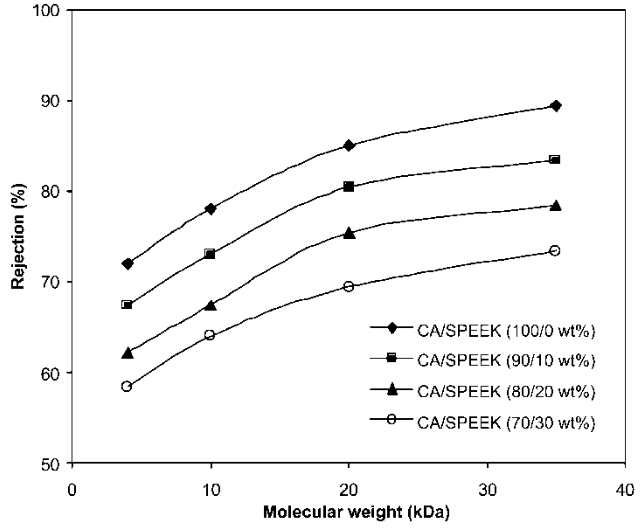

Figure 5. Effect of molecular weight of PEGs on rejection (%) for different CA/SPEEK membrane compositions.

Effect of Transmembrane Pressure on Membrane Performance

The membranes with different compositions were analysed for permeate flux and rejection by varying the transmembrane pressure as 69, 138, 207, 276, and 345 kPa for PEG (M_w 35000 Da) with a feed concentration of 50 ppm. It is evident that the increase in the transmembrane pressure results in the increase of permeate flux at a linear rate for all the membranes as shown in Fig. 6. The flux first increased with increasing pressure and finally reached a limiting value leading to a pressure-independent filtration. As the SPEEK content increases in the membrane composition, the permeate flux improves drastically reaching the maximum of 35 $l/(m^2h)$ at 345 kPa. This is primarily due to the formation of macro voids upon blending of SPEEK with CA in the wet phase inversion technique (25). On the other hand, the rejection (%) decreases upon increase in the transmembrane pressure as well as increase in the SPEEK content in the membranes as shown in Fig. 7. A higher rejection of 98% is achieved using a 100 wt% CA membrane at a transmembrane pressure of 69 kPa, however with a lower permeate flux.

Determination of Mass Transfer Coefficient, Diffusion Coefficient, and Radius of Gyration

A plot of $\ln[(1 - R_0)/R_0]$ versus J_v was made for PEG (M_W 35000 Da) at all the selected concentration levels for different compositions of CA/SPEEK

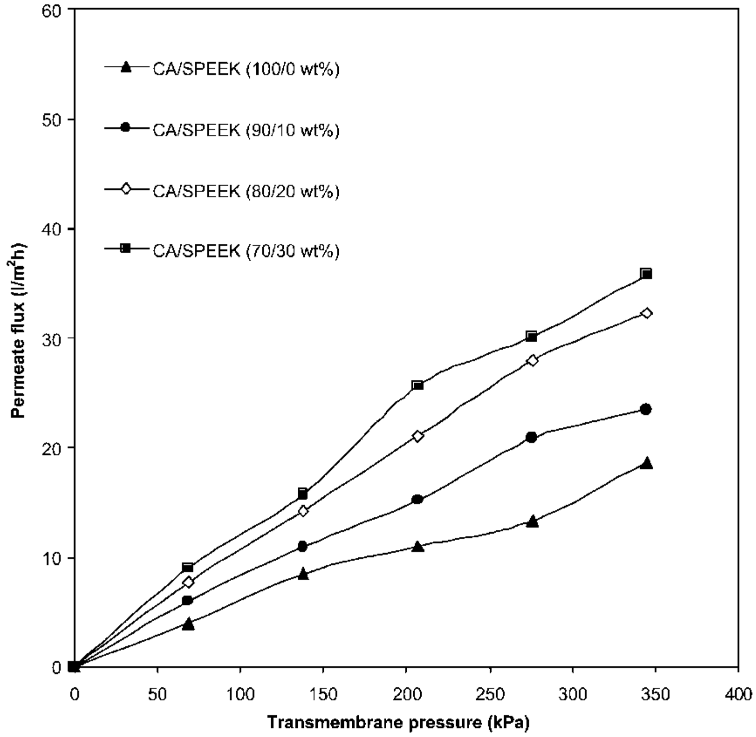

Figure 6. Effect of transmembrane pressure on permeate flux of PEG ($M_w = 35{,}000$) for different CA/SPEEK membrane compositions.

