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Development of high flux thin-film composite membrane for water desalination: a statistical study using response surface methodology

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ABSTRACT

High flux thin-film composite reverse osmosis membranes for brackish water desalination have been fabricated by interfacial polymerization based on aromatic polyamide chemistry. A response surface methodology was used to optimize the concentrations of the monomers, 1,3-Diaminobenzene (MPDA) and 1,3,5-Benzenetricarbonyl trichloride (TMC), and a flux-enhancing additive, Dimethyl sulfoxide (DMSO). The membranes prepared showed a salt rejection of more than 95%. The membranes produced with DMSO additive exhibited a four- to five-fold higher flux rate as compared to the membranes without additive. Quadratic mathematical models have been proposed and verified using diagnostic plots, which adequately describe the flux rate and rejection ability within the limits of the factors investigated. The membrane rejection ability was contributed by a first-order effect of the membrane preparation parameters MPDA, TMC, and DMSO concentration, a quadratic effect of TMC and DMSO concentration, and an interaction effect between TMC and DMSO concentrations. For flux rate, first-order effect of TMC, MPDA, and DMSO concentration, and quadratic effect of MPDA concentration were significant model terms.

Keywords: Desalination; Dimethyl sulfoxide; High flux; Thin film composite membrane; Response surface methodology; Interfacial polycondensation

1. Introduction

Today, cross-linked fully aromatic polyamide is widely regarded as the most effective and reliable material for reverse osmosis (RO) application. Over the years, research efforts have resulted in tremendous improvements in the performance of these membranes [1]. The composite membranes developed till the late 80's already had four to five times larger water flux and five times higher product water quality than those of cellulose acetate membranes used for RO. Since 1987, membrane performance has been further drastically improved. On the basis of these developments, cross-linked fully aromatic polyamide composite membranes have emerged as the materials of choice for brackish water desalination.

The polyamide layer that determines the performance of thin film composite (TFC) membrane is formed by interfacial polycondensation (IP), and therefore the conditions of this polymerization may be expected to have a significant influence on the functional performance of the resulting membrane.

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Different additives in the aqueous phase have been tried to improve the water flux of the TFC membrane without significant loss of their salt rejection capability. These additives function by influencing one or more of the following: monomer solubility, diffusivity, hydrolysis, and protonation. Some capture the reaction by-product formed during IP [2]. Kuehne et al. [3] reported that the addition of the organic salt of triethylamine with camphorsulfonic acid could considerably increase the water flux, with no loss of salt rejection ability. The use of hydrophilic compounds as additives during interfacial polymerization is another means of increasing the TFC membrane water flux [3]. Kong et al. could increase the water flux of TFC polyamide membrane by adding acetone in the organic phase [4]. Patent literature discloses the use of alcohol, ethers, water-soluble polymers, or polyhydric alcohols in the aqueous phase to improve membrane permeability [5–8]. Effect of DMSO additive on the structural properties of TFC membranes has also been reported [9,10]. These studies highlight the potential of additives in improving the performance of polyamide RO membranes. However, it is to be noted that additives that improve the water flux also have the potential to decrease the salt rejection. It is therefore necessary to carefully optimize the concentration of additives used in synthesis steps. Further, the monomer concentrations themselves can also influence properties of the membrane, such as hydrophilicity, and therefore it becomes necessary to study the effect of additives in the context of other preparation parameters.

Response surface methodology (RSM), in conjunction with experimental designs (DoE), has been extensively used in the membrane literature to identify significant variables that influence membrane performance and to arrive at optimal combination of synthesis variables. RSM is a statistical/mathematical methodology that can be used for studying the effect of several factors at different levels and their influences on each other [11]. The objective of RSM is to optimize the response based on the factors investigated. Idris et al. [12] employed RSM to predict the optimum composition of the aqueous phase for production of TFC membrane. RSM was also used by Xiangli et al. [13] to optimize the preparation conditions of polydimethylsiloxane/ceramics composite membranes. The proposed RSM regression model in that study showed that the polymer concentration was the most significant variable among the three, so far as the influences on permeation and rejection rate were concerned. Ismail and Lai [14] presented RSM and 2^k factorial design to optimize dope formulation (polymer concentration; solvent ratio) and preparation conditions (forced convective evaporation time; casting shear rate) for asymmetric polysulfone membrane preparation. Yi et al. [15] used RSM in conjunction with a central composite rotatable design to manipulate the preparation conditions of vinyltriethoxysilane-modified silicate/polydimethylsiloxane hybrid membranes. The result showed that the main effect of silicate loading was the most significant factor that influenced the hybrid membrane's selectivity followed by the quadratic effect of silicate loading, the main effect of cross-linker/prepolymer weight ratio and polymer concentration.

