Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2016.1141323

57 (2016) 24222–24230
November

Taylor & Francis
Taylor & Francis Group

Optimization of reverse osmosis treatment process to reuse the distillery wastewater using Taguchi design

K. Thirugnanasambandham[a], V. Sivakumar[b,*], K. Shine[c]

[a]*Department of Chemistry, Excel College of Engineering and Technology, Komarapalayam, Namakkal 637 303, TN, India*
[b]*Department of Chemical Engineering, AC Tech Campus, Anna University, Chennai 60005, TN, India, Tel. +91 4294 226606; Fax: +91 4294 220087; email: drvsivakumar@yahoo.com*
[c]*Department of Botany and Microbiology, College of Science, King Saud University, Post box 2455, Riyadh 11451, Saudi Arabia*

Received 18 March 2015; Accepted 1 January 2016

ABSTRACT

In this study, the reverse osmosis (RO) treatment of distillery wastewater was studied using various operating conditions including pH, temperature, transmembrane pressure, and feed flow rate. Taguchi method was used to design the experiments and optimize the RO treatment process for the maximum permeate flux and removal of salinity and COD. L_9 (3^4) orthogonal array for experimental design and the larger-the-better response category was applied to find optimum parameters. The optimum conditions were found to be as pH (6), temperature (45˚C), transmembrane pressure (10 bar), and flow rate (300 l/h). Under these conditions, 80 l/hm^2 permeate flux was achieved with 91% of salinity removal and 85% of COD removal. The obtained results revealed that RO treatment technique was an effective treatment technology for the reuse of distilleries wastewater.

Keywords: Distillery wastewater; Reverse osmosis; Optimization; Taguchi design; Validation

1. Introduction

The large quantity of aqueous waste discharged by industries has become a major environmental issue in all over the world because of its harmful nature [1]. Especially, wastewater discharged by distillery industry caused negative impact on environment when it is discharge into environment without a pretreatment. Distillery industries are one of the top users of water, with consumption ranging from 25 to 175 L/L production of alcohol [2]. Distillery industries required raw water for both process and non-process application. Water requirement in process applications include molasses preparation, steam generation, yeast propagation, etc., ranging from 14.5 to 21.4 L/L production of alcohol [3]. Then again, water usage in non-process applications include cooling water, steam generation, making potable liquor etc., is ranging from 102.65 to 240 L/L production of alcohol. Large quantities of high-toxic effluent have been discharged by the most distillery industries in India [4]. Hence, the Central Pollution Control Board (CPCB) in India listed distillery industries as one of the 17 top polluting industries. For each liter of alcohol production, the molasses-based distillery industries would generally discharge around 8–15 L of wastewater with bulk of organic matters and dark brownish color. Treatment scheme of distillery wastewater is shown in Fig. 1 [5]. But this method shows the ineffective treatment of distillery industry wastewater.

---

*Corresponding author.

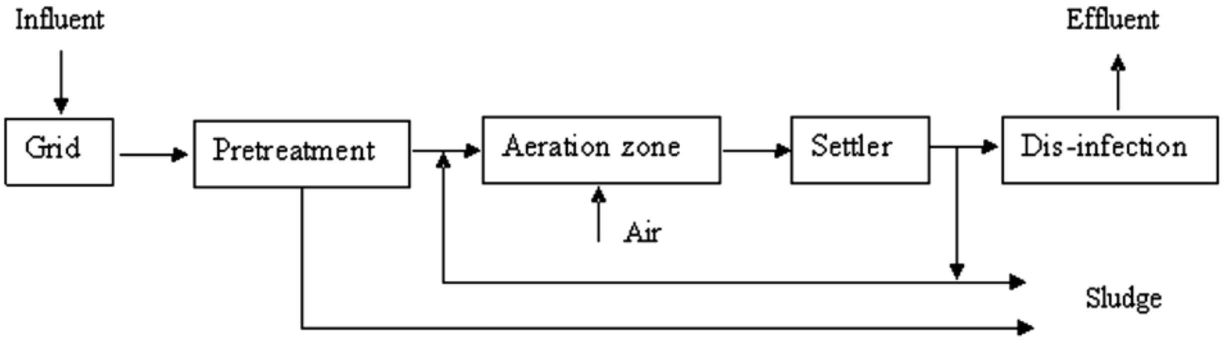

Fig. 1. Treatment scheme of distillery wastewater.

