A green membrane distillation system for seawater desalination: Response surface modelling and optimization

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Abstract. In this work, response surface methodology (RSM) was applied for modelling and optimization of operating parameters for water desalination by direct contact membrane distillation (DCMD) system using cost effective and green ceramic hollow fibre membrane prepared from rice husk waste. Operating parameters including feed temperature, permeate temperature and feed flow rate were selected and the optimum parameters were determined for DCMD salt rejection. The developed model for permeate flux response was statistically validated by analysis of variance (ANOVA) which showed a high value coefficient of determination value ($R^2 = 0.9954$). In this study, the obtained optimum operating parameters were found to be 70.15°C of feed temperature, 19.84°C permeate temperature and 0.58 LPM of feed flow rate.

1. Introduction

“Fresh water is the world’s first and foremost medicine”. The world is poised at the brink of a severe global crisis especially lack of fresh water. Accordingly, seawater as alternative source for fresh water has been explored due to its large percentage (>97%) of total water available on earth, leading increasing number of studies on seawater desalination. Among all technologies, membrane distillation (MD) system has received increasing attention, especially after the rapid advances in MD membranes and solar energy. There are four main configuration in MD [1]: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD), and vacuum membrane distillation (VMD). Consequently, DCMD is the most studied configuration because the condensation step is carried out inside the membrane module leading to a simple operation mode without the need of external condensers like in SGMD and VMD configurations. In DCMD configuration, the hot feed solution is in direct contact with the inlet membrane side surface. Therefore, evaporation takes place at the feed membrane surface. Vapor is moved by the pressure difference across the membrane to the permeate side and condenses inside membrane module [2]. Because of the hydrophobic nature of the membrane, the liquid feed cannot penetrate inside dried membrane pores. Up to the present, DCMD has been applied, in major part, for the production of fresh water, in wastewater treatment and reuse and in the food industry.
The major part of the reported researches on membrane distillation was carried out using conventional method of experimentation, changing one of the independent parameters while maintaining the others at fixed levels. Conventional method involves an elevated number of experimental runs, which are time consuming, expensive and ignore the interaction effects between the dominant parameters affecting the performance of the membrane distillation process. Research Surface Methodology (RSM) has been applied successfully in scientific and research areas such as chemistry, physics, biochemistry, biology, environment, etc. In the field of membrane distillation, few studies have been carried out using RSM.

Khayet et al. [3] used the RSM for the optimization of DCMD process together with prediction of permeate fluxes when using NaCl salt aqueous solution. The considered factors were the stirring velocity, the feed temperature and the NaCl concentration. It was found that the DCMD parameters affect the permeate flux in the same way as observed from the classical method of experimental flux. There is a good agreement between the experimental and the predicted permeate fluxes by RSM. However, the use of polymeric membranes, like PVDF, is severely restricted for the long term operation at elevated temperatures or drastic chemical conditions. Thus, ceramic membranes offer several advantages over polymeric membranes especially due to its non-swelling behaviour and thermal stability, which is desirable in MD application [4-6]. In our previous work [7], we have successfully prepared a cost effective and green ceramic hollow fibre membrane derived from rice husk waste and prepared through phase inversion and sintering technique. In was revealed that a very highest flux more than 35 kg/m²h (data in review), comparable to the polymeric one, could be obtained. Unfortunately, there is some drawbacks using ceramic membrane in MD which is its poor wettability properties. Other than its hydrophobicity/hydrophilicity effect, other major reason is due to the microfiltration pores of ceramic membrane that can lead salt in feed solution to pass through. Thus, the objectives of this study is to model and optimize the DCMD parameters using green ceramic hollow fibre membrane prepared from rice husk waste towards salt rejection performance.

2. Materials and Methods

2.1. DCMD experimental setup

Experiments were carried out using a green ceramic hollow fibre membrane (CHFM) made of rice husk waste that converted into silica through calcination process. Then, ceramic suspension containing silica based rice husk ash as main material, polyethersulfone (PESf) as binder and 1-methylpyrrolidone (NMP) as solvent. Tap water was used as both external and internal coagulation bath. After that, sintering process was conducted to produce final CHFM. The detail preparation of the CHFM was reported in our previous work [7]. Accordingly, the prepared CHFM was modified from its natural hydrophilicity into hydrophobic through FAS grafting process [8].