In this paper, a systematic investigation aimed at increasing the flux of polyamide TFC membranes is carried out using RSM. This study employs DMSO as an additive, and for the first time highlights the effect of concentration of the additive at varying levels of monomer concentrations. RSM-based modeling is the methodology employed. A three-factor central composite design (CCD) has been used for the experiments and the results of statistical analysis of the data thus obtained have been combined with RSM-based modeling. The conclusions allow an optimization of membrane performance for brackish water desalination, as determined by the permeation flux and salt rejection.

2. Experimental

2.1. Materials and methods

Polysulfone (PSF) base support membrane was supplied by Dow Chemicals (USA). MPDA and TMC were purchased from Aldrich Chemicals Co. (USA). DMSO, n-Hexane, Triethylamine, Camphorsulfonic acid, and Sodium carbonate were procured from Merck & Co. (USA). All the chemicals were used without further purification.

Calculated amounts of DMSO, camphor sulfonic acid and triethylamine were added in water and sonicated for 5 min. To this solution, the required amount of MPDA was added. TMC solution of the required concentration was prepared by dissolving TMC in n-Hexane. TFC membranes were fabricated as follows: the PSF base support was soaked with MPDA solution for 3 min. The excess solution was drained for 8–10 min and the PSF membrane was contacted with TMC solution in hexane for 50 s. After draining TMC solution for 15 s, the membrane was heat treated at 80 °C for 5 min. The resulting TFC membrane was first washed with hot water at 50 °C for 3 min and subsequently washed with a solution of sodium carbonate (0.2 w/v%) for 3 min.

The desalination performance of the membranes was determined using a flat sheet cross-flow

permeation cell (Sterlitech Corporation, USA) with an active area of 42 cm^2. A feed solution of 2000 ppm NaCl was passed at a feed-side pressure of 1.55 MPa. To get a stable operation and constant values of rejection and flux rate, the cell was operated for 1 h. After this period, the permeate was collected to calculate the flux rate. The concentrations of NaCl in permeate (C_p) and feed (C_f) were measured using a previously calibrated conductivity meter. The salt rejection (R_s) of membrane was calculated as follows:

$$R_s(\%) = \left(1 - \frac{C_p}{C_f}\right) \times 100 \quad (1)$$

2.2. Experimental design

Investigation of formulation compositions for TFC membrane preparation was carried out using CCD for RSM. RSM allows for the statistical analysis and modeling of a process in which a response of interest may be influenced by several variables and can be used to determine the optimum set of such variables [16]. Further, it also helps us to obtain the surface contour, which provides a good way for visualizing interaction among the factors studied. The three key steps in RSM are experimental design, model analysis, and condition optimization. Experimental designs, such as CCD, are convenient when the number of variables and levels is not too large because, under such circumstances, they do not require an excessive number of experimental runs. For example, the total number of experiments to be performed in 3-factor 2-level CCD study is given as sum of the 2^3 runs (factorial design), 2×3 axial or star runs, and 3 center runs [17].

In this study, RSM for TFC membrane preparation was carried out using three independent process variables, namely: TMC concentration (0.08–0.17 w/v%), MPDA concentration (1.83–2.67 w/v%), and DMSO concentration (0.49–3.01 w/v%). A previous study [18] exploring the effect of monomer concentrations in the absence of any additives, also based on CCD and RSM, helped fix the ranges for monomer concentrations in this study. The RSM designed in this study was based on CCD in which the factorial portion was a full factorial design with all combinations of the three factors at two levels, where the factor levels are coded to the usual low (−1) and high (+1) values, the axial or star points for which all but two factors were set at level 0, and the one factor was set at the outer value corresponding to an alpha value of 1.682. The center points (coded level 0), which were the midpoints between the high and low levels, were repeated three times to provide an estimate of the experimental error variance. The design involved 17 runs and the response variables measured were the flux ($lm^{-2}d^{-1}$) and the rejection (%). The operating ranges and the levels of the variables considered are shown in Table 1.

3. Results and discussion

3.1. Preparatory investigation of DMSO effect on membrane performance

Screening experiments were performed to identify the design variables that have a significant effect for further investigation. The values of MPDA (2 w/v%) and TMC (0.1 w/v%) concentrations used in these experiments were chosen based on the earlier RSM study [18] mentioned above on the effect of these variables without any additives. Fig. 1 shows the flux and rejection rate of TFC membrane produced at different DMSO concentrations in aqueous phase. A significant increase in flux was observed with increasing content of DMSO. The salt rejection remained unchanged for DMSO concentration up to 2 (w/v%), but showed a decreasing trend at higher levels. The flux of membranes obtained using DMSO could be further improved when salts of camphorsulfonic acid and triethylamine (CSA–TEA) were used as additives. Further, addition of CSA–TEA improved vastly, the consistency and reproducibility in flux and rejection rate of the membranes synthesized. A 3.4 w/v% concentration of TEA:CSA (1.1:2.3) showed an improvement in flux and a high rejection value of 98.5%. With this preliminary result, a CCD was employed to study the effect of monomer concentrations and DMSO concentration, and their interactions, on membrane performance.

