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Estimation of solute transport parameter and mass transfer coefficient in nanofiltration for solvent-diluted palm oil

K M Lim^{1,2} and N F Ghazali^{3*}

¹ Conversion & Separation Technology iKohza (SHIZEN), Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Malaysia

² Malaysia-Japan International Institute of Technology (MJIT), Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Malaysia

³ Department of Bioprocess Engineering, School of Chemical and Energy Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

*nazlee@utm.my

Abstract. Membrane technology has been gradually used as an alternative to the conventional purification method in various industries. In the advancement of membrane technology, important elements such as volumetric flux, rejections, mass transfer and interaction parameters have been frequently considered, particularly in the prediction of membrane performance. In this study, an estimation of mass transfer and solute transport parameters in organic solvent was conducted by using the Levenberg-Marquardt least squares method. Combined film theory/ solution-diffusion model was selected and was correlated with experimental data to estimate the parameters. The mass transfer and solute transport parameters for the nanofiltration of acetone-diluted palm oil in SolSep 030306 membrane were successfully estimated. The correlation of mass transfer coefficient with feed concentration and stirrer speed were additionally described.

1. Introduction

In recent years, solvent resistant nanofiltration or organic solvent nanofiltration (OSN) has becoming theoretically practicable through the development of solvent resistant commercial polymeric membranes. Membrane technology has a great potential to match with the conventional energy-intensive process such as distillation. As membrane nanofiltration is able to separate organic solvents and solutes in the range of 200 Da to 1000 Da, it is often compared with distillation processes in the literature [1]. The properties of the nanofiltration membrane lie between ultrafiltration and reverse osmosis membrane. Nanofiltration membranes are usually composites of polymer layers with a selective layer of thickness which is approximately 1 μm that controls all rejection and flow properties [2]. The development of nanofiltration