membranes as shown in Fig. 8. The data were fit for linear models with a slope of $1/k_m$ and an intercept of $\ln [(1 - R)/R]$ as per the Eq. (5). The calculated values of mass transfer coefficient (k_m), diffusion coefficient and true retention coefficient (R) are given in Table 1. It is seen that the values of mass transfer and diffusion coefficients are increasing up on increase in the concentration of PEG (M_w 35000 Da) while the true retention coefficient values are decreasing. It is seen that as the concentration increases from 10 to 50 ppm, the true retention coefficient decreases, which leads to an increase in the solute mobility, thereby increasing the mass transfer and diffusion coefficients (26–28).

The effect of the molecular weight of PEGs on the mass transfer, diffusion, and true retention coefficients was studied at 50 ppm concentration for different compositions of CA/SPEEK membranes by plotting $\ln [(1 - R_0)/R_0]$ versus J_v as shown in Fig. 9. The data were fit for linear models and the derived mass transfer, diffusion, and true retention coefficients are given in Table 2. It is seen that the increase in the molecular weight of PEGs results in the decrease of mass transfer and diffusion coefficients as expected,

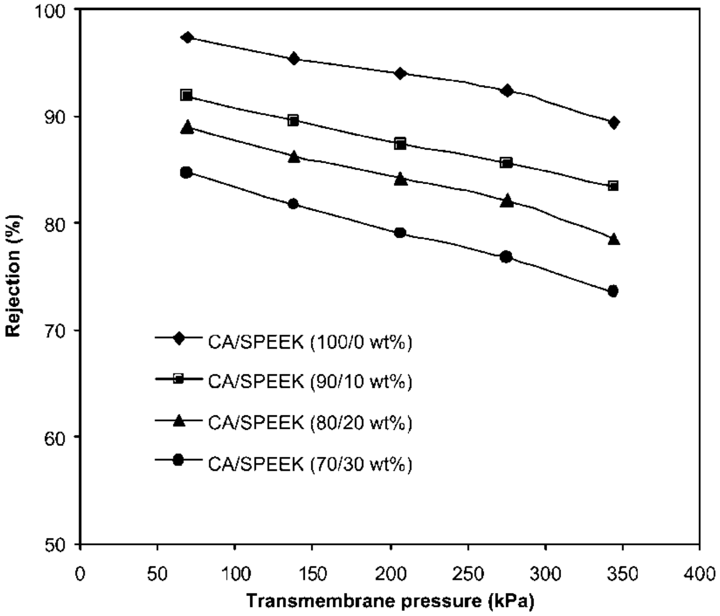

Figure 7. Effect of transmembrane pressure on rejection (%) of PEG ($M_w = 35,000$) for different CA/SPEEK membrane compositions.