3.2. Response surface and Analysis of variance (ANOVA) analysis

The responses of CCD were systematically examined using Design Expert version 8.0.3.1 (State-Ease

Table 1
Values of the processing parameters at different levels in the CCD employed

Factors (w/v%)	Real values of coded levels				
	$-\alpha$	−1	0	+1	$+\alpha$
(*A*) TMC concentration	0.08	0.1	0.13	0.15	0.17
(*B*) MPDA concentration	1.83	2.0	2.25	2.5	2.67
(*C*) DMSO concentration	0.49	1.0	1.75	2.5	3.01

Note: TEA:CSA was kept constant at 1.1: 2.3 (w/v%) for all membrane formulations.

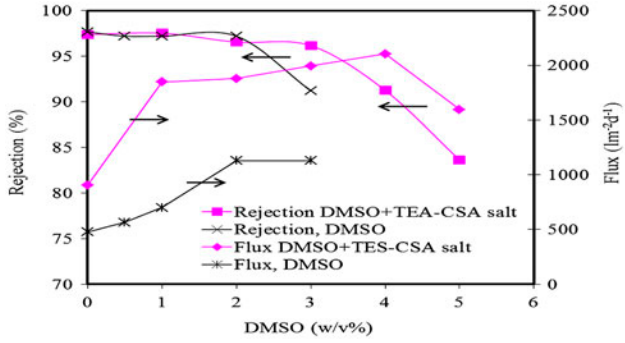

Fig. 1. Screening experiments showing effect of DMSO concentration (with and without the salt of CSA–TEA) on membrane flux and rejection.

Inc., Minneapolis, MN, USA, Trial Version) to study the effect and interactions of (*A*) TMC, (*B*) MPDA, and (*C*) DMSO concentrations on membrane permeation flux and rejection. The CCD layout and corresponding responses are summarized in Table 2. Contour plots were fitted to analyze the interaction between independent process factors and the desired responses, based on a statistical analysis of the experimental data. Any interactions among the membrane preparation parameters – TMC concentration, MPDA concentration, and DMSO concentration – which could induce significant effects on membrane performance were also determined.

3.3. Statistical models to aid selection of membrane synthesis recipes

Following the above procedure, the model summary statistics suggested a quadratic model for both response variables, viz., membrane permeation flux and rejection rate. To reduce the insignificant model terms and/or large block effect, a backward elimination regression was followed with an alpha exit value of 0.1 to get reduced quadratic models for the response surfaces.

Table 3 shows the analysis of variance for the reduced quadratic model for membrane permeation flux. The first-order effect of TMC concentration (*A*), MPDA concentration (*B*), DMSO concentration (*C*), and quadratic effect of MPDA concentration (B^2) showed up as significant model terms, where prob > *F* values were less than 0.05. The model *F*-value of 21.41 and corresponding value of Prob > *F* (<0.0001) implied that the model is significant, in that there is only a 0.01% chance that a model *F*-value this large could occur due to noise. The lack-of-fit *F*-value of 4.75 implies that the lack-of-fit is not significant relative to pure error. There is 18.25% chance that a lack-of-fit *F*-value this large could occur due to noise. The empirical model of permeation flux (Table 4) showed good validity and reliability as the value of regression coefficient (R^2), 0.8771, was reasonably close to 1. Predicted R^2 of 0.7591 is in reasonable agreement with

Table 2
Design layout and corresponding response of CCD

Run no.	Variable factors						Response	
	TMC (w/v%)	Level A	MPDA (w/v%)	Level B	DMSO (w/v%)	Level C	Rejection (%)	Flux ($lm^{-2}d^{-1}$)
1	0.15	+1	2	−1	1	−1	94.64	2,768
2	0.125	0	2.25	0	1.75	0	96.1	3,396
3	0.125	0	2.25	0	3.01	+α	93.6	3,396
4	0.125	0	2.67	+α	1.75	0	96.9	3,453
5	0.125	0	2.25	0	0.49	−α	97.41	2,910
6	0.125	0	2.25	0	1.75	0	95.9	3,287
7	0.1	−1	2	−1	2.5	+1	94.52	3,167
8	0.125	0	2.25	0	1.75	0	97.2	3,340
9	0.1	−1	2	−1	1	−1	97.2	2,996
10	0.125	0	1.82	−α	1.75	0	96.52	2,882
11	0.1	−1	2.5	+1	1	−1	97.3	3,253
12	0.1	−1	2.5	+1	2.5	+1	95.32	3,738
13	0.15	+1	2.5	+1	1	−1	95.76	2,939
14	0.15	+1	2	−1	2.5	+1	94.36	3,224
15	0.15	+1	2.5	+1	2.5	+1	95.22	3,396
16	0.083	−α	2.25	0	1.75	0	95.48	3,196
17	0.17	+α	2.25	0	1.75	0	93.62	2,739