2.2. Experimental design and statistical analysis

In this study, the Central Composite Design (CCD) was applied for the optimization of DCMD process used for desalination of NaCl salt solution. This is due to the design that is suitable for fitting a quadratic surface and helps to optimize the effective parameters with minimum number of experiments, at the same time, analyzing the interaction between parameters. Three main parameters in DCMD have been chosen which are feed temperature, permeate temperature and feed flow rate, to study their interaction towards better salt rejection. In CCD, DCMD parameters and performances were termed as variables and responses, respectively. Consequently, mathematical function is assumed for the response in terms of the significant independent variables. A quadratic model corresponding to the following second order equation was built to describe the response [9]:

\[
Y = b_0 + \sum_i b_i X_i + \sum_i b_i^2 X_i^2 + \sum_{ij} b_{ij} X_i X_j
\]  

(1)
where $Y$ is the response, $b_o$ the constant coefficient, $b_i$ the linear coefficients, $b_a$ the quadratic coefficients, $b_j$ the interaction coefficients and $X_i$, $X_j$ the coded values of the variables.

The statistical significance of the models was justified through analysis of variance (ANOVA) for polynomial model with 95% confidence level, and residual plots were used to examine the goodness of models fit. The quality of the fit polynomial model was also expressed by the coefficient of determination $R^2$. In this study, three factors including feed temperature, permeate temperature and feed flow rate with five levels were employed for response surface modeling and optimization of DCMD process. According to CCD, a total number of 32 experiments have been performed with 3 replicate points to estimate the experimental error.

3. Results and Discussions
Among all desalination technologies, direct contact membrane distillation (DCMD) is a recent method that received worldwide attention due to its excellent desalination performance at cost which appears to be reasonable compared to reverse osmosis (RO) membrane [10]. In addition, newly-developed green ceramic membrane from waste rice husk was used in DCMD, for the first time, for desalination application. In DCMD process, the hot solution (feed) is in direct contact with the hot membrane side surface. Therefore, evaporation takes place at the feed-membrane surface. The vapour is moved by the pressure difference across the membrane to the permeate side and condenses inside the membrane module. Accordingly, an optimum parameters of the process such as feed temperature, permeate temperature and feed flow rate should be investigated to produce high water vapor flux. An optimization using Research Surface Methodology (RSM) method has been performed. Table 1 presents the regression model developed for the prediction of salt rejection using analysis of variance (ANOVA). It was observed that the model F-value of 20.67 implies the model is significant. In addition, value of Prob > F less than 0.0500 indicate that the model are significant. Therefore, in this case, C and $C^2$ which are both represent feed flow rate are significant model terms. The “lack of fit F-value” is 1.23 implies the lack of fit is not significant to the pure error There is a 33.85% chance that a “lack of fit F-value” this large could due to noise. Based on Myers et al., non-significant lack of fit presents a good model, which is obtained for this work [11]. It should be mentioned that the p-value obtained for this work is less than 0.05 (p-value = <0.0001). This means that the obtained salt rejection model is highly significant and the regression model gives a good prediction of the experimental data. The coefficient of determination ($R^2$), which is the proportion of variation in the response attributed for the regression model, should be close to 1. The $R^2$ of the regression model is 0.9954, meaning that more than 99.54% of the data deviation can be explained by the empirical model, which indicates that the regression model is statistically significant. Moreover, it is preferred to use the adjusted $R^2_{Adj}$ values to evaluate the model adequacy since it is adjusted for the number of terms in the model. The $R^2_{Adj}$ value is 95.1%, indicating a high degree of correlation between the experimental and predicted responses.

