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Investigation of Applying NF Membrane for Dye Removal Using Multivariate Regression Model

Ghada A. Al Bazedi*, Marwa M. Elsayed , Mona A. Abdel-Fatah

Chemical Engineering and Pilot Plant Department, National Research Center,

El-Bohouth Street, Dokki, Egypt

Abstract: Recently, Nanofiltration plays an important role in dye wastewater treatment. The efficiency of membrane separation system for wastewater treatment plant is complicated due to different operating conditions involved in the separation process. Estimation of the Nanofiltration membrane performance is essential to evaluate the final water quality for further reuse. This work focus on the development of a simplified predictive model for membrane performance estimation under a set of parameters. A linear multiple regression model with coefficient of determination (R^2) of 1 has been developed for COD values in the permeate stream, while (R^2) of 0.85 for membrane rejection estimation. Seven investigated parameters have been related to formulate the model. The model has been applied for dye wastewater scheme comprising Nanofiltration with proper pretreatment. The model has been validated with an average deviation of 15% for membrane rejection and almost no deviation for COD level estimation in the permeate stream.
Keywords: Nanofiltration, Membrane separation, Wastewater Treatment, predictive model, Reactive dye.

1. Introduction

Textile dyeing industry is one of the largest water consuming industries. The effluent wastewater produced from the dyeing industries contains different chemicals and colouring agents and hence this effluent requires proper treatment before its discharge (Do & Chem[1]). The dye house effluents are difficult to be treated because of the variable composition in the waste stream (Najmeh et.al.[2]). In addition to color, the textile wastewater the characteristics of high pH, high COD and low biodegradability (Mohammed[3]). Textile wastewater has a dramatic environmental impact and cannot be directly discharged to a receiving water body (Marrot & Roche[4]); therefore, textile wastewater treatment is mandatory due to the high water consumption rate and the environmental impacts (Chaia-Hung et.al.[5]).

The major sources of color in textile effluents are the dyes from the dyeing operations. There are three groups of textile dyes dependent on their state in solution as well as their charge: group N; neutral dyes such as disperse, vat and Sulphur dyes which are insoluble in water; group C; cationic dyes like basic dyes and group A; anionic dyes that is to acid, direct and reactive dyes which are soluble in water (Ahmad et.al.[6]). Furthermore, as a result of low biodegradability of most of the dye groups used in textile industry, biological treatment by activated sludge is not always effective due to aerobic biological treatment and oxidizing agent’s resistance (Jiraratananon et.al.[7]). An advanced treatment technique is required, especially if the treated wastewater will be recycled/reused (Kapadan & Kargi [8]). Membrane separation is an optimal solution to remove color, COD and salinity (Aouni et.al.[9]).

Nanofiltration (NF), is a newly developed membrane technology for various water treatment and purification purposes (Koyuncu et.al.[10]); (Valentina et.al.[11]). Although, membrane processes require proper pretreatment techniques, many approaches have been studied to minimize membrane fouling and reduce the costs (Koyuncu et.al.[12]). Pretreatment of feed water includes optimization of operating conditions such as pH as well as chemicals consumption and recovery ratio (Hong & Elinlech [13]; Koyuncu et.al [14]). Microfiltration (MF) and ultrafiltration (UF) can be used as a pretreatment step for Nanofiltration (Gozalvez-Zafrilla et.al.[15]; Debik et.al.[16]; Ong et.al.[17]; Barredo-Damas et.al.[18]; Alventosa-delara et.al.[19]).

Many studies have been carried out on color removal from textile wastewater by Nanofiltration processes. Lebrun et al., investigated electric field enhanced Nanofiltration process by BQ01 and NF45 membranes in dye removal. The results showed variations in dynamic permeability in the presence of electrolytes and according to the electrical potential applied (Lebrun et.al. [20]). Fuchs et al., studied membrane bioreactor (MBR) performance in textile wastewater treatment, and explored its ability to achieve water quality meeting reuse criteria. COD removal was found to vary between 60 and 95 % and COD levels reduced at lower volumetric loading rates that was tested (Fuchs et.al. [21]). Das Gupta et al., studied Nanofiltration with MWCO = 400 Dalton to treat the effluent from a textile plant. Reactive black and red dyes were used and the membrane rejection was up to 92-94 % of the two dyes were achieved respectively, while COD removal was up to 94 % in cross-flow cell (Das Gupta et.al.[22]).

De et.al, studied unstirred batch and cross flow Nanofiltration with MWCO = 400 Dalton to separate dye from aqueous solutions. Using the experimental results, and model parameters (i.e. the diffusivity of the solute (D) and real retention (R_r)) of the membrane were evaluated by optimizing the experimental flux and permeate concentration profiles (De et.al. [23]). Chaudhari et.al, investigated the color removal of commercially azo dyes under anaerobic conditions in wastewater. Color removal was achieved up to 99 % in both the dye containing reactors, whereas COD removals were up to 95 %, 92 % 94 % in control, orange and black dye containing reactors, respectively (Chaudhari et.al.[24]).