Table 3
ANOVA table for flux rate

Source	SS	DF	MS	*F*-Value	Prob. > *F*
Model	1042368.29	4	260592.07	21.41	<0.0001 significant
A-MPDA	332613.6	1	332613.6	27.32	0.0002
B-TMC	186417.26	1	186417.26	15.31	0.0021
C-DMSO	416982.75	1	416982.75	34.26	<0.0001
B^2	106354.68	1	106354.68	8.74	0.012
Residual	146074.18	12	12172.85		
Lack-of-fit	140132.18	10	14013.21	4.72	0.1875 not significant
Pure error	5,942	2	2,971		
Cor total	1188442.47	16			

Note: SS: sum of square; DF: degree of freedom; MS: mean square.

the Adjusted R^2 of 0.8367. Adequate precision measures the signal-to-noise ratio. A ratio greater than 4 is desirable. The value of Adequate precision of 14.96 indicated an adequate signal. The larger the *F*-value and the smaller the prob > *F*, the more significant the corresponding factor is. The ranking of the significant factors was $C > A > B > B^2$.

For membrane rejection ability as the response variable, the results of quadratic model in the form of analysis of variance are given in Table 5. As can be seen from this table, the model *F*-value of 17.27 and the low probability value (Prob > 0.0001) indicate that the model is very significant. The membrane rejection ability was contributed by the first-order effect of the membrane preparation parameters–MPDA concentration (*A*), TMC concentration (*B*), DMSO concentration (*C*), quadratic effect of TMC concentration (B^2) and DMSO concentration (C^2), and interaction effect between TMC concentration and DMSO concentration (*BC*). These parameters have significant model terms, where prob > *F* values were less than 0.05. The analysis of variance shows that $p < 0.05$ was statistically significant with the 95% confidence level in the range studied. The lack-of-fit analysis shows $p > 0.05$ and indicates that there was an adequate goodness-of–fit. The significance ranking in this study was $C > B^2 > B > BC > C^2 > A$. The empirical model (Table 4) was satisfactory and has shown reasonable validity and reliability for membrane rejection prediction, with R^2 (0.9120) and Adjusted R^2 (0.8592). The Adequate precision ratio of 13.04 indicates an adequate signal and shows that this model can be used in the design space to predict the membrane rejection ability.

Table 4
Summary of ANOVA and regression analysis for membrane flux rate and rejection ability

Response model	R^2	Adjusted R^2	Predicted R^2	Adequate precision
For flux rate				
Quadratic model	0.8771	0.8361	0.7591	14.9621
For rejection ability				
Quadratic model	0.912	0.8595	0.7550	13.04

The model for permeation flux (J, $\mathrm{lm^{-2}d^{-1}}$) and rejection ability ($R_{s,\%}$) in terms of coded variables have been expressed by the following equations:

$$J = 3253.76 + 156.06 \times A - 116.83 \times B + 174.74 \times C - 90.35 \times B^2 \quad (2)$$

$$R_s = 96.54 + 0.26 \times A - 0.55 \times B - 0.87 \times C + 0.48 \times BC - 0.69 \times B^2 - 0.35 \times C^2 \quad (3)$$

In terms of actual factors, the empirical model were

$$J = -233.08 + 624.24 \times \text{MPDA concentration} + 31466.42 \times \text{TMC concentration} + 232.98 \times \text{DMSO concentration} - 144,559 \times \text{TMC concentration}^2 \quad (4)$$

$$R_s = 85.54 + 1.03 \times \text{MPDA concentration} + 207.79 \times \text{TMC concentration} - 2.19 \times \text{DMSO concentration} + 25.6 \times \text{TMC concentration} \times \text{DMSO concentration} - 1,098 \times \text{TMC concentration}^2 - 0.62 \times \text{DMSO concentration}^2$$

These models can be used to predict the membrane permeate flux and rejection coefficient within the limits of experimental parameters employed in this study.

Table 5
ANOVA table for response rejection ability of membrane

Source	SS	DF	MS	*F*-value	Prob. > *F*
Model	23.5	6	3.92	17.27	<0.0001 significant
A-MPDA	0.91	1	0.91	4	0.0734
B-TMC	4.11	1	4.11	18.1	0.0017
C-DMSO	10.35	1	10.3	45.63	<0.0001
BC	1.84	1	1.84	8.13	0.0172
B^2	5.82	1	5.82	25.65	0.0005
C^2	1.5	1	0.23	6.62	0.0277
Residual	2.27	10	0.23		
Lack-of-fit	1.29	8	0.16	0.33	0.896 not significant
Pure error	0.98	2	0.49		
Cor total	25.8	16			

3.4. Verification of regression models on diagnostic plot

A plot of the normal% probability vs. residual is shown in Fig. 2 for flux rate and rejection ability. The normal probability plot of the residuals is an important diagnostic tool in the residual analysis of response surface design and is useful to check the assumptions of normality of distribution of errors and their independence from each other, which are made in the analysis. As seen from the figure, there is no obvious indication of non-normality of the experimental results, as most of the residuals fall close to the diagonal line.

