

# Direct contact membrane distillation applied to saline wastewater: parameter optimization

Sana Abdelkader, Ali Boubakri, Sven Uwe Geissen and Latifa Bousselmi

## ABSTRACT

Freshwater availability is increasingly under pressure from growing demand, resource depletion and environmental pollution. Desalination of saline wastewater is an option for supplying households, industry and agriculture with water, but technologies such as reverse osmosis, evaporation or electrodialysis are energy intensive. By contrast, membrane distillation (MD) is a competitive technology for water desalination. In our study, response surface methodology was applied to optimize the direct contact membrane distillation (DCMD) treatment of synthetic saline wastewater. The aim was to enhance the process performance and the permeate flux  $J_p$  ( $L/m^2 \cdot h$ ) by optimizing the operating parameters: temperature difference  $\Delta T$ , feed velocity  $V_f$ , salt concentration  $[NaCl]$ , and glucose concentration  $[Gluc]$ . The results are a high permeate quality, with 99.9% electrical conductivity reduction and more than 99.9% chemical oxygen demand (COD) removal rate. The predicted optimum permeate flux  $J_p$  was  $34.1 L/m^2 \cdot h$  at  $\Delta T = 55.2^\circ C$  and  $V_f = 0.086 m/s$ , the two most significant parameters. The model created showed a high degree of correlation between the experimental and the predicted responses, with high statistical significance.

**Key words** | desalination, direct contact membrane distillation, feed velocity, response surface methodology, temperature difference, wastewater

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## INTRODUCTION

The increasing domestic and industrial demand for clean fresh water is creating an ever-growing pressure on global security, since water has numerous interlinkages with all the aspects in our lives in terms of economic development, energy demand, environmental security and industrial growth. This pressure is becoming even more critical with climate change (UN World Water Development 2015).

Water desalination, and wastewater treatment and desalination in particular, could provide a valuable alternative to partially overcome the need for more water resources by reusing the treated discharged effluents and integrating them into industrial or domestic water cycles via multiple desalination technologies such as membrane distillation (MD). MD offers the possibility of producing high-quality permeates with significant water recovery rates with lower energy consumption compared to the conventional and well-established desalination processes such as reverse osmosis, multiple effect distillation, etc. (Miller 2003; Samblebe 2006).

Based on its high separation performance, MD is being investigated and applied for water, and in some cases, nutrient recovery from various types of effluents, taking into consideration the effect of experimental parameters (Izquierdo-Gil *et al.* 1999; Jia *et al.* 2017). In addition to high salt rejection rates, MD is characterized by good rejection rates of non-volatile and low adsorptive organic compounds (Carnevale *et al.* 2016; Plattner *et al.* 2017). The enhancement of MD has drawn attention to the improvement of membrane hydrophobicity using multiple techniques such as the use of hydrophobic surface-modifying macromolecules on poly(vinylidene fluoride) hydrophobic composite membranes, which has led to promising results from MD treatment of sea water (Prince *et al.* 2014a). Moreover, multiple studies have focused on the enhancement of the driving force for water vapor transport in MD through the development of hydrophobic membranes that have high resistance to pore wetting, which has led to significantly higher flux

doi: 10.2166/wst.2018.274

levels along with high rejection rates (Prince *et al.* 2012, 2014b; Efome *et al.* 2016). The interest in MD systems has led to more emphasis on process optimization in order to achieve the highest possible permeate fluxes with higher yields and distillate quality. In their study, Zhang *et al.* (2016) found that the optimization of saline wastewater treatment by two-stage vacuum MD using polypropylene (PP) hollow fiber membrane increased the water recovery percentage to 88.6%, with an improvement of the permeate quality in terms of electrical conductivity and total suspended solids removal, in comparison to one-stage vacuum MD.

Some mathematical theoretic models have been developed to simulate the MD process for multiple membrane module configurations in order to enhance their performance (Laganà *et al.* 2000; Boukhriss *et al.* 2015; Deshpande *et al.* 2017). According to Khayet *et al.* (2007), the application of response surface methodology (RSM) to optimize salt aqueous solution treatment by direct contact membrane distillation (DCMD) was suitable for assessing the permeate fluxes for both commercial- and laboratory-made membranes. Moreover, they proposed an algorithm that can help in the searching step for optimum localization considering factors such as the stirring rate, the feed temperature and the salt concentration. Another optimization study carried out by Cheng *et al.* (2016) using RSM (via the application of quadratic rotation-orthogonal composite design) showed that both operating conditions and membrane module parameter optimization can lead to a remarkable improvement in process performance. The recorded enhancements were in average permeate flux, water productivity per unit volume of module, water production per unit energy consumption, and comprehensive index.

Within this framework, our study investigated the optimization of synthetic saline wastewater treatment by DCMD by maximizing the process response in terms of permeate flux through the application of RSM. The optimum levels of the independent operating variables, namely, the temperature difference, feed velocity, NaCl concentration and glucose concentration were investigated.

## MATERIAL AND METHODS

### Wastewater composition

The synthetic saline wastewater solution used in this paper was composed of  $\text{NH}_4\text{Cl}$  (1 g/L),  $\text{KH}_2\text{PO}_4$  (0.3 g/L),  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  (2 g/L),  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$  (0.2 g/L), and  $\text{C}_2\text{H}_5\text{NaO}_2 \cdot 3\text{H}_2\text{O}$  (1 g/L) (Kapdan & Erten 2007). In addition, various concentrations of salt (NaCl) and glucose ( $\text{C}_6\text{H}_{12}\text{O}_6$ ) were added to simulate different saline wastewater compositions. The chosen ranges of variation of COD and [NaCl] had average levels and were as follows: the chemical oxygen demand (COD) varied from 0.3 to 10  $\text{gO}_2/\text{L}$  and the [NaCl] from 10 to 30 g/L. The COD and [NaCl] variation ranges included different effluent qualities in terms of salinities and organic loads (Kapdan & Erten 2007). A new feed solution was prepared for each experiment and all chemical reagents used were analytical grade.