Woei-Jye & Ismail[25], studied the performances of some commercial NF membranes and examined dye rejection, permeate flux and COD rejection as well as studying the transport mechanisms in NF membrane with a brief review of transport models of NF membrane. Jahangiri & Aminian[26], compare experimental results with neural network simulation in rejection estimation for two different nanofiltration membranes (NF-90 & DK-5). Arlindo & Isolina[27], studied the influence of osmotic pressure and solute adsorption on permeate flux during nanofiltration (NF) in dye wastewater treatment. He also predicts the rejection coefficient using the solution-diffusion model (Tahri et.al. [28]).

2. Methodology

The adopted methodology to come up with the performance indicators of applying nanofiltration membranes for dye wastewater is as follows:

- Collection and compiling of nanofiltration (NF) current practice in dye wastewater treatment.
- Determination of the governing process parameters.
- A mathematical correlation through multivariate regression to estimate the performance of using NF membrane in dye wastewater treatment.
- Validation of the regression model using published cases.

3. Results and Discussion

NF data for dye wastewater treatment have been reviewed and analyzed according to technical characteristics. Different data have been collected and excluded due to lack of total information needed regarding reactive dye separation using nanofiltration membranes. Table (1), presents compiled data regarding NF separation systems. The collected data have been refined to determine the parameters governing the separation process. Membranes used in determining this approach are NF90, NF 270, NF 200, SR90, DK2540F and 40-40 TS80 TSF. Multiple regression method is used to predict nanofiltration dye rejection and COD in the permeate values. The correlation developed are based on published data (Arlindo et.al.[27]; Chia-Hung et.al.[5];

Table (1) Different NF membrane types characteristics for dye wastewater treatment

Dye type	NF membrane Type	pH	Feed water conductivity (ms/cm)	Flux (l/m^2h)	Transmembrane Pressure (bar)	Membrane pore size (Da)	COD_{in}	COD_{out}	Dye rejection	Ref
reactive dye	DK2540F	10.6	17	49	12	200	2450	612	98	[30]
reactive dye	DK2540F	9.2	14	40.5	12	200	2160	324	95	[31]
reactive dye	NF200	7.1	6	1.5	15	250	708	212	97	[32]
reactive dye	NF270	7.1	6	1.1	15	250	708	140	98	[32]
reactive dye	SR90	7.1	10	50	14	250	890	700	50	[33]
reactive dye	NF90	7.1	10	50	14	250	890	448	75	[33]
reactive dye	40-40 TS80 TSF	6.5	42	9	5	200	20	9	90	[34]

Hassani et.al.[29]; Woei-Jye & Ismail [25]; Tahri et.al.[28]; Najmeh et.al.[2]; Jahangiri & Aminian [26]; Ahmed et.al.[6]) assessed and assembled to formulate the regression model. Eight parameters were selected and investigated, six are considered as independent variables of the multivariate analysis, and two dependent variables. The independent variables are membrane flux, pH, dye rejection, trans-membrane pressure, membrane pore size, COD (in feed and permeate). Tables (2) and (3) shows selected compiled data used for model formulation, showing a strong relationship between the investigated variables and the estimated values.

The final forms of the formulated correlation are mentioned below in equation (1) and (2)

$$\mathbf{Dye\ Rejection}\ (\%) = (\mathbf{-880.248}) + (\mathbf{104.552 * Cond}) + (\mathbf{-25 * Flux}) + (\mathbf{914.11 * Pore\ size}) + (\mathbf{-56.218 * pH}) + (\mathbf{24.686 * COD\ feed}) \quad \textbf{...... (1)}$$

$$\mathbf{COD\ rejection = 82655.46 + (5618.55 * pH) + (-912 * Feed\ Conductivity) + (154.8 * Flux) + (-404.63 * Pore\ size) + (-21.12 * Feed\ COD) + (-10.08 * Dye\ rejection)} \textbf{..... (2)}$$

Where:

Cond.= Feed conductivity (ms/cm)/10,

Flux = Flux (l/m^2h)/10,

Pressure = Pressure (bar) /10,

COD feed = COD feed/100, and

Pore size = Pore size (Da)/200

Table (2) Coefficient of determination (R2) and the error Probability (P-value), Summary output for Dye Rejection estimate using NF (Chia-Hung et.al.[5]; Hassani et.al. [29]; Tahri et.al.[28], Ahmed et.al.[6])

Regression Statistics				
Multiple R	0.91514			
R Square	0.857481			
Adjusted R Square	-0.97511			
Standard Error	17.67767			
Observations	7			
ANOVA				
	df	***SS***	***MS***	***F***
Regression	6	1610.357	268.3929	1.030629
Residual	1	312.5	312.5	
Total	7	1922.857		
	Coefficients	***Standard Error***	***t Stat***	***P-value***
Intercept	-880.248	34379.25	-0.0256	0.983704
pH	-56.2185	2188.507	-0.02569	0.98365
Feed Conductivity	104.5524	3720.019	0.028105	0.982112
Flux	-25	625	-0.04	0.974549
Pressure	0	0	65535	
Pore size	914.1117	33370.39	0.027393	
COD feed	24.68629	854.8369	0.028878	0.981621

The results showed that the rejection of reactive dyes by NF membranes highly dependent on the variation in the salt content of the feed stream and the membrane type (pore size) and independent on the applied pressure. These results are compatible with the published results of (Chollom et.al.[35]). Salt rejections and permeate flux rates are dependent on feed pressure, however, for the studied range; no influence was noticed on the studied variables.