Extensive study reports are available on the treatment of distilleries wastewater such as chemical coagulation, electrocoagulation, aerobic, anaerobic, oxidation, adsorption, and UASB reactor treatment. But these treatment methods show the lower removal efficiency (RE) of salinity and COD, generate secondary pollutants that is solid sludge which needs a further treatment for safe removal [6]. So developing an effective distilleries wastewater treatment method for reuse of wastewater is a critical need due to increase in water scarcity.

Last few decades, membrane separation using reverse osmosis (RO) as one of the most promising technologies for treatment of wastewater because of its great efficiency, ease and low-cost of operation. RO treatment process enable to discharge safe water and reuse the industrial wastewaters by giving treated water with good quality [7]. Several literature review shows that various industrial wastewater are treated using RO. Besides, the optimization of parameters for RO treatment process such as temperature, pH, transmembrane pressure, and flow rate may increase the water treatment efficiency and in addition it also creates the chance to know the detailed mechanism behind in it [8]. The conventional optimization method of varying one factor at a time to study the effects of variables on the response is extremely time consuming and costly for large number of variables [9]. Statistical design of experiments is a useful method for studying the RO treatment process by conducting least number of trials. Response surface methodology (RSM) coupled with Taguchi design (TD) is a powerful statistical approach used for the analysis of multivariable systems.

An extensive literature search reveals that there is no report of previous studies on the treatment of distilleries wastewater using RO via RSM coupled with TD, which may give the crucial solution to environment-related problems [10]. Hence the present research has been undertaken to find out and optimize the process variables such as pH, temperature, transmembrane pressure, and flow rate for the maximum permeate flux, salinity removal, and COD removal from distillery wastewater using RO process via L_9 (3^4) orthogonal array for experimental design.

2. Materials and methods

2.1. Raw wastewater and chemicals

The distilleries wastewater were collected from distillery industry located in Erode, Tamil Nadu, India and were stored at 4°C. Chemicals (NaOH and HCl) used in this experiment were of analytical grade and bought from local dealers from Erode, Tamil Nadu.

2.2. Experimental method

Image of RO experimental setup was shown in Fig. 2. RO experiments were carried out in a lab-scale RO plant in cross-flow operation, which was bought from New venus, Chennai, India. The filtration module made up of polyamide having effective area of 50 cm^2 was made up of stainless steel. High-pressure pump was used in the experiment setup. The characteristics of membrane are summarized in Table 1. To attain stable membrane structure, each membrane was first pre-compacted with pure water for 5 h at the transmembrane pressure of 15 bar. 1 M HCl and 1 M NaOH were used to adjust the pH, and thermoheater was used to maintain the temperature of the feed wastewater. Experimental runs were performed with a feed volume of 5 L at the beginning of each run. All the experiments were performed in concentration mode of filtration (CMF). Permeate was collected in a beaker, concentrate was circulated back to the feed vessel. Permeate water was used to determine the salinity and COD reduction.

Fig. 2. Reverse osmosis experimental setup.

Table 1
Membrane specifications

Membrane specifications	
Material	Polyamide
Surface area	2 m^2
Structure	Asymmetric
Surface property	Hydrophilic
Pore size (nm)	<0.1
Max. P (bar)	15
Max. T (°C)	50

2.3. Analytical method

COD measurement of the wastewater was carried out according to the APHA Standard methods. The pH of the samples was monitored by a Li-113 model pH meter. Salinity of the wastewater was determined using digital TDS meter (LKU-5i5). The following equation were used to calculate RE of salinity and COD [11]:

$$\mathrm{RE} = \left(\frac{c_0 - c_e}{c_0}\right) \times 100 \tag{1}$$

where c_0 and c_e is the initial and final concentrations of salinity and COD respectively.