### DCMD unit and experimental parameters

The DCMD apparatus used in this study was a bench-scale DCMD set-up composed mainly of one heating and one cooling thermostatic water bath connected to the membrane cell via two peristaltic pumps. The two thermostatic cycles (heating closed loop for feed and cooling closed loop for permeate) were connected to a membrane cell made of Plexiglas with an effective membrane surface area of 0.0032  $\text{m}^2$ . A new hydrophobic PTFE membrane, with a nominal pore size of 0.45  $\mu\text{m}$  (Table 1), was placed inside the module for each experiment. Permeate electrical conductivity and temperatures of the feed and permeate solutions were measured continuously during the experiments with variation ranges of [26–80 °C] and [18–23 °C], respectively. All experiments were conducted for 90 minutes in a counter current configuration to maintain the temperature gradient.

The permeate quality was tested by measuring the COD according to *Standard Methods for the Examination of Water and Wastewater* (Rodier 1984) and the electrical conductivity using an S823 pH/conductivity meter.

Table 1 | Membrane characteristics

Material	Nominal pore size ( $\mu\text{m}$ )	Thickness ( $\mu\text{m}$ )	Bubble point (bar)	LEP <sup>a</sup> (bar)	Contact angle <sup>a</sup> (°)	Support material
PTFE	0.45	160 ± 40	0.55–0.76	2.25	123.36	PP

Source: membrane data sheet.

<sup>a</sup>Measured parameters.

The permeate flux ( $J_p$ ) ( $L/m^2 h$ ) is by definition the flux of water vapor that crosses the hydrophobic membrane through its pores and it is proportional to the vapor pressure difference across the membrane following Darcy's law:

$$J_p = Bm (P_f - P_p) \quad (1)$$

where  $Bm$  is the membrane coefficient,  $P_f$  is the vapor pressure (bar) at the feed side water/vapor interface and  $P_p$  is the vapor pressure at the permeate side water/vapor interface. It is also worth noting that the water vapor is an exponential function of the operating temperature according to the Antoine equation:

$$P^0 = \exp\left(23.238 - \frac{3.841}{T - 45}\right) \quad (2)$$

where  $P^0$  is the pure water vapor (Pa) and  $T$  is the water temperature (K).

The volumetric permeate flux ( $L/m^2 h$ ) is expressed as follows:

$$J_p = \frac{\Delta V}{S \Delta t} \quad (3)$$

where  $\Delta V$  is the permeate volume (L),  $S$  is the effective membrane surface ( $m^2$ ) and  $\Delta t$  is the DCMD operating time (h).

### Membrane characteristics

Flat sheet PTFE hydrophobic membranes were purchased from Membrane Solutions to conduct the DCMD experiments. Table 1 gives the details of the membrane characteristics as provided by the manufacturer (material, nominal pore size, thickness and bubble point) and two measured hydrophobicity parameters (liquid entry pressure (LEP) and contact angle).

### Liquid entry pressure

The LEP was measured using 0.5% NaCl solution to which we applied a gradually increasing pressure perpendicularly to the active membrane surface with a fixed pressure slope ( $dp/dt = 0.01 \text{ bar/s}$ ). As soon as the liquid solution broke through the membrane, the corresponding pressure (LEP) was recorded (Table 1). The membrane LEP value shows a good membrane resistance against water breakthrough across the membrane pores (membrane wetting).

### Contact angle

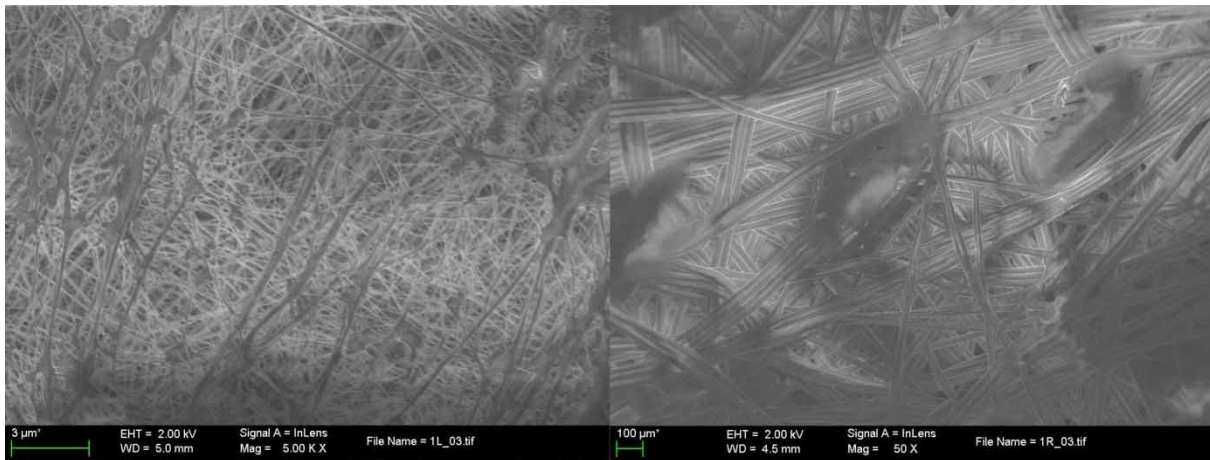
The contact angle characterization (CA) of the membrane used was achieved via the automated method of DataPhysics Contact Angle System OCA and the DataPhysics SCA20 software. The contact angle measurements were performed by the automatic generation of water drops using a pure water dispenser ( $5 \mu\text{m}$ ) on new membrane samples at  $22^\circ\text{C}$  (Table 1). The measured contact angle ( $>90^\circ$ ) shows that the membrane has a good hydrophobic nature that forms a physical barrier to the feed water stream. According to the literature, the membrane used has a good contact angle value in comparison to other synthesized and modified hydrophobic membranes in which the value varies between  $112.7^\circ$  and  $154.2^\circ$  (Prince *et al.* 2012, 2014b; Efome *et al.* 2016). Moreover, according to the aforementioned references, this parameter could contribute to the improvement of DCMD performance.