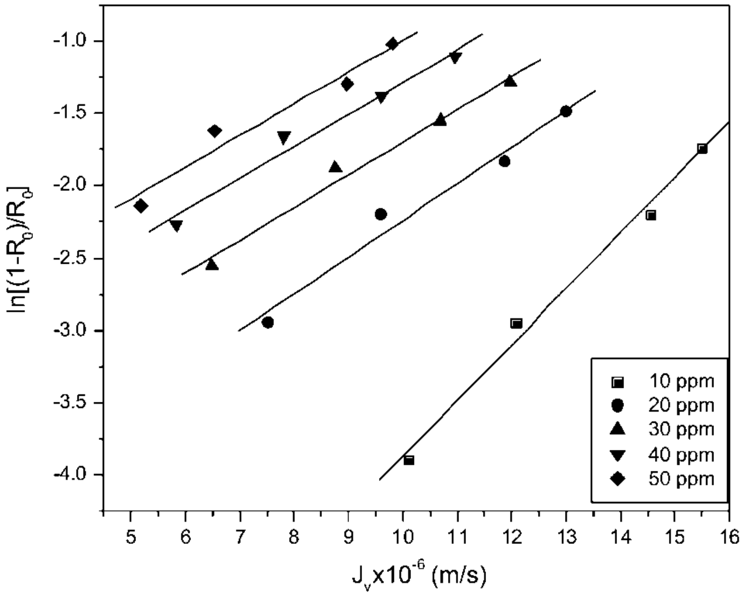

Figure 8. A plot of ln $[(1 - R_0)/R_0]$ versus J_v for varying concentration of PEG ($M_w = 35,000$) for different CA/SPEEK membrane compositions.

Table 1. Relation between concentration of PEG ($M_w = 35$ kDa) for varying compositions of CA/SPEEK membranes and mass transfer, diffusion and true retention coefficients

Concentration of PEG (ppm)	$K_m \times 10^6$ (m/s)	$D_m \times 10^6$ (m^2/s)	R
10	2.59	0.57	0.9996
20	3.94	0.87	0.9916
30	4.43	0.97	0.9812
40	4.53	1.00	0.9705
50	4.57	1.01	0.9603

while the true retention coefficient increases. The increase in true retention coefficient denotes decrease in solute mobility, which lead to the decrease in the mass transfer and diffusion coefficients. In order to find out the relation between true retention coefficient and the molecular weight of PEGs as well as radius of gyration of PEG molecule, the log of M_w of PEGs and radius of gyration of PEGs using Eq. (7) were calculated and presented in Table 2. The true retention coefficients for various molecular weight of PEGs are plotted in a logarithmic scale to obtain a standard retention curve as shown in Fig. 10. It is seen that as the molecular weight of PEG increases the true retention coefficient also increases linearly

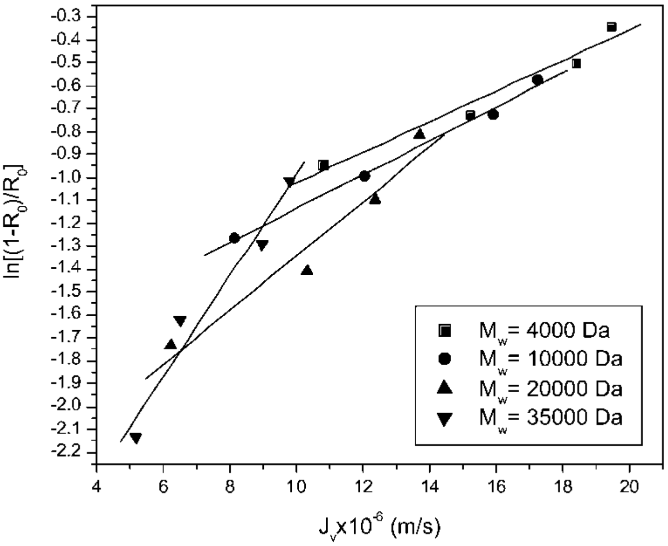

Figure 9. A plot of ln $[(1 - R_0)/R_0]$ versus J_v for varying molecular weight of PEGs (at 50 ppm) for different CA/SPEEK membrane compositions.

Table 2. Relation between molecular weight of PEGs at 50 ppm concentration for varying compositions of CA/SPEEK membranes and mass transfer, diffusion and true retention coefficients and radius of gyration

M_w (Da)	$K_m \times 10^6$ (m/s)	$D_m \times 10^6$ (m^2/s)	R	Log M_w	$r_g \times 10^{18}$ (m)
4000	14.93	3.3	0.845	3.6	4.32
10,000	13.54	3.0	0.867	4.0	10.80
20,000	8.44	1.9	0.926	4.3	21.60
35,000	4.53	1.0	0.961	4.5	37.80