2.4. Taguchi design

In this present study, RSM coupled with Taguchi method was applied to design the experiments. The TD utilizes fractional factorial design called orthogonal arrays to minimize the experimental number as well as operating cost and raw material [12]. The selection of an appropriate orthogonal array which will depend on the number of variables and their levels. The parameters and their levels are presented in Table 2. Design-Expert 8.0.7.1 (State-Ease Inc., USA) statistical analysis software was used to analyze the experimental data. L_9 (3^4) orthogonal array was used for experimental design and pH (A), temperature (B), pressure (C), and flow rate (D) were selected as independent variables; whereas permeate flux (Y_1), salinity removal (Y_2), and COD removal (Y_3) were considered as responses. Then, the experimental data was fitted to the additive model in order to describe the RO process. The balanced characteristic of orthogonal array can be used to predict the performance value corresponding to optimum parameters using the following equation [13]:

$$Y_i = \mu + X_i + e_i \tag{2}$$

where μ is the overall mean of the performance value, X_i is the fixed effect of the quantity level arrangement in ith experiment, and e_i is the random error in the ith experiment. Analysis of variance (ANOVA) was done to find out the influence and relative importance of variables. Later, developed mathematical models were used to visualize the three-dimensional (3D) contour graphs to study the effect of independent factors on the responses. Lastly, numerical optimization was used to optimize the process parameters for maximum permeate flux and higher removal efficiencies of salinity and COD [14].

Table 2
Process variables and their ranges

Variable (unit)	Factors	Level		
	X	–1	0	1
pH	A	3	5	7
Temperature	B	25	35	45
Transmembrane pressure	C	8	10	12
Flow rate	D	300	450	600

3. Results and discussions

3.1. Predicted model and statistical analysis

TD and the corresponding results to find out the effects of the four independent factors such as pH, temperature, transmembrane pressure, and flow rate are presented in Table 3. The multiple regression analysis (Table 4) on the TD experimental values shows that linear model can explain the present treatment process robustly due to high *F*-value and lower *p*-value (<0.05). Mathematical models for the predicted responses such as permeate flux (Y_1), salinity removal (Y_2), and COD removal (Y_3) could be interpreted using the following equation (in the form of actual factors):

$$Y_1\,(\mathrm{l/hm^2}) = 37.4166 + 1.50000 \times \mathrm{pH} + 0.68333 \times \mathrm{Temperature} + 0.66667 \times \mathrm{Transmembrane\ pressure} - 0.014444 \times \mathrm{Flow\ rate} \tag{3}$$

$$Y_2\,(\%) = 48.18250 + 1.38167 \times \mathrm{pH} + 0.73350 \times \mathrm{Temperature} + 0.53417 \times \mathrm{Transmembrane\ pressure} - 0.015656 \times \mathrm{Flow\ rate} \tag{4}$$

$$Y_3\,(\%) = 43.32917 + 1.12750 \times \mathrm{pH} + 0.72867 \times \mathrm{Temperature} + 0.59083 \times \mathrm{Transmembrane\ pressure} - 0.016056 \times \mathrm{Flow\ rate} \tag{5}$$

ANOVA was performed for the statistical validation of the developed mathematical model. The Table 5 presented the ANOVA result for the fitted linear models of RO process. The models show the value of the determination coefficient (R^2) was greater than 0.9500 for responses, which implied that 95% of the variations could be explained by the fitted mathematical models [15]. For a suitable statistical model, adj-R^2 should be close to R^2. As presented in Table 5, adj-R^2 was greater than 0.90 for the responses indicates that a high degree of correlation between the predicted and experimental values. A lower value of coefficient of variation (CV) and adequate precision (AP) revealed a reliability of the experimental data [16]. The corresponding variables would be of higher significance if the *F*-value turns higher and the *p*-value turns smaller. Analysis of results showed that the developed models have high *F*-values and low *p*-values which show the significance of developed model. The suitability of developed models to represent the RO process technique was explained by creating diagnostic plots such as experimental vs. predicted plot (Fig. 3). The data points align very close to the diagonal line and it clearly illustrates a good relationship between actual and predicted value. From the results, it was validated that the developed mathematical models have the power to explain the RO process significantly [17].