Table (3) Coefficient of determination (R2) and the error probability (P-value), Summary output for COD value estimate in permeate by using NF

Regression Statistics			
Multiple R	1		
R Square	1		
Adjusted R Square	65535		
Standard Error	0		
Observations	7		
ANOVA			
	df	***SS***	***MS***
Regression	8	380845.4286	47605.67857
Residual	0	0	65535
Total	8	380845.4286	
	Coefficients	***Standard Error***	***t Stat***
Intercept	82655.45788	0	65535
-	0	0	65535
pH	5618.553443	0	65535
Conductivity	-9120.01731	0	65535
Flux	1548	0	65535
Pressure	0	0	65535
Pore size	-80926.39431	0	65535
COD feed	-2112.941251	0	65535
Membrane Rejection	-10.08	0	65535

Figure (1), shows the deviation percent between estimated values and published case studies/data (Nur et.al.[36], Arlindo et.al.[27], Chia-Hung et.al.[5], Hassani et.al.[29], Mehmet et.al.[37], Woei-Jye [25], Tahri et.al.[28], Aouni et.al.[30], Najmeh et.al.[2], Jahangiri & Aminian[26], Chollom et.al.[35], Reza et.al.[33], Najmeh et.al.[34], and Ahmed et.al. [6]). The values show that the percent deviation for COD value estimation is almost neglected, while for rejection estimation of about 15%.

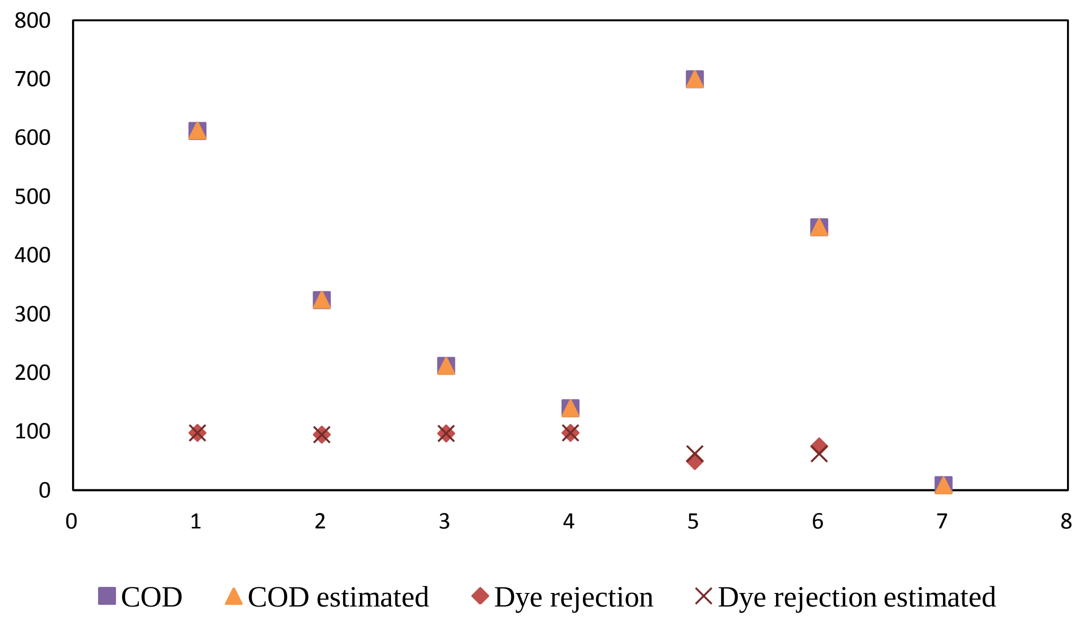

Figure (1) Comparison between estimated and published values

4. Conclusion

Due to the environmental concerns regarding treatment and recycling of dye wastewater. Nanofiltration presents a reliable option for dye wastewater treatment and presenting an option of water recycling. Multivariate regression models have been developed to predict membrane performance regarding reactive dyes rejection as well as COD concentration in the permeate stream. Eight parameters have been selected and investigated; six of them set were as the independent variables of the multivariate analysis and two dependent variables. Validation of the presented models by comparing the results with published data to give a reliable estimation on COD and rejection values of the membrane used under the pre-mentioned parameters.

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