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OPTIMIZATION OF PES-SURFACTANT MEMBRANES USING RESPONSE SURFACE METHOD

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ABSTRACT

In this study, Sodium Dodecyl Sulfate (SDS) and Cetyl Trimethyl Ammonium Bromide (CTAB) surfactant were used to investigate the effect of cationic and anionic surfactant on the performance of dye rejection and permeate flux for the removal of methyl blue dye from aqueous solutions. For this purpose, dye concentration and polymer concentration parameter level was statistically optimized using historical data from response surface methodology (RSM) to determine the optimum conditions for dye rejection and permeate flux. The results revealed high dye rejection efficiency over 99.9 and 99.2 %, accompanied with reasonable permeate flux over 45.4 and 43.9 $L/m^2 \cdot h$ under optimal process conditions for SDS and CTAB surfactant, respectively. The estimated results were elucidated graphically through response surface (RS) plots.

Keywords: Response Surface Methodology (RSM), Surfactant, Nanofiltration Membrane, Optimization

INTRODUCTION

Textile industries traditionally use a huge amount of water which is normally discharged after the wastewater treatment system to decrease the pollution load. To address these challenges, such treatment systems have been enhanced due to increasing in regulatory pressure and demand in cost reduction of water and chemicals[1]. From a number of different textile process streams, this operation allows for the recovery of the valuable chemical components and water. For membrane separation process, a lot of articles have discussed about Ultrafiltration, Microfiltration and Reverse Osmosis and Nanofiltration (NF)[2]. NF frequently becomes the chosen treatment of membrane separation process due to inefficient of conventional treatment systems.

Moreover, nanofiltration offers significant advantages such as lower osmotic pressure difference, higher permeate flux, higher retention of multivalents salts and molecular weight compounds, relatively low investment and low operation and maintenance costs. In terms of dye retention, salt rejection, permeate flux and chemical oxygen demand (COD) retention, many researchers have evaluated the performance of NF membranes[3-5]. Different operating conditions of wastewater and membrane properties have become the factors that have been systematically studied for NF membranes.

For the treatment of textile wastewater, the results have proven that NF membranes are suitable separation process to be employed and generally showed an acceptable rejection. It is necessary to use a suitable pre-treatment in order to prevent fouling and severe module damage to maintain the efficiency of NF membranes at a reasonable operating cost. For every kilogram source in textile refining processes, it will produces substantial amounts of water, mineral salts and reactive dyes. Thus, from its daily operation, they generate a large amount of wastewater which contains complex contaminants[6]. On the other sides, containing unfixed dyes along with salts and auxiliary chemicals such as emulsifying agents, the dyeing process wastewater is a highly colored stream. Difficulties in the effluent treatment arise from its non-degradable property by aerobic digestion. The alternatives ways for effluent treatment of dyes wastewater is through membrane filtration processes. The fouling of NF and RO membranes has been widely investigated throughout the recent years. Thus, modification on the membrane has been developed in order to prevent the fouling phenomena[7].

RESPONSE SURFACE METHODOLOGY

Response surface methodology (RSM) is a series of statistical and mathematical practices that are employed for the progress, enhancement and optimization of certain processes in which a response of interest is affected by several process varieties and the objective is to optimize this response. RSM has a multitude of application selections in the design, development and formulation of new processes and products, or the improvement of an existing product[8–13]. RSM provides more data from a reasonably little number of experiments as compared with conventional optimization procedures, which is less costly and time consuming[14].

In precise, in more multifaceted treatment systems such as photochemical advanced oxidation processes, collaborative and synergistic effects are quite common making these applications suitable candidates for RSM[15]. In this present study, the historical data of response surface methodology type design was employed to analyze the effect of surfactant types on nanofiltration membrane for textile wastewater because it effective for optimize the operating parameter for a process without limitation in the number of design factors besides utilize the data that already exists.

MATERIALS AND METHODS

Materials

Polyethersulfone (PES) was supplied from SOLVAY Advanced Polymers Company (RADEL A-300). 1-Methyl-2-Pyrrolidone (NMP) with analytical purity of 99.5%, ethanol, n-hexane, Polyethylene Glycol (PEG) 600 g/mol and Methyl blue dye was supplied from Merck. Sodium Dodecyl Sulfate (SDS) and Cetyl Trimethyl Ammonium Bromide (CTAB), which were purchased from EMD Chemicals.

Membrane Preparation and Fabrication

NMP was heated at about 50 °C and PES was inserted gradually. PEG 600 was added when polymer has been dissolved. The solution was stirred for 8 hours to get uniform solution. Then, surfactant (17, 19, 21 wt%) was added 3 hours before the polymer solution was finished. Casting process will take place in fabricated thin film of membrane.

Slight amount of polymer solution was poured onto glass plate as support layer with casting knife setting at 150 µm. After the membrane has been casted, the glass plate was submerged into the coagulation bath. The membrane was submerged in water bath for 24 hours before it was submerged in ethanol for another 24 hours. Finally, the resulted membrane was drenched in n-hexane for 2-3 hours before dried at room temperature at least for 24 hours.