3.2. Effects of process conditions on RO efficiency

The correlation between dependent and independent factors are depicted by the two-dimensional response surface contours plots created by the developed mathematical models and they are presented in Fig. 4. In the present study, the model has more than two variables. So the response surface contour plots are drawn by keeping one variable at a constant level, while the other two variables were assorted in their level.

3.2.1. Effect of pH

pH is one of the crucial parameter in RO process, which significantly influences the treatment process.

Table 3
TD with experimental designs

Run	*A*	*B*	*C*	*D*	Permeate flux ($\mathrm{l/m^2\,h}$)	Salinity removal (%)	COD removal (%)
1	7	45	10	300	80	89.74	84.03
2	5	25	10	600	60	67.86	63.56
3	5	35	12	300	75	84.56	78.56
4	3	25	8	300	60	70.54	65.42
5	3	35	10	450	65	75.45	68.56
6	3	45	12	600	72	82.65	77.54
7	7	25	12	450	65	75.24	68.56
8	5	45	8	450	74	85.26	79.69
9	7	35	8	600	70	80.24	72.46

Table 4
Sequential model sum of squares for response

Source	Sum of squares	DF	Mean Square	*F*-value	Prob. > *F*	Remarks
Sequential model sum of squares for permeate flux						
Mean	42,849.00	1	42,849.00			
Linear	373.00	4	93.25	28.69	0.0033	Suggested
2FI	10.43	3	3.48	1.35	0.5470	Aliased
Quadratic	2.57	1	2.57			Aliased
Cubic	0.00	0				Aliased
Residual	0.00	0				
Total	43235.00	9	4,803.89			
Sequential model sum of squares for salinity removal						
Mean	56,254.35	1	56,254.35			
Linear	408.57	4	102.14	28.93	0.0033	Suggested
2FI	13.35	3	4.45	5.76	0.2952	Aliased
Quadratic	0.77	1	0.77			Aliased
Cubic	0.00	0				Aliased
Residual	0.00	0				
Total	56677.04	9	6,297.45			
Sequential model sum of squares for COD removal						
Mean	48,162.69	1	48,162.69			
Linear	392.26	4	98.07	33.36	0.0025	
2FI	3.82	3	1.27	0.16	0.9119	Suggested
Quadratic	7.94	1	7.94			Aliased
Cubic	0.00	0				Aliased
Residual	0.00	0				Aliased
Total	48,566.71	9	5,396.30			

Table 5
ANOVA results for response

	Permeate flux (l/m^2 h)		Salinity removal (%)		COD removal (%)	
Source	F-value	*p*-value	*F*-value	*p*-value	*F*-value	*p*-value
Model	28.69	0.0033	28.93	0.0033	33.36	0.0025
A	16.62	0.0151	12.98	0.0227	10.38	0.0322
B	86.21	0.0007	91.43	0.0007	108.38	0.0005
C	3.28	0.1443	1.94	0.2361	2.85	0.1666
D	8.67	0.0422	9.37	0.0376	11.84	0.0263
C.V. %	2.61		2.38		1.95	
PRESS	59.34		70.24		68.35	
AP	15.62		15.80		22.03	
R^2	0.9792		0.9806		0.9965	
Adj-R^2	0.9585		0.9613		0.9752	

The pH value of wastewater can affect the membrane surface charge because of the disassociation of membrane's functional groups. To find out its effect in RO treatment process, the experiments were performed at different pH range (3–5–7) and the results were presented in Fig. 4(a)–(c). The result showed that permeate flux and removal efficacies of salinity and COD were improved with increasing pH throughout the experiment. This can be described by the fact that electrostatic interaction between the solutes and membrane in the wastewater occurred throughout the pH range studied [18].