### Scanning electron microscopy

Scanning electron microscopy (SEM), coupled with energy dispersion spectrometry (EDS) was used for membrane characterization. Membrane samples were observed on both sides (PTFE active surface and PP non-woven support surface) with a Zeiss Merlin FE-SEM. SEM images of the membrane active surface and the support layer are shown in Figure 1. The SEM pictures show the differences in structure between the PTFE membrane's active surface, which has a tight net-like appearance (nominal pore size =  $0.45 \mu\text{m}$ , magnitude =  $5.10^3$  times), and the PP support layer, which has a larger net structure with bigger spaces (magnitude = 50 times).

### Experimental design and statistical analysis

RSM is applied in this study to optimize the DCMD process for saline wastewater treatment through one of the most commonly used forms of RSM: central composite design (CCD). This method, which is suitable for a quadratic surface, is generally used to optimize the effective parameters of a process and to identify their existing interactions and their extent with a minimum number of experiments. CCD is a second-order design which is based on adding a number of ( $2k$ ) axial-points experiments and a number of center-points replications ( $n_0$ ) to a simple first order design ( $2^k$ ) where  $k$  is the number of factors. The added experiments help to get more information about the response



**Figure 1** | SEM images of the PTFE membrane active surface (left) and the PP support layer (right).

surface and the optimization of the process through a second-order model (Khuri & Mukhopadhyay 2010).

The CCD model is a mathematical polynomial function expressed as follows in the case of four independent variables:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_4 + b_{11}X_1^2 + b_{22}X_2^2 + b_{33}X_3^2 + b_{44}X_4^2 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{14}X_1X_4 + b_{23}X_2X_3 + b_{24}X_2X_4 + b_{34}X_3X_4 \quad (4)$$

In our study, with a factor number of  $k = 4$ , we carried a total number of 28 experiments that included  $2^4$  orthogonal design points, eight axial points ( $\alpha = \pm 1.682$ ) and four replications of the center-points (Table 3).

Analysis of variance (ANOVA) was applied to determine the statistical significance of the model and to study the quality of the model's fit. The statistical and graphical analysis tool used in this work was Minitab<sup>®</sup> 17 software by Minitab Inc.

In our study, the volumetric permeate flux  $J_p$  (L/m<sup>2</sup>·h) was considered to be the main response (Y) to be optimized by

CCD through the optimization of four different factors [ $X_1$ : temperature difference  $\Delta T$  (°C),  $X_2$ : feed velocity  $V_f$  (m/s),  $X_3$ : NaCl concentration [ $NaCl$ ] (g/L) and  $X_4$ : glucose concentration [ $Gluc$ ] (g/L)]. The first step in applying the CCD is to identify the factors and their different levels. Table 2 illustrates the four chosen independent factors in our study with their respective codes and actual values. The choice of the variation ranges of each parameter was made according to preliminary tests with respect to the volumetric permeate flux ( $J_p$ ), it being the main response monitored during this study.

## RESULTS AND DISCUSSION

### Permeate flux stability

The stability of the DCMD process during the operation time for synthetic saline wastewater treatment was checked through preliminary tests to evaluate the permeate flux evolution and the quality of the permeate obtained (Figure 2). The volumetric permeate flux maintained an almost

**Table 2** | CCD variables levels in their coded and actual values

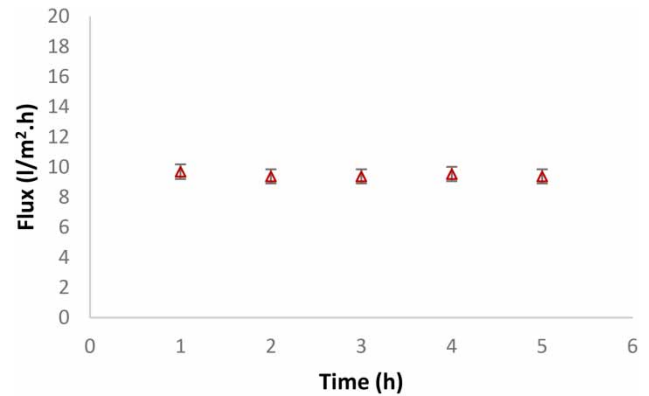
Factors	Symbols	Actual values of the coded levels				
		$-\alpha$	$-1$	$0$	$+1$	$+\alpha$
Temperature difference $\Delta T$ (°C)	$X_1$	4.8	15	30	45	55.2
Feed velocity $V_f$ (m/s)	$X_2$	0.027	0.039	0.057	0.075	0.086
Initial NaCl concentration [ $NaCl$ ] (g/L)	$X_3$	10.1	14.1	20	25.9	29.9
Initial glucose concentration [ $Gluc$ ] (g/L)	$X_4$	0.5	4.5	10.3	16.1	20