A plot of studentized residual vs. predicted response is shown in Fig. 3 and shows comparable scatter above and below the x-axis, thus indicating the absence of any serious pattern and unusual structures. From the figure, it is apparent that the model proposed by RSM analysis is adequate. There is no reason to suspect any violation of the assumptions of independence or constant variance.

Fig. 4 shows a plot of predicted vs. actual values of the response variables. They were compared to check the goodness-of-fit of the model via the correlation coefficient (R^2). The figure shows that the empirical models are reliable to predict the membrane performance in terms of permeation flux and rejection.

3.5. Membrane performance analysis

The effect of MPDA, TMC, and DMSO concentrations interaction on variable response for membrane permeation flux is shown in Fig. 5 in the form of three-dimensional response surface and contour plots. As seen from the figure, high flux TFC membranes could be synthesized by choosing high MPDA and DMSO concentration, and low TMC concentration.

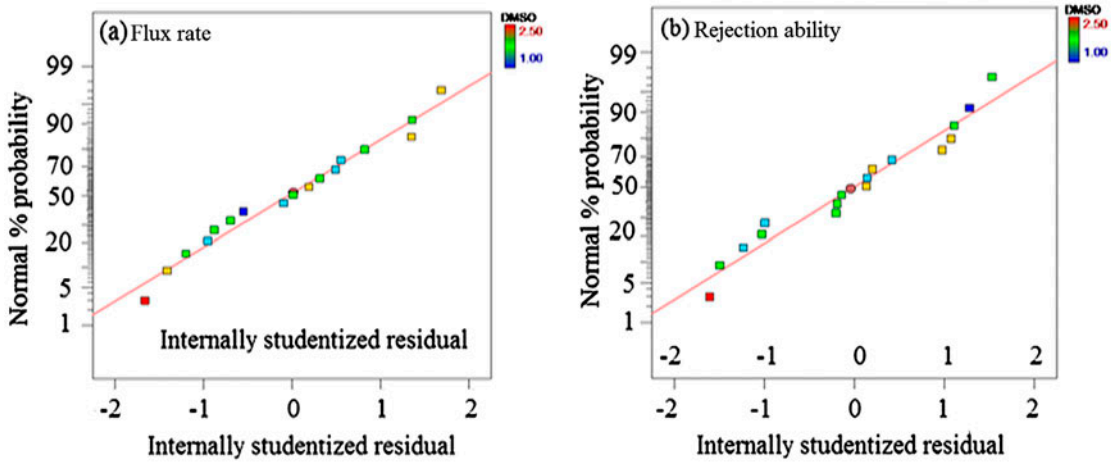

Fig. 2. Plots of normal probability vs. residual.

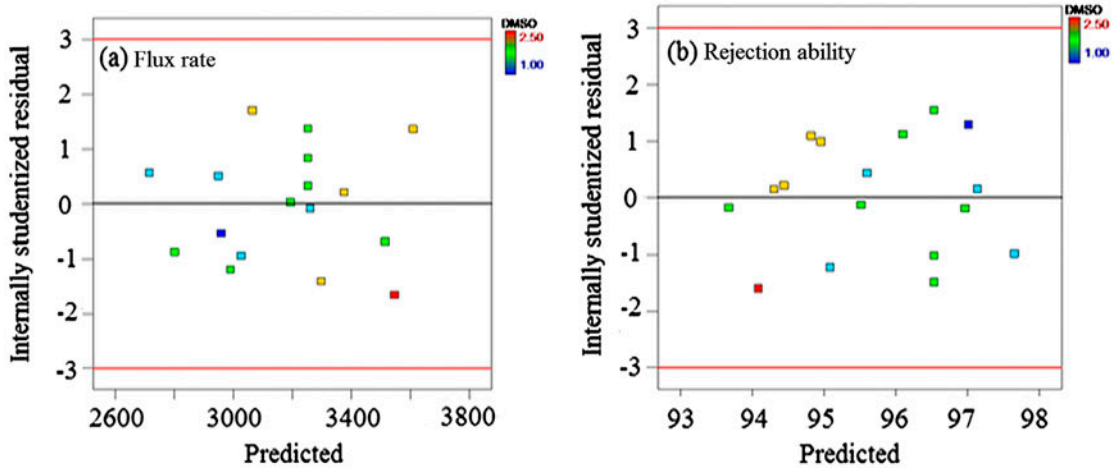

Fig. 3. Plot of residual vs. predicted response.