Membrane Performance Evaluation

The rejection characteristics were described by determining observed rejection, R_{obs} and real rejection, R_{real}. In the membrane separation processes, the concentration on the membrane surface is always higher than in the bulk. A concentration on the membrane surface is not directly obtained from experiment thus the following equation is applied:

$$R = \left(1 - \frac{Cp}{Cf}\right) \times 100\% \quad (1)$$

Where, C*p* represents the methyl blue concentration (mg/L) in the collected permeate sample and the concentration of the dye in the feed solution (mg/L) is denoted by C_f. The permeation flux (J_w) was calculated as:

$$Jw = \left(\frac{Q}{At}\right) \quad (2)$$

Where, J_w denotes the permeation flux (L/m^2 h), Q represents the volume (L) of the permeate collected in sampling time t (h), and A refers to the effective membrane surface area (m^2).

Response Surface Methodology

The main objective of RSM is to determine the optimum operational conditions of the system. The application of statistical experimental design techniques in adsorption process development can result in improved product yields, reduced process variability, reduced development time and overall costs[16]. In RSM, the technique used was RSM historical data where the experiment was run according to the sequence as suggested in Design Expert software.

RESULTS AND DISCUSSION

The experimental design has been developed to 9 different experimental conditions and both responses of the experiments for both SDS and CTAB surfactant are presented in Table 1.

Table 1: Experimental design matrix and the value of dye rejection and flux responses

Run	Polymer Concentration (%)	Dye Concentration (ppm)	Surfactant Types			
			SDS		CTAB	
			Rejection (%)	Flux ($L/m^2{\cdot}h$)	Rejection (%)	Flux ($L/m^2{\cdot}h$)
1	17	10	98.3	43.962	99.2	44.565
2	17	15	99.3	36.692	98.2	28.396
3	17	20	99.2	19.360	98.7	14.837
4	19	10	98.8	38.513	99.5	32.395
5	19	15	99.2	24.621	98.6	27.953
6	19	20	99.2	17.836	99.6	12.255
7	21	10	99.2	14.491	99.5	13.682
8	21	15	98.7	13.832	99.2	9.432
9	21	20	99.2	12.135	99.9	8.972

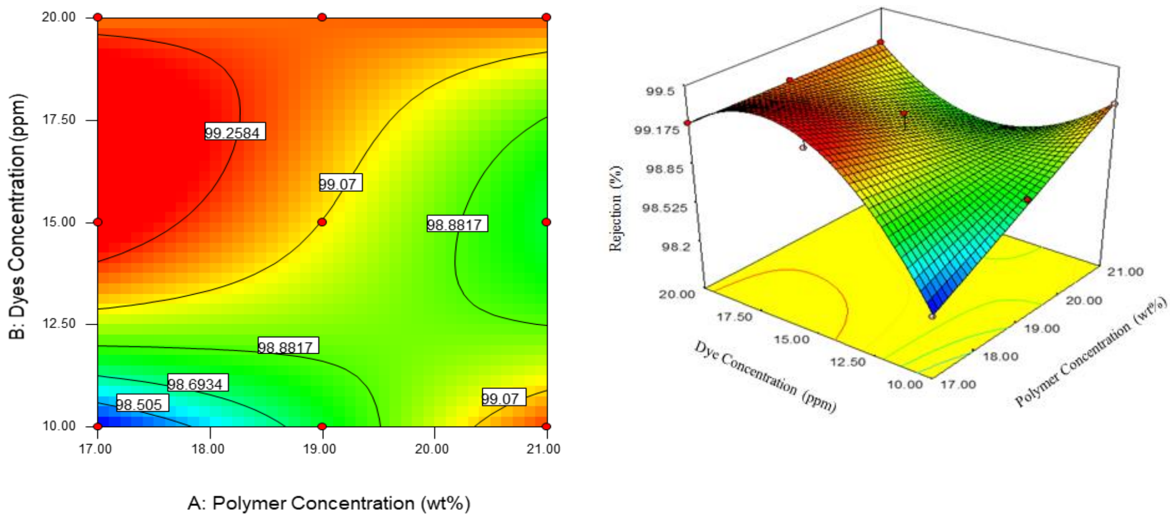
Statistical Analysis

In order to confirm a suitable model fit, the significance of the regression model need to be examined[11,17]. A "Prob>F" value less than 0.05 indicates that the design model is statistically significant. It is obvious that the Prob>F values of the models are varying between 0.0012 and 0.0182, indicating that the models perceived for the surfactants SDS and CTAB are significant. The precision of a model can be examined by the determination of the coefficient of variation (R^2). The R^2 values for SDS and CTAB dye rejection efficiencies were analyzed as 0.9688 and 0.9577, respectively. Meanwhile The R^2 values for SDS and CTAB permeate flux were evaluated as 0.9412 and 0.9488, respectively.