**Table 3** | CCD with predicted and experimental DCMD results

Experiment number	$X_1$	$X_2$	$X_3$	$X_4$	Predicted flux (L/m <sup>2</sup> ·h)	Experimental flux (L/m <sup>2</sup> ·h)
1	-1	-1	-1	-1	3.1	3.1 ± 0.4
2	1	-1	-1	-1	14.3	12.8 ± 0.3
3	-1	1	-1	-1	3.8	4.4 ± 0.7
4	1	1	-1	-1	17.6	17.5 ± 0.1
5	-1	-1	1	-1	4.0	2.8 ± 0.2
6	1	-1	1	-1	13.5	14.1 ± 0.4
7	-1	1	1	-1	4.4	4.1 ± 0.1
8	1	1	1	-1	16.5	15.9 ± 0.4
9	-1	-1	-1	1	4.2	5.6 ± 0.8
10	1	-1	-1	1	14.9	14.1 ± 0.5
11	-1	1	-1	1	10	6.6 ± 0.9
12	1	1	-1	1	23.6	27.5 ± 0.3
13	-1	-1	1	1	3.0	1.9 ± 0.2
14	1	-1	1	1	12.1	12.5 ± 0.3
15	-1	1	1	1	8.6	13.0 ± 0.5
16	1	1	1	1	20.3	19.1 ± 0.4
17	- $\alpha$	0	0	0	2.7	2.3 ± 0.2
18	$\alpha$	0	0	0	21.9	22.5 ± 0.2
19	0	- $\alpha$	0	0	6.8	8.1 ± 0.3
20	0	$\alpha$	0	0	14.2	13.1 ± 0.2
21	0	0	- $\alpha$	0	11.2	10.8 ± 0.3
22	0	0	$\alpha$	0	9.3	10.0 ± 0.2
23	0	0	0	- $\alpha$	9.0	10.5 ± 0.1
24	0	0	0	$\alpha$	13.1	11.9 ± 0.4
25	0	0	0	0	11.4	11.3 ± 0.1
26	0	0	0	0	11.4	11.4 ± 0.1
27	0	0	0	0	11.4	11.3 ± 0.1
28	0	0	0	0	11.4	11.3 ± 0.1

constant level, with an average value of 9.5 L/m<sup>2</sup>·h during 5 hours of treatment, under the following operating conditions:  $\Delta T = 30^\circ\text{C}$ ,  $V_f = 0.051\text{ m/s}$ ,  $[\text{NaCl}] = 10\text{ g/L}$  and  $[\text{Gluc}] = 5\text{ g/L}$ .

Concerning the permeate quality, the electrical conductivity reduction from the feed side to the permeate side was 99.9% and for COD the reduction was higher than 99.9%. These reduction rates confirm the ability of the DCMD process with the PTFE membrane (pore size = 0.45  $\mu\text{m}$ ) to treat and desalinate synthetic saline wastewater, with a stable performance over time (5 hours of treatment). Following this observation, an operation time of 1 hour was selected for all optimization experiments via RSM. Following each

**Figure 2** | Permeate flux evolution over the operation time.

experiment performed in this study, both COD and permeate conductivity were measured and the removal rates were all higher than 99.9%, with very low variations. Therefore, only the permeate flux was considered to be the main response in our work.

### Response surface methodology application in DCMD

The results of CCD experiments in terms of experimental and predicted permeate flux values are shown in Table 3. As can be seen in the table, the predicted response values are highly comparable to the experimental data, which indicates that the regression model developed provides a good fit to the experimental results.

The predicted flux was calculated using the following regression equation, which presents the empirical model written in terms of actual variables (Equation (5)). As illustrated in Table 3, the predicted permeate flux values have a large range of variation following changes in the parameters from 3 to 23 L/m<sup>2</sup>·h, which indicates the significant effect of the chosen parameters on the DCMD process.

$$\begin{aligned}
 J_p = & 11.38 + 5.706\Delta T + 2.21V_f - 0.549[\text{NaCl}] \\
 & + 1.209[\text{Gluc}] + 0.313\Delta T^2 - 0.321V_f^2 - 0.405[\text{NaCl}]^2 \\
 & - 0.127[\text{Gluc}]^2 + 0.646\Delta T V_f - 0.417\Delta T[\text{NaCl}] \\
 & - 0.098\Delta T[\text{Gluc}] - 0.074V_f[\text{NaCl}] + 1.292V_f[\text{Gluc}] \\
 & - 0.503[\text{NaCl}][\text{Gluc}]
 \end{aligned} \quad (5)$$

When studying the coefficients given in Equation (5), we can conclude that temperature difference ( $\Delta T$ ) has the most significant influence on mass transfer in DCMD followed by feed velocity, with coefficient values of  $b_1 = 5.706$  and



$b_2 = 2.21$ , respectively. Both variables did had a positive effect on permeate flux ( $J_p$ ) since increasing one or both of them did lead to an increase in the permeate flux. In addition,  $\Delta T^2$  also has a significant influence on permeate flux and indicates the non-linearity of  $J_p$  with respect to temperature  $T$ .

The significant effect of both temperature difference and feed flow rate, and therefore the feed velocity, have been shown to be some of the major parameters on which DCMD process performance depends. Following the temperature difference and feed flow rate variations, significant high or low permeate fluxes could be achieved and this could be attributed to the nature of the process driving force (Hou et al. 2010; He et al. 2011; Ashoor et al. 2016).

The main effects of the investigated operating conditions, which are summarized in Figure 3, confirm the previous results. According to Equation (2), the water vapor, and consequently the permeate flux, are an exponential function of the temperature, which can be seen in Figure 3. Moreover, Figure 3 shows that the temperature difference ( $\Delta T$ ) has the greatest effect on DCMD treatment of synthetic saline wastewater in comparison to the rest of the variables, followed by feed velocity ( $V_f$ ) as previously shown.