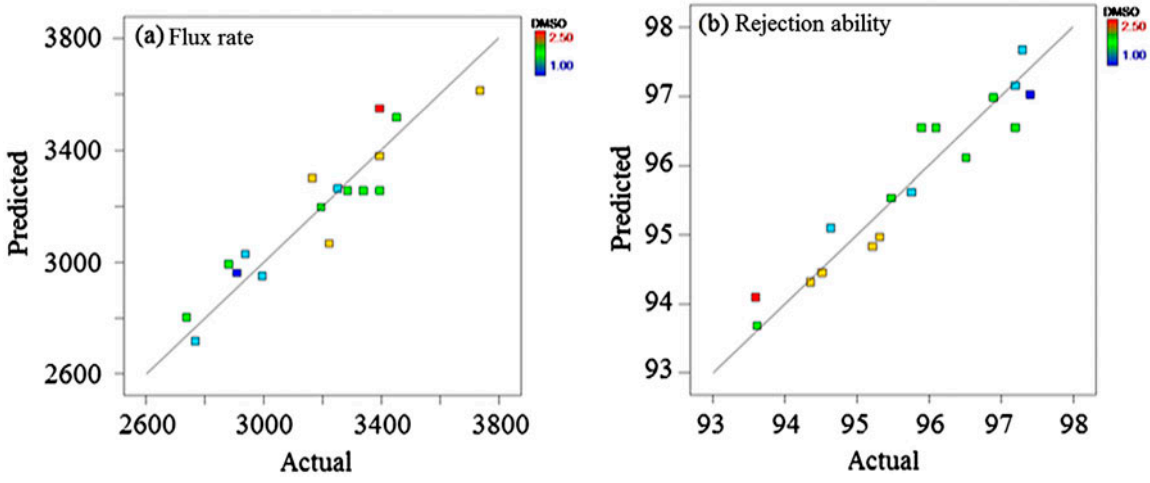

Fig. 4. Plot of predicted vs. actual flux rate and rejection ability.

MPDA concentration has a positive effect on membrane permeation flux in presence of DMSO. A linear increase of permeation flux with the MPDA concentration was observed. The phenomenon of increasing flux of TFC membranes with increase in MPDA concentration without any additives in aqueous phase was reported in [18]; however, the flux of these membranes were four to five times lesser than that obtained here using DMSO additive. The rise in permeation flux rate with MPDA concentration is probably a result of (i) decreases in width of IP reaction zone, which in turn results in a decrease in the PA layer thickness, and (ii) formation of a loose network [19,20]. Membranes fabricated with high TMC concentrations exhibit lesser permeation flux. This fact may be similarly attributed due to increases in thickness and cross-link density of PA layer with increase in TMC concentration [21]. The effect of increasing DMSO concentration in aqueous phase resulted in higher permeation flux of membrane. The increase in flux with DMSO concentration could be explained by considering that water permeation of TFC membranes takes place in two types of pores, network pores and aggregate pores. The observed effect can thus be explained by (i) an increase in the size and the number of network pores, and (ii) an increase in the size of aggregate pores [22].

The surface and contour plots for rejection ability of TFC membranes as a function of MPDA, TMC, and DMSO concentrations are shown in Fig. 6. Salt transport in solution-diffusion membranes is a complex phenomenon and is still a subject of fundamental study. Selectivity is governed by pore-size distribution, porosity, and specific interactions within the pore fluid. The radius of hydrated Na^+ and Cl^- is ca 4.75 Å [22]. If there are pores larger than this value (large