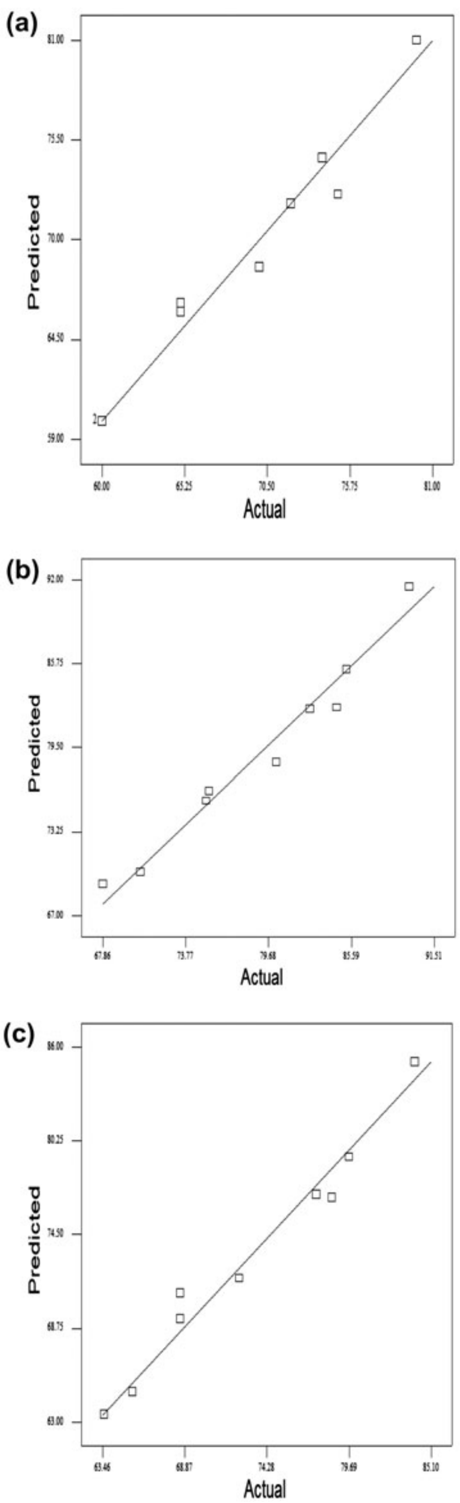

Fig. 3. Actual vs. predicted plot: (a) Permeate flux, (b) Salinity removal, and (C) COD.

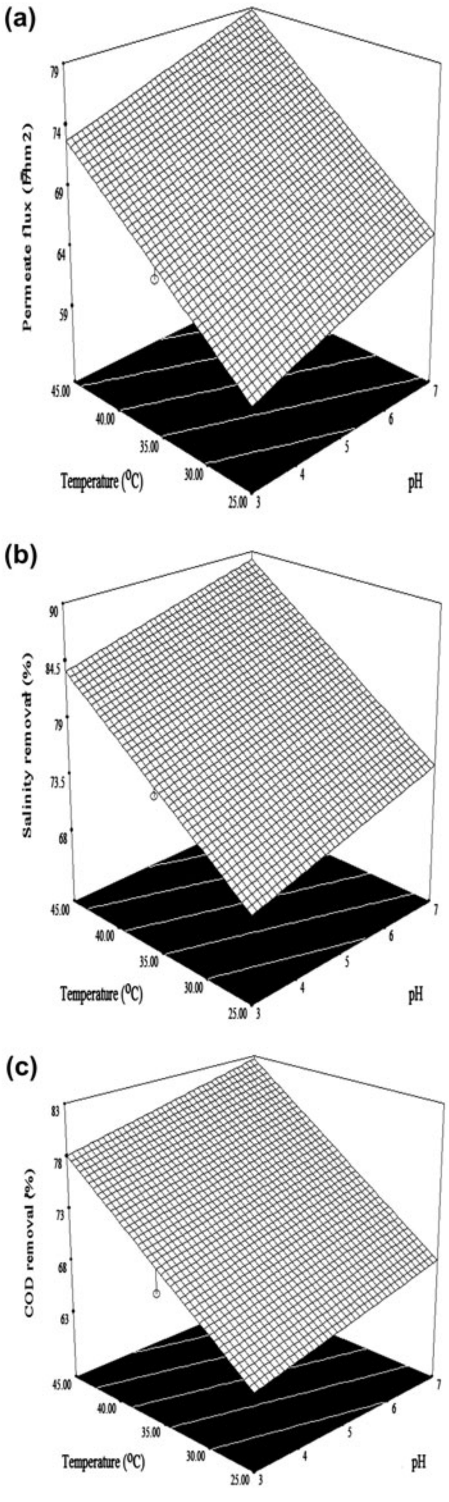

Fig. 4. Effect of process variables (A and B) RO treatment process.