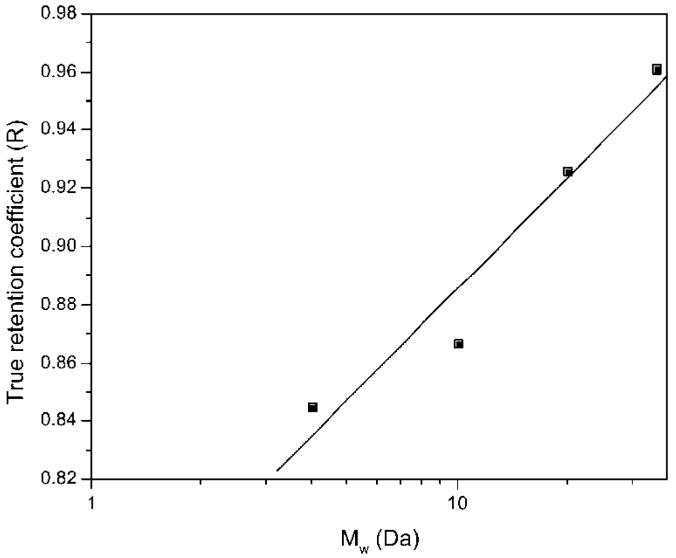

Figure 10. A plot of true retention coefficients versus log of molecular weight of PEGs.

confirming the earlier results. It is also evident from Table 2 that as the radius of gyration of the PEG molecule increases, the true retention coefficient also increases.

CONCLUSIONS

An attempt has been made in the present work to study the retention characteristics of PEGs of various molecular weights using CA/SPEEK blend ultrafiltration membranes. The CA/SPEEK blend membranes with various compositions offer a very good potential for the selection of appropriate retention of PEGs of various molecular weights. Various factors affecting the rejection and permeate flux such as molecular weight of PEGs, concentration of the solute, composition of the membranes, and transmembrane pressures have been studied. The mass transfer and diffusion coefficients have been obtained from the measured flux and retention for applied pressure of 345 kPa in the framework of film theory for the concentration polarization phenomena. It has been shown that the increase in the molecular weight of PEGs results in the decrease of mass transfer and diffusion coefficients. The standard retention curve shows a linear relation between the true retention coefficient values and the molecular weight of PEG molecules.

REFERENCES

1. Cheryan, M. (1986) *Ultrafiltration Handbook*; Technomic Publishing Company: Lancaster, UK.
2. Malaisamy, R., Mahendran, R., and Mohan, D. (2002) Cellulose acetate and sulfonated polysulfone blend ultrafiltration membranes II pore statistics, molecular weight cut off and morphological studies. *J. Appl. Polym. Sci.*, 84: 430.
3. Sivakumar, M., Mohan, D., and Rangarajan, R. (1998) Preparation and performance of cellulose acetate-poly urethane blend membranes and their applications Part-I. *Poly. Int.*, 47: 311.
4. Mahendran, R., Malaisamy, R., and Mohan, D. (2002) Cellulose acetate and epoxy resin blend ultrafiltration membranes: Preparation, characterization, and applications. *J. Macromol. Sci.*, 39: 1025.
5. Arthanareeswaran, G., Srinivasan, K., Mahendran, R., Mohan, D., Rajendran, M., and Mohan, V. (2004) Studies on cellulose acetate and sulphonated poly(etheretherketone) blend ultrafiltration membranes. *Eur. Polym. J.*, 40: 751.
6. Pradanos, P., Arribas, J.I., and Hernandez, A. (1995) Mass transfer coefficient and retention of PEGs in low pressure cross-flow ultrafiltration through asymmetric membranes. *J. Membrane Sci.*, 9: 1.
7. Brink, L.E.S., Elbers, S.J.G., Robertsen, T., and Both, P.J. (1993) The anti-fouling action of polymers preadsorbed on ultrafiltration and microfiltration membranes. *J. Membrane Sci.*, 76: 281.
8. Sivakumar, M., Malaisamy, R., Sajitha, C.J., Mohan, D., Mohan, V., and Rangarajan, R. (2000) Preparation and performance of cellulose acetate-poly urethane blend membranes and their applications-II. *J. Membrane Sci.*, 169: 215.
9. Sabde, A.D., Trivedi, M.K., Ramachandran, V., Hanra, M.S., and Misra, B.M. (1997) Casting and characterisation of cellulose acetate butyrate based ultrafiltration membranes. *Desalination*, 114: 223.
10. Mahandran, R., Malaisamy, R., Arthanareeswaran, G., and Mohan, D. (2004) Cellulose acetate-polyethersulfone blend ultrafiltration membranes. II. Application studies. *J. Appl. Polym. Sci.*, 92: 3659.
11. Colton, C.K., Friedman, S., Wilson, D.E., and Lees, R.S. (1972) Ultrafiltration of lipoproteins through a synthetic membrane. *J. Clin. Invest.*, 51: 2472.
12. Kimura, S. and Nakao, S. (1975) Fouling of cellulose acetate tubular reverse osmosis modules treating the industrial water in Tokyo. *Desalination*, 17: 267.
13. Jonsson, G. (1986) Transport phenomena in ultrafiltration: membrane selectivity and boundary layer phenomena. *Pure Appl. Chem.*, 58: 1647.