Meanwhile, the NaCl and glucose concentrations showed less influence on the permeate flux in the respective ranges of variation. The NaCl concentration had a slightly negative influence ( $b_3 = -0.549$ ) on the response value

( $J_p$ ). Regarding its influence on DCMD performance, the NaCl concentration increase slightly affected the water activity on the feed side and therefore the driving force, which led to a small decrease in the permeate flux (Martinez & Rodriguez-Marot 2007; Boubakri et al. 2014).

The glucose concentration showed a small influence on the response, with a coefficient value of  $b_4 = 1.209$ . In previous studies on DCMD using sucrose aqueous solutions, an increase in the feed concentration led to a small drop in the permeate flux that was linked to change in the feed water activity. However, this influence was less than obtained with saline feed solutions at the same concentration. It is important to note the close link between the feed viscosity and its water activity since when adding sucrose as a solute to the feed stream, viscosity increases, especially when the feed solution temperature is relatively low ( $\leq 35^\circ\text{C}$ ) and glucose concentrations are high (100–500 g/L) (Schofield et al. 1990; Izquierdo-Gil et al. 1999; Martinez & Rodriguez-Marot 2007). However, in our study the glucose concentration did not exceed 20 g/L as a maximum value, which is much lower than the concentrations used in the aforementioned investigations carried out on sucrose aqueous solutions, which started with 100 g/L as the lowest sucrose concentration. This difference in glucose concentration could be the reason behind the different observed effects on the permeate flux. According to our model, at high temperature differences the positive effect of glucose concentration decreases.

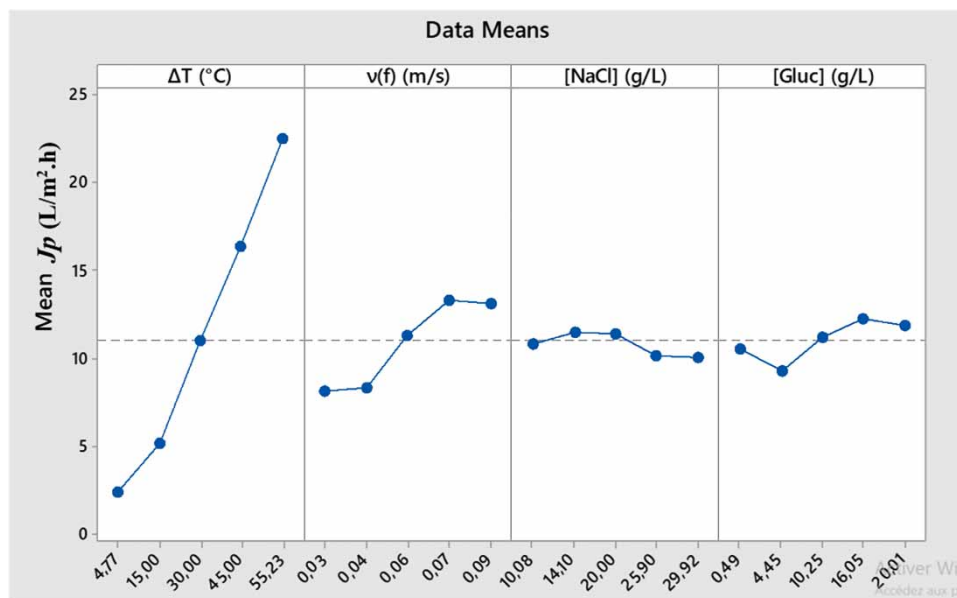


Figure 3 | Main effects plot for the permeate flux ( $J_p$ ).

## Analysis of variance

Variance analysis (ANOVA) was used to investigate the statistical significance of the regression model as well the significance of the effects of the various parameters and their interactions. Statistical significance is mainly reflected by both the  $p$ -value ( $p$ -value  $< 0.05$  for significant influence) and the  $R^2$  value, which indicates the proportion of variation in the regression model response; a value close to 1 for a model shows good effectiveness and predictive ability (Boubakri *et al.* 2014).

With respect to the different operating variables and their interactions, the ANOVA results showed that the temperature difference and the feed velocity had most significant positive influence on the DCMD response, with low probability values for both of them ( $p = 0.000$ ). Concerning the NaCl concentration, the variance analysis indicated that it had an insignificant effect on the permeate flux response ( $p = 0.119$ ). However, the glucose concentration had a relatively significant influence on the process response ( $p = 0.003$ ). According to the ANOVA results, the only significant variables interaction in this case were between the feed velocity and the glucose concentration ( $p = 0.005$ ). The increase in the feed flow rate, and consequently its velocity, decreased the temperature difference between the inlet and outlet of the DCMD module. This decrease led to higher  $\Delta T$  which affected the feed stream viscosity and consequently caused the increase in the permeate flux (Schofield *et al.* 1990; Gryta 2012).

In our study, the model had a high  $R^2$  level of 0.967, which indicated that the empirical model could explain more than 96.7% of the data deviation and therefore is statistically significant. This observation was strengthened by the obtained  $R^2_{\text{Adj}}$  value: 0.932 which meant that significant terms had been included in the empirical model (Khayet *et al.* 2007). In addition, the regression model had a very low  $p$ -value (0.000) which reflected its high significance and its ability to provide a good prediction of the response permeate flux ( $J_p$ ). All the ANOVA results confirmed the effectiveness and significance of the model.