Interactive Effects of Process Independent Variables

The behaviors of the quadratic model approximated response surface with rejection as the response, when subjected to the concentration of dye and polymer concentration, was graphically represented by means of the 2-dimensional contour and 3-dimensional response surface plots shown in Figure 1. The dye rejection efficiency (%) for SDS and CTAB, have been demonstrated by the response surface (RS) plots shown in Figure 1 and Figure 2 respectively. In each of the cases, the dye rejection efficiency for SDS and CTAB is ranged from 98.3% and 99.9%; the significantly high dye rejection, was attributed to the higher polymer concentration thus thickening the membrane surface that could retain high number of dye molecules[18].

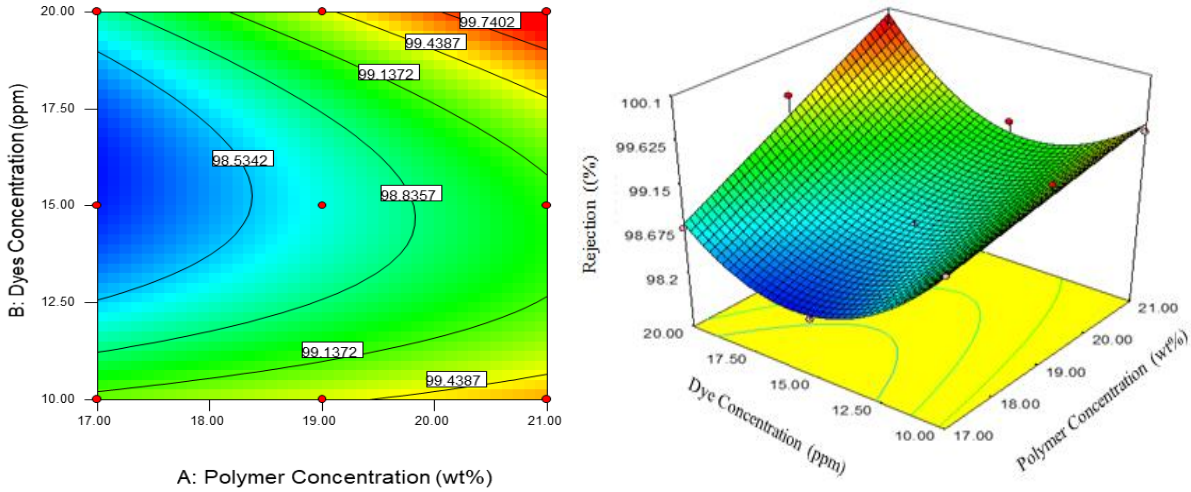
From Figure 1 it is obvious that dye concentration values had a vivid positive effect on rejection rate of SDS surfactant formulations. Improving the dye concentration of the SDS surfactant solution resulted in a succession of dye rejection efficiencies in the investigated ranges. This can be explained by the effects of steric hindrance and electrostatic action that influence the rejection characteristic of the nanofiltration membrane charged to solute charged.



(a) (b)

Figure 1: (a) 2D contour and (b) 3D Plot showing interactive effects of varying dye concentration and polymer concentration values on percent dye rejection efficiencies for SDS surfactant

Figure 2 shows that the highest rejection rate is at maximum point of dye and polymer concentration. The increasing of dye rejection is due to the adsorption of dye molecule on the membrane surface where first effects the permeation flux and increases the solution rejection. The adsorption occur was the affects from the physicochemical interactions or due to charges between dye solution and membrane surface thus can be concluded that, tallying of cationic surfactant in polymer solution will gives great impact by producing membrane with high performance and experimental data shows that higher rejection was due to the adsorption of molecules on the membrane surface[19].



(a) (b)

Figure 2: (a) 2D contour and (b) 3D Plot showing interactive effects of varying dye concentration and polymer concentration values on percent dye rejection efficiencies for CTAB surfactant

Optimization

Optimization process was done by RSM to determine the optimum operating conditions for dyes removal under which the removal efficiency reach its maximum point. The predicted and experimental efficiencies for optimal operating conditions are shown in Table 2.

Table 2: Optimum conditions for dye rejection

Optimal Operating Conditions	Surfactants	
	SDS	CTAB
Desirability	0.884	0.747
Polymer Concentration (wt%)	17	17
Dye Concentration (ppm)	15	10
Predicted rejection by RSM	99.1	99.0
Obtained rejection by experimental	99.2	99.3

CONCLUSION

In this research work, the application of response surface method for modelling the polymer and dye concentration using SDS and CTAB surfactant on dye rejection efficiencies and permeate flux is successfully developed. Analysis Of Variance (ANOVA) disclosed that the recognised factorial design models were statistically significant and described dye rejection and permeate flux at satisfactory levels. From the 2D contour and 3D plots displaying the collaborative interactions between polymer concentration, dye concentration and dye rejection, it is considered that for SDS surfactant dye concentration had significant effect on dye rejection. Meanwhile, for CTAB surfactant, dye and polymer concentration had a dramatic positive effect on rejection rate.

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