3.2.2. *Effect of temperature*

RO treatment process is highly influenced by temperature because it plays a major role in removal of salinity and COD from distillery wastewater. So as to investigate its effect on RO treatment process, experiments were carried out at various temperature (25–35–45˚C) and the observations are presented in Fig. 4. The result showed that the permeate flux and percentage of salinity and COD removal efficiencies were improved with increasing temperature. The effects of permeate flux with temperature is mainly explained by solvent diffusion coefficient in the membranes, solvent viscosity, and thermal expansion of the membrane material [19]. The reduction in solvent viscosity results increases in solvent diffusion coefficient and higher temperature of experiment may expand the membrane structure and solutes can easily pass through the membrane, thus treatment efficiency is enhanced.

3.2.3. *Effect of transmembrane pressure*

The transmembrane pressure is a major parameter to determine the efficiency of RO treatment process to distillery wastewater. To find out its effects on RO treatment process, experiments were done at various transmembrane pressure (8–10–12 bar) and the results are illustrated in Fig. 4(a) and (b). The result showed that the treatment efficiency of RO process increases with increasing transmembrane pressure. This can explained by the fact that, by increasing the transmembrane pressure, a phenomenon known as concentration polarization on the membrane surface happens, which makes the concentration change between the both sides of the membrane rises [20]. So the diffusion-driven force rises, more particles cross the membrane module, thus permeate flux, salinity and COD removal efficiencies are increased.

3.2.4. *Effect of flow rate*

In the RO treatment process, the flow rate is a major factor which significantly affects the treatment efficiency. To find out the effect of flow rate on the RO treatment process, experiments were carried out at flow rate (300–450–600 l/h) and results are presented in graphs Fig. 5(a), (b) and (c). The result showed that the permeate flux, percentage removal of salinity, and COD were decreased with higher flow rate. This can explained by the fact that at higher flow rate there is