By considering both the effectiveness of the model and its good fit to the experimental data by the comparison between the predicted and the experimental responses, we can conclude that its statistical validation is well established. The regression model could be applied to the description and optimization of DCMD permeate flux response through the optimization of the chosen variables ( $\Delta T$  ( $^{\circ}\text{C}$ ),  $V_f$  (m/s),  $[\text{NaCl}]$  (g/L) and  $[\text{Gluc}]$  (g/L)) in their aforementioned respective ranges of variation.

## Response surface and contour-line plots

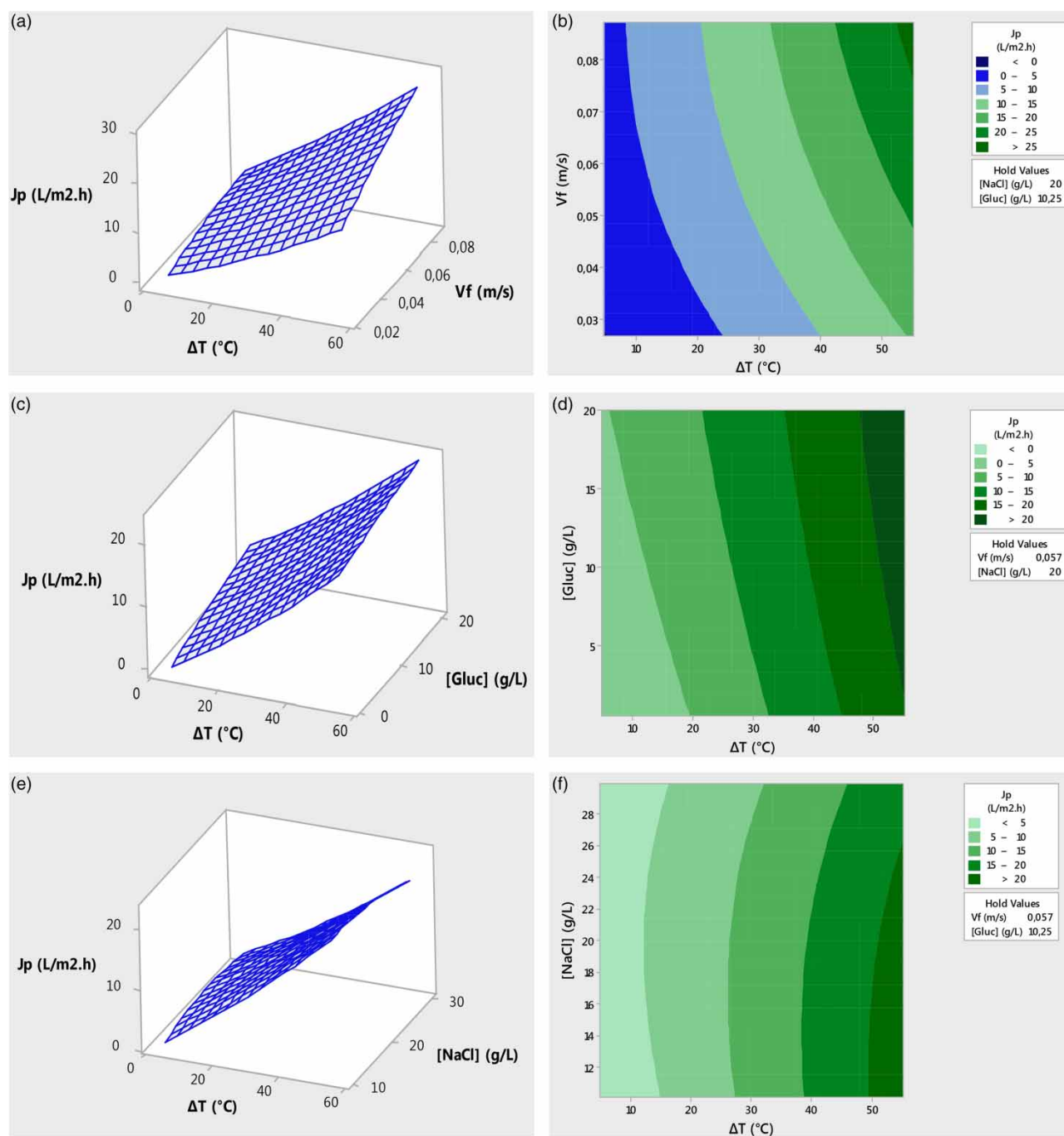
In order to study the interactions between the different variables, response surface and contour-line plots are presented in Figure 4. These helped to identify optimum levels for the variables, which helped to achieve the highest possible permeate flux value. For each plot, two of the four variables were kept constant and the regression model was used to calculate the permeate flux and follow its evolution as a function of the two other variables.

Figure 4(a) and 4(b) show the simultaneous effect of  $\Delta T$  and  $V_f$  on  $J_p$  at constant glucose and NaCl concentrations (maintained at their center points). As both variables had the most significant effects, with a more pronounced influence of  $\Delta T$  ( $b_1 = 5.706 > b_2 = 2.21$ ), high permeate fluxes ( $> 25 \text{ L/m}^2\cdot\text{h}$ ) were obtained at their highest levels ( $\Delta T \geq 50 \text{ }^{\circ}\text{C}$  and  $V_f \geq 0.081 \text{ m/s}$ ). At a low temperature difference the increase in the feed velocity did not significantly improve the permeate flux, which shows the importance of providing a higher driving force by increasing  $\Delta T$  (Nakoa *et al.* 2014). It is important to note that there was no significant interaction between the temperature difference and the feed velocity ( $p > 0.05$ ).