| Sources                | Sum of squares | df | Mean square | F-Value | P-Value |
|------------------------|----------------|----|-------------|---------|---------|
| Model                  | 106.90         | 9  | 11.87732399 | 20.67   | < 0.0001|
| A-Feed Temperature     | 0.00           | 1  | 0           | 0.00    | 1.0000  |
| B-Permeate Temperature | 0.00           | 1  | 0.002857143 | 0.00    | 0.9444  |
| C-Feed Flow Rate       | 66.88          | 1  | 66.88285714 | 116.38  | < 0.0001|
| AB                     | 0.72           | 1  | 0.72        | 1.25    | 0.2751  |
| AC                     | 0.00           | 1  | 0           | 0.00    | 1.0000  |
| BC                     | 0.00           | 1  | 0.005       | 0.01    | 0.9265  |
| A^2                    | 0.67           | 1  | 0.671836228 | 1.17    | 0.2913  |
Figure 1 shows the 3D response curves that presenting the interaction of the variables and optimum level of each variables and optimum level of each variable for maximum response. Figure 1(A) shows the response of salt rejection with varying permeate temperature and feed temperature. As can be seen, both factors have a slight effect on DCMD salt rejection. At center value of PT and FT a maximum response value was obtained, the salt rejection is 100% as calculated by regression model. Whereas, Figure 1(B) shows the responses of DCMD salt rejection with varying FT and Q_F. It is observed that the effect of feed flow rate (Q_F) on salt rejection is highly significant than the effect of feed temperature (FT). This is highly due to the pore size of ceramic membrane that in microfiltration range [12]. The combined effects of PT and Q_F on salt rejection is shown in Figure 1(C). As expected, the PT do not give any effect on salt rejection. As a result, similar trend also obtained by the effect of feed flow rate on salt rejection. Therefore, it was revealed that the feed flow rate (Q_F) has the major effect towards DCMD salt rejection.

To confirm the model adequacy and the validity of optimization procedure, experiments were carried out under optimized conditions and the results were compared with the predicted values of response using the model equation. The experiments were conducted in triplicates and mean values are reported. The optimization reveals that to obtain the highest 100% of salt rejection using hydrophobic ceramic membrane made from rice husk waste, the three main parameters in DCMD which are feed temperature, permeate temperature and feed flow rate should set at 70.15℃, 19.84℃ and 0.58 LPM, respectively. These results represent 0.5% as deviation between experimental (100%) and predicted values (100.519%). In fact, the highest salt rejection cannot be more than 100%, which means that the experimental value is the best value obtained. This indicates the suitability of the developed regression model and it may be noted that these optimal values are valid within the specified range of DCMD process variables.

Figure 1. 3D response surface plot of salt rejection as function of operating factors; (A) feed temperature vs permeate temperature, (B) feed temperature vs feed flow rate, and (C) permeate temperature vs feed flow rate
4. Conclusions
This study investigates the modeling and optimization of lab scale DCMD for aqueous NaCl salt solution for the purpose of seawater desalination application using experimental design and response surface methodology. Effects of operating conditions which are feed temperature, permeate temperature and feed flow rate, were studied to optimize DCMD salt rejection using ceramic membrane made from rice husk waste. As a result, an optimization has been obtained by setting the feed temperature, permeate temperature and feed flow rate of 70.15°C, 19.84°C and 0.58 LPM, respectively.

The response surface model for DCMD permeate flux was statistically validated by ANOVA and provided good quality to predict the response, whose $R^2 = 0.9954$ and $R^2_{Adj} = 0.9510$. It was also found that the feed flow rate giving the most significant effect towards DCMD salt rejection.

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Acknowledgement
The authors gratefully acknowledge the financial support from the Ministry of Education Malaysia under the Higher Institution Centre of Excellence Scheme (Project Number: R.J090301.7846.4J192), Universiti Teknologi Malaysia under the Research University Grant Tier 1 (Project number: Q.J130000.2546.12H25), Nippon Sheet Glass Foundation for Materials Science and Engineering under Overseas Research Grant Scheme (Project number: R.J130000.7346.4B218) and Kurita Water and Environment Foundation under KWEF Research Grant Program.