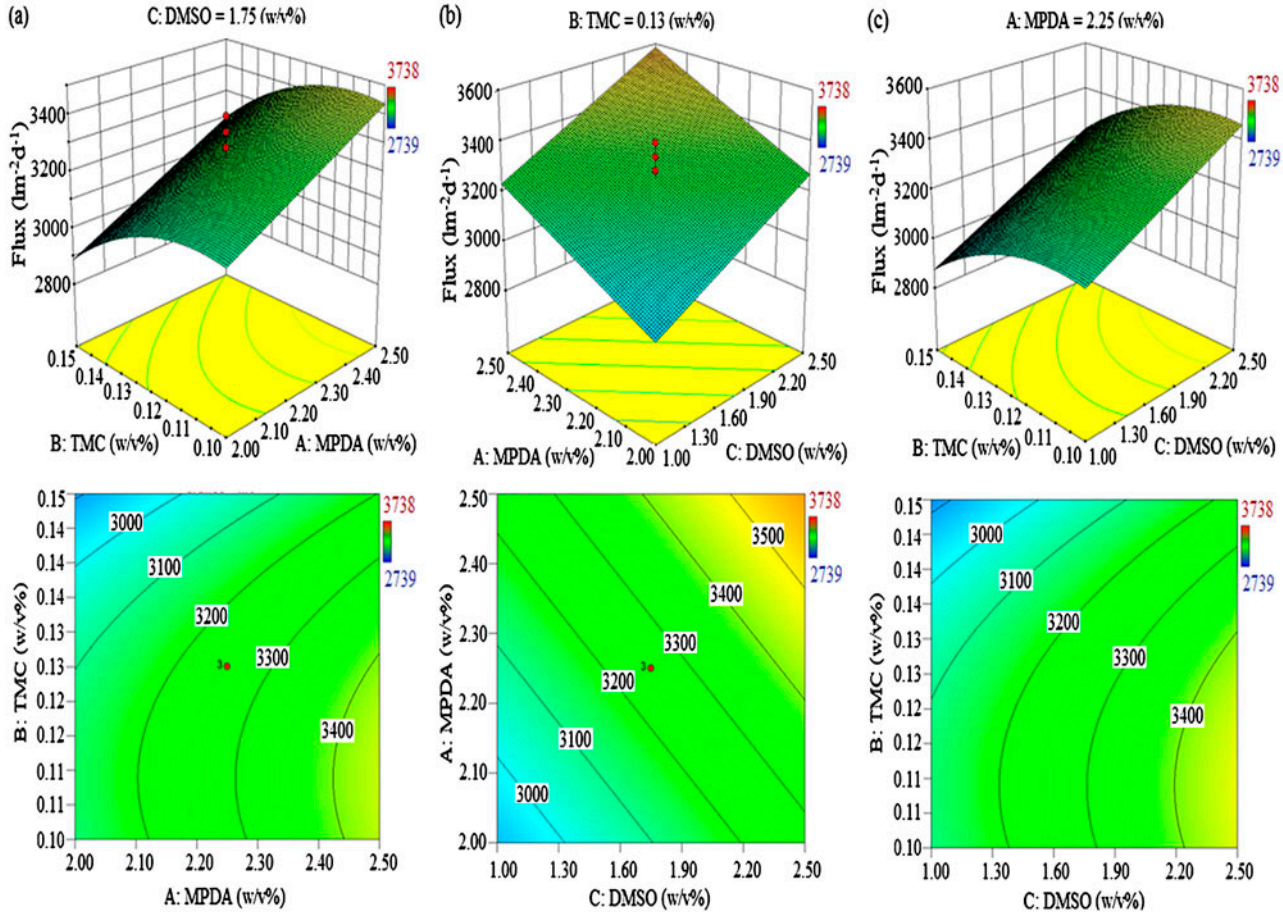

Fig. 5. Response surface and contour for membrane flux rate plotted on (a) TMC concentration: MPDA concentration, (b) MPDA concentration: DMSO concentration, and (c) TMC concentration: DMSO concentration.

defect pores), some direct flow of solute can occur. The nonadditive version of polyamide, BW-30, and laboratory-prepared TFC membranes showed high salt rejection abilities. This is because the pores are smaller than the size of hydrated ion. The use of DMSO additive during membrane formation process, however, increases not only the number of pores but also the size of pores. This is the reason why selectivity of TFC membranes deteriorates with an increase in DMSO concentration in aqueous phase [23].

3.6. Confirmation experiments

In order to confirm the model, three confirmation experiments were performed. Table 6 shows the conditions and the experimental results. The predicted values of the flux rate and salt rejection were calculated according to Eqs. (4) and (5), respectively. The percent error between the actual and predicted values for the flux rate and salt passage varied from −0.20 to 4.73 and −0.31 to 0.11, respectively. The results of the confirmation experiments, thus, indicate that the regression models obtained were reasonably accurate and can be used to predict the salt rejection and flux rate of polyamide TFC membranes.

3.7. Optimal synthesis parameters and benchmarking against commercial membranes

The visualization of surface and contour plots (Figs. 5 and 6) suggest that the membranes produced with MPDA, TMC and DMSO concentration in the range of 2–2.7 (w/v%), 0.1–0. 13 (w/v%), and 0.5–1.75 (w/v%), respectively, produced TFC membranes which showed highest flux rate in the range 2,880–3,450 $lm^{-2}d^{-1}$ with rejection rate of 96.5–97.4%. To see how these values compare against commercially available desalination membranes for brackish water service, a BW30 (FilmTec Corporation) was procured from Dow Chemicals and its performance was evaluated using the same test cell and test conditions as employed in this work. The values obtained were