The effect of the temperature difference and the glucose concentration is shown in Figure 4(c) and 4(d) where the feed velocity and the NaCl concentration had constant values (24.46 L/h and 20 g/L, respectively). As expected, the temperature difference had the greater influence on the flux response, which reached the highest values ( $> 15 \text{ L/m}^2\cdot\text{h}$ ) at  $\Delta T > 40 \text{ }^{\circ}\text{C}$  for all glucose concentrations varying from 0 to 20 g/L. The pronounced effect of  $\Delta T$  in comparison to  $[\text{Gluc}]$  was confirmed by both their coefficients in the regression model ( $b_1 = 5.706 > b_4 = 1.209$ ) and their calculated  $p$ -values (0.000 and 0.003, respectively). In addition, the plotted curve presents a parallel aspect that could lead to the assumption that there was no significant interaction between the two variables.

At constant feed velocity (0.057 m/s) and glucose concentration (10.25 g/L), the permeate flux in Figure 4(e) and 4(f) increased significantly when increasing the temperature difference. A slight decrease could be identified when increasing the NaCl concentration. The observed results could be attributed to the induced reduction of the partial vapor pressure over the membrane surface as shown previously (Schofield *et al.* 1990; Kamrani *et al.* 2014). The plotted response surface suggested that the interaction between  $\Delta T$  and  $[\text{NaCl}]$  was negligible.

As illustrated in Figure 4(g) and 4(h), the plotted responses show the variation in  $J_p$  when varying both the



**Figure 4** | Response surface plots (a, c, e, g, i and k) and contour-line plots (b, d, f, h, g and l) of the predicted DCMD permeate flux ( $J_p$ ) as function of: temperature difference and feed velocity (a and b), temperature difference and glucose concentration (c and d), temperature difference and NaCl concentration (e and f), feed velocity and glucose concentration (g and h), NaCl concentration and glucose concentration (i and j) and feed velocity and NaCl concentration (k and l). (Continued.)

feed velocity and glucose concentration with constant  $\Delta T$  and [NaCl] at 30 °C and 20 g/L, respectively. At low feed velocities ( $V_f < 0.046$  m/s) the permeate flux did not exceed 10  $L/m^2 \cdot h$  for all glucose concentrations. A simultaneous increase of both variables led to a rise in  $J_p$  that could exceed 17.5  $L/m^2 \cdot h$ . This increase might have

been induced by the decrease of the temperature polarization effect caused by the increase in the heat transfer on the membrane surface that was led by the feed flow rate increase (Al-Asheha *et al.* 2006). The interaction surface plot between  $V_f$  and glucose concentration could indicate a significant interaction between these variables, which is



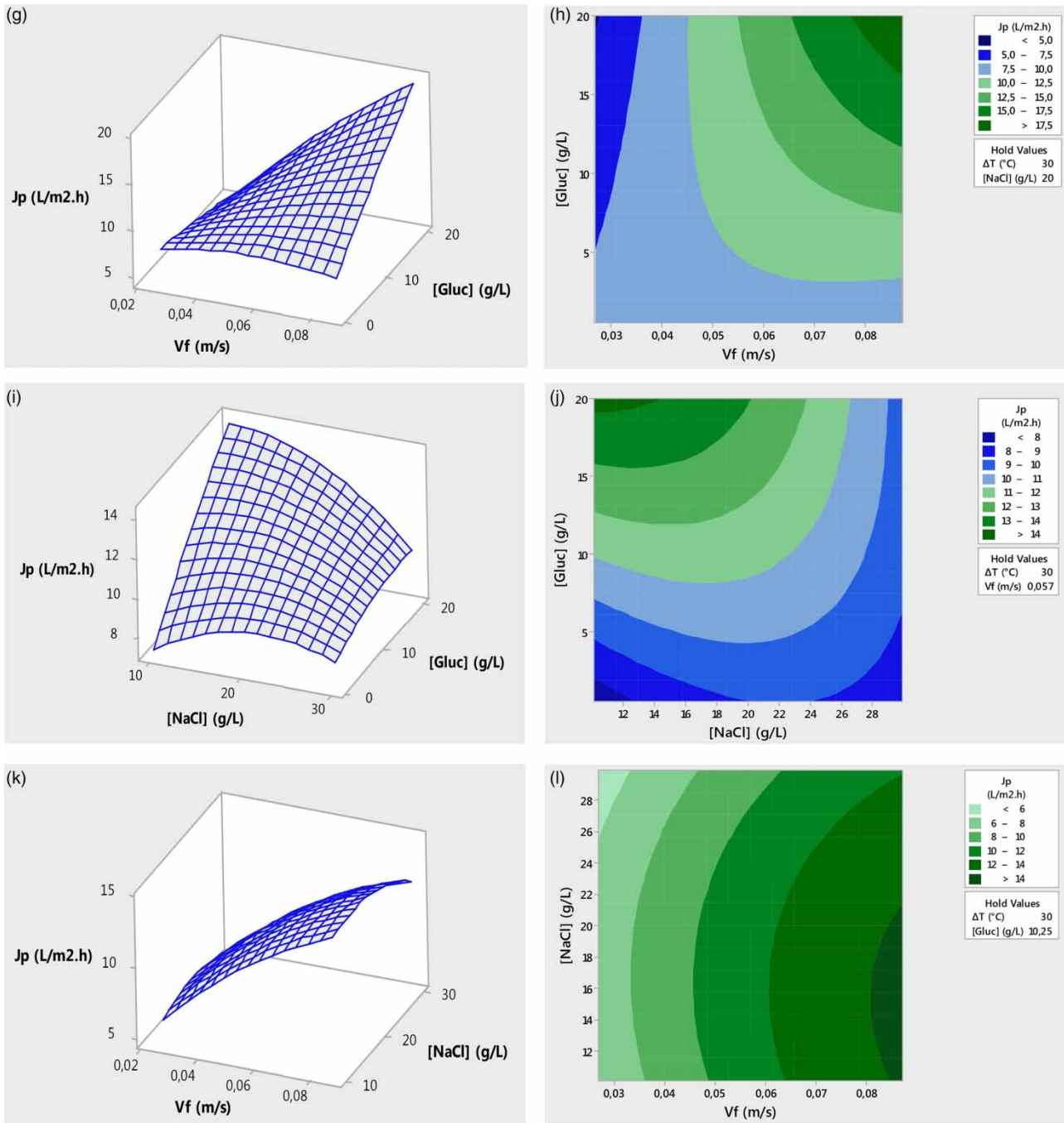


Figure 4 | Continued.