14. Jonsson, G. and Boesen, C.E. (1977) Concentration polarization in a reverse osmosis test cell. *Desalination*, 21: 1.
15. Jonsson, G. and Christensen, P.M. (1986) *Separation Characteristics of Ultrafiltration Membranes*; pp. 179–190; Drioli, E. and Nakagaki, M. (eds.); Membranes and Membrane Processes: Plenum New York, USA.
16. Kulkarni, S.S., Funk, E.W., and Li, N.N. (1992) *Ultrafiltration Membrane Handbook*; New York, USA.
17. Christopher, T.C., Thomas, F.S., and Amy, K.Z. (2002) Standardized membrane pore size characterization by poly(ethylene glycol) rejection. *J. Environ. Eng.*, 128: 399.
18. Akgemci, E.G., Ersoz, M., and Atalay, T. (2004) Transport of formic acid through anion exchange membranes by diffusion dialysis and electro-electro dialysis. *Sep. Sci. Tech.*, 39: 165.
19. Flory, P.J. (1953) *Principles of Polymer Chemistry*; Cornell University Press: New York, USA.
20. Shen, J.S. and Probstien, R.F. (1977) On the prediction of limiting flux in laminar ultrafiltration of macromolecular solution. *Ind. Eng. Chem. Fund.*, 16: 459.
21. Tsapiuk, E.A. (1993) Determination of molecular weight retention curves of ultrafiltration membranes. *Khimiya i Tekhnologiya*, *Vody*, 15: 171.
22. Tsapiuk, E.A. (1989) On the possible cause of increasing the rejection of low molecular weight solutes in the course of the dynamic membranes formation. *Kaloidn. Zh.*, 51: 113.
23. Tsapiuk, E.A. (1992) Displacement of molecular weight retention curves of ultrafiltration membranes in gel formation regime. *Khimiya i Tekhnologiya Vody*, 14: 532.
24. Tsapiuk, E.A. and Bryk, M.T. (1993) An interpretation of the separation of low and high molecular weight solutes by ultrafiltration. *J. Membrane Sci.*, 79: 227.
25. Nakao, S., Nomura, T., and Kimura, S. (1979) Characteristics of macromolecular gel layer formed on ultrafiltration tubular membrane. *J. Amer. Inst. Chem. Eng.*, 25: 615.
26. Minnikanti, V.S., Dasgupta, S., and De, S. (1999) Prediction of mass transfer coefficient with suction for turbulent flow in cross flow ultrafiltration. *J. Membrane Sci.*, 157: 227.
27. Nguyen, Q.T., Aptel, P., and Neel, J. (1980) Characterisation of ultrafiltration membranes Part-II. Mass transport measurements for low and high molecular weight synthetic polymers in water solution. *J. Membrane Sci.*, 7: 141.
28. Pradanos, P., Arribas, J.I., and Hernandez, A. (1992) Hydraulic permeability, mass transfer and retention of PEGs in cross-flow ultrafiltration through a symmetric microporous membrane. *Sep. Sci. Tech.*, 27: 2121.