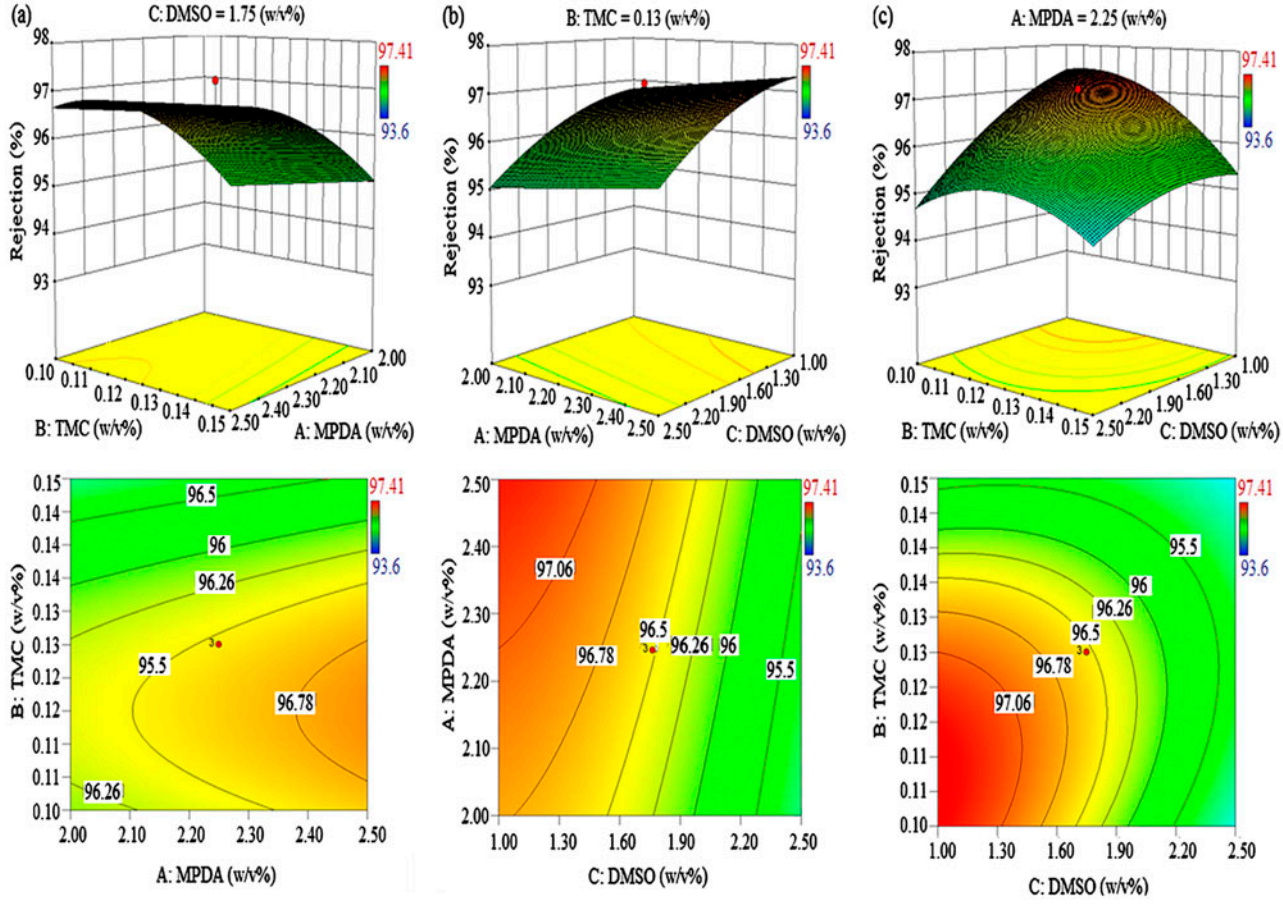

Fig. 6. Surface and contour for rejection ability of membrane plotted on (a) TMC concentration: MPDA concentration, (b) MPDA concentration: DMSO concentration and (c) TMC concentration: DMSO concentration.

Table 6
Experimental conditions and results for confirmation experiments

Factors			Predicted		Actual			
TMC (*A*)	MPDA (*B*)	DMSO (*C*)	J ($lm^{-2}d^{-1}$)	Rs (%)	J ($lm^{-2}d^{-1}$)	Error (%)	Rs (%)	Error (%)
0.125	2.25	1.75	3,254	96.5	3,100	4.73	96.8	−0.31
0.1	2	1	2,949	97.2	2,956	−0.2	97.3	−0.1
0.15	2.25	2.5	3,377	94.82	3,298	2.33	94.93	0.11

1,100 $lm^{-2}d^{-1}$ for water flux and 97.4% for salt rejection, respectively. Thus, the membranes fabricated in this work are comparable in terms of salt rejection but much superior in terms of water flux.

4. Conclusions

The main objective of this study was to investigate the effect of monomer concentrations and that of a flux-enhancing additive, DMSO, on TFC membrane performance, and hence to evolve a methodology for optimizing the preparation recipe for such membranes. The results revealed that the TFC membranes for brackish water desalination, with performance superior to commercially available ones, could be successfully fabricated using DMSO additive. The membranes showed enhanced water flux with salt rejection ability in the range of RO. A RSM was used to study the effect, individually and in combination, of the concentrations of the monomers and the additive on membrane performance, and hence to identify the values for optimal membrane performance.

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