confirmed with the probability  $p$ -value obtained by ANOVA ( $< 0.05$ ).

The combined variation effect of  $[NaCl]$  and  $[Gluc]$  on the permeate flux at constant  $\Delta T$  (30 °C) and feed velocity (0.057 m/s) is shown in Figure 4(i) and 4(j). At low levels of both variables, the permeate flux had a minimum of 8 L/m<sup>2</sup>·h and it increased gradually with the glucose

concentration to reach 14 L/m<sup>2</sup>·h at low NaCl concentration, ranging between 10 L/m<sup>2</sup>·h and 11 L/m<sup>2</sup>·h for the highest  $[NaCl]$ . We could assume from the response plot that there was no significant interaction between the two variables.

Figure 4(k) and 4(l) illustrate the influence of feed velocity and salt concentration on the permeate flux variation when keeping the rest of the variables constant ( $\Delta T = 30$  °C and

**Table 4** | Optimum operating conditions and their corresponding predicted and experimental responses

Variable	$\Delta T$ (°C)	$V_f$ (m/s)	[NaCl] (g/L)	[Gluc] (g/L)	$J_p$ (L/m <sup>2</sup> ·h) predicted	$J_p$ (L/m <sup>2</sup> ·h) experimental	Composite desirability
Value	55.23	0.086	10.08	20.01	34.14	35.69	0.975

[Gluc] = 10.25 g/L). The plotted response reveals the effect of  $V_f$  increase which led to the rise in  $J_p$ , which exceeded 14 L/m<sup>2</sup>·h when  $V_f$  was around 0.081 m/s and [NaCl] < 21 g/L. At low feed velocities (<0.035 m/s) the calculated flux was less than 8 L/m<sup>2</sup>·h for all salt concentrations in the range investigated. As can be seen in the plotted responses, the interaction between  $V_f$  and [NaCl] was negligible.

### DCMD response optimization and model verification

As a result of the RSM application synthetic saline wastewater, a maximum response ( $J_p$ ) should be predicted. In order to identify the optimum operating conditions leading to the aforementioned response, all the effects and interactions were taken into consideration using Minitab software (Table 4). A maximum permeate flux of 34.14 L/m<sup>2</sup>·h was predicted at the following optimum operating conditions: temperature difference  $\Delta T = 55.23$  °C, feed velocity  $V_f = 0.086$  m/s, NaCl concentration [NaCl] = 10.08 g/L and glucose concentration [Gluc] = 20.01 g/L. The composite desirability was calculated to assess how well the combination of the defined optimum variables met the desired goal (maximizing the permeate flux  $J_p$ ). In this study we had a good desirability value of 0.975 (1 is the maximum).

In order to verify the optimization procedure and results, experiments were conducted at the optimum conditions obtained and the responses were compared to the predicted maximum response. Table 4 illustrates both the experimental permeate flux mean value after three repeated experiments at optimum operating conditions:  $35.69 \pm 0.6$  L/m<sup>2</sup>·h and the maximum permeate flux predicted by the regression model at the same conditions: 34.14 L/m<sup>2</sup>·h. The deviation of 4.3% indicates and confirms the validity of the optimization procedure and the DCMD optimization model in addition to high performance in terms of COD removal and conductivity rejection.

### CONCLUSIONS

RSM was applied for the optimization of synthetic saline wastewater DCMD treatment and desalination via the

optimization of the permeate flux ( $J_p$ ) with a remarkably high permeate quality in terms of electrical conductivity reduction (99.9%) and COD removal rate (>99.9%). In this study, four different variables were considered for creating the optimization model, namely: temperature difference  $\Delta T$ , feed velocity  $V_f$ , NaCl concentration [NaCl] and glucose concentration [Gluc]. The regression model we developed demonstrated a good level of effectiveness and ability to predict the process response with  $R^2 = 0.967$  and  $R_{Adj}^2 = 0.932$ . The statistical evaluation and analysis of the model showed that it had a high significance and that the most significant variable in the ranges investigated were the temperature difference and the feed flow rate followed by the glucose concentration, which had a less significant effect. The optimization procedure led to the prediction of the maximum permeate flux (34.14 L/m<sup>2</sup>·h) that could be achieved under the following optimum experimental conditions:  $\Delta T = 55.23$  °C,  $V_f = 0.086$  m/s, [NaCl] = 10.08 g/L and [Gluc] = 20.01 g/L. As a final step, experimental verification of the regression model showed its validity and suitability within the range of variables studied.

### ACKNOWLEDGEMENTS

This work was supported by the International Foundation for Science (IFS), Stockholm, Sweden, through a grant to Ms Sana Abdelkader (grant number: W/5585-1) and the Contract Program funded by the Tunisian Ministry in charge of research.

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First received 20 February 2018; accepted in revised form 7 June 2018. Available online 18 June 2018