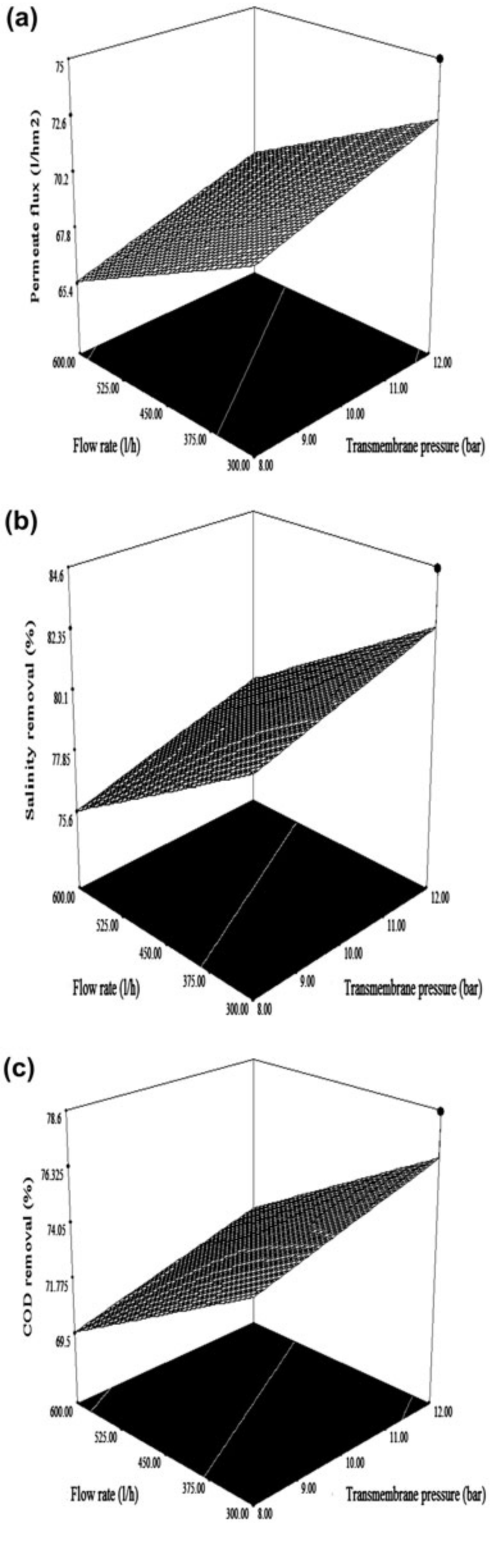

Fig. 5. Effect of process variables (C and D) RO treatment process.

Table 6
Characteristics of wastewater after and before treatment

Parameters	Raw wastewater	After RO treatment
pH	5.68	6.84
Salinity (mg/L)	4,586	154
COD (mg/L)	6,524	78

an ineffective interaction between membrane modules and feed wastewater, thus permeate flux, removal efficiencies of salinity, and COD are decreased [21].

3.3. Optimization of RO treatment process

Numerical optimization technique is used to predict the optimum response values and it was found to be pH (6), temperature (45˚C), transmembrane pressure (10 bar), and flow rate (300 l/h). Under these conditions, 80 l/hm^2 permeate flux are achieved with 91% of salinity removal and 85% of COD removal. Finally, confirmation testing is used to verify the optimum condition. The final experiment was carried out using the optimum parameters to correlate the results with the predicted values [22–24]. The obtained results are within a ±5% error range. Also the results showed a good correlation between the experimental and predicted values. Then, the quality of the composite permeate attained from confirmation test was compared with raw wastewater in Table 6. This results indicated that RO-treated distillery wastewater may be reused in inplant application and land irrigation.

4. Conclusions

TD coupled with RSM is used to optimize the parameter for RO treatment process to treat distilleries wastewater. Linear mathematical models were developed with good coefficient of determination values ($R^2 > 0.95$) for permeate flux, COD removals, and salinity. ANOVA revealed the significant effect of each factors and the optimum parameters for RO treatment method were found to be pH (6), temperature (45˚C), transmembrane pressure (10 bar), and flow rate (300 l/h). Under these conditions, 80 l/hm^2 permeate flux was achieved with 91% of salinity removal and 85% of COD removal.

Acknowledgements

This Project was supported by King Saud University, Deanship of Scientific Research, College of Sciences Research Center.

References

[1] J.D. Jacobson, M.D. Kennedy, G. Amy, J.C. Schippers, Phosphate limitation in reverse osmosis: An option to control biofouling? Desalin. Water Treat. 5 (2009) 198–206.
[2] K. Thirugnanasambandham, V. Sivakumar, J. Prakash Maran, Treatment of egg processing industry effluent using chitosan as an adsorbent, J. Serb. Chem. Soc. 79 (2014) 743–757.
[3] K. Thirugnanasambandham, V. Sivakumar, J. Prakash Maran, Analysis of efficiency of bacillus subtilis to treat bagasse based paperand pulp industry wastewater—A novel approach, J. Korean Chem. Soc. 58 (2014) 198–204.
[4] K. Thirugnanasambandham, V. Sivakumar, J. Prakash Maran, Optimization of electrocoagulation process to treat biologically pretreated bagasse effluent, J. Serb. Chem. Soc. 79 (2014) 613–626.
[5] L.A. Ioannou, D. Fatta-Kassinos, Solar photo-Fenton oxidation against the bioresistant fractions of winery wastewater, J. Environ. Chem. Eng. 1 (2013) 703–712.
[6] S.G. Yiantsios, D. Sioutopoulos, A.J. Karabelas, Colloidal fouling of RO membranes: an overview of key issues and efforts to develop improved prediction techniques, Desalination 183 (2005) 257–272.
[7] L.D. Nghiem, A.I. Schäfer, Fouling autopsy of hollow-fibre MF membranes in wastewater reclamation, Desalination 188 (2006) 113–121.
[8] W.S. Ang, S. Lee, M. Elimelech, Chemical and physical aspects of cleaning of organic-fouled reverse osmosis membranes, J. Membr. Sci. 272 (2006) 198–210.
[9] K. Thirugnanasambandham, V. Sivakumar, J. Prakash Maran, Chitosan based grey wastewater treatment—A statistical design approach, Carbohydr. Polym. 99 (2014) 593–600.
[10] J.S. Vrouwenvelder, J.W.N.M. Kappelhof, S.G.J. Heijrnan, J.C. Schippers, D. van der Kooija, Tools for fouling diagnosis of NF and RO membranes and assessment of the fouling potential of feed water, Desalination 157 (2003) 361–365.
[11] A. Ritter, R. Muñoz-Carpena, Performance evaluation of hydrological models: Statistical significance for reducing subjectivity in goodness-of-fit assessments, J. Hydrol. 480 (2013) 33–45.
[12] L. Wu, R. Munoz-Carpena, B. Gao, W. Yang, Y. Pachepsky, Colloid filtration in surface dense vegetation: Experimental results and theoretical predictions, Environ. Sci. Technol. 48 (2014) 3883–3890.
[13] M.H. Li, Reducing specific energy consumption in Reverse Osmosis (RO) water desalination: An analysis from first principles, Desalination 276 (2011) 128–135.
[14] S. Coker, P. Sehn, Four years field experience with fouling resistant reverse osmosis membranes, Desalination 132 (2000) 211–215.
[15] M.A. Kuehne, R.Q. Song, N.N. Li, R.J. Petersen, Flux enhancement in TFC RO membranes, Environ. Prog. 20 (2001) 23–26.
[16] K. Thirugnanasambandham, V. Sivakumar, J. Prakash Maran, Efficiency of electrocoagulation method to treat chicken processing industry wastewater—Modeling and optimization, J. Taiwan Inst. Chem. Eng. doi: 10.1016/j.jtice.2014.04.011.

[17] K. Thirugnanasambandham, V. Sivakumar, J. Prakash Maran, Modeling and optimization of biogas production from rice mill effluent by anaerobic digestion process using up flow anaerobic sludge blanket reactor, J. Renewable Sustainable Energy 6 023129 (2014), doi: 10.1063/1.4873400

[18] M.A. Barakat, E. Schmidt, Polymer-Enhanced Ultrafiltration Process for Heavy Metals Removal from Industrial Wastewater, The 3rd International Conference on Water Resources and Arid Environments (2008).

[19] Z. Beril Gönder, S. Arayici, H. Barlas, Advanced treatment of pulp and paper mill wastewater by nanofiltration process: Effects of operating conditions on membrane fouling, Sep. Purif. Technol. 76 (2011) 292–302.

[20] C. Charcosset, A review of membrane processes and renewable energies for desalination, Desalination 245 (2009) 214–231.

[21] P. Navarro, J. Sarasa, D. Sierra, S. Esteban, J.L. Ovelleiro, Degradation of wine industry wastewaters by photocatalytic advanced oxidation, Water Sci. Technol. 51 (2005) 113–120.

[22] J. Prakash, V. Maran, K. Sivakumar, Thirugnanasambandham, Artificial neural network and response surface methodology modeling in mass transfer parameters predictions during osmotic dehydration of *Carcia papaya* L., Alexandria Eng. J. 52 (2013) 507–516.

[23] P.L. Ritger, N.A. Peppas, A simple equation for description of solute release II. Fickian and anomalous release from swellable devices, J. Controlled Release 5 (1987) 37–42.

[24] N.L. Yong, L. Mohammad, A. Wahab, C. Yin, R. Rosiah, Optimization of polymeric membrane characteristics through thermal treatment and deposition of polyelectrolyte layers using response surface modeling, Adv. Polym. Technol. 8 (2014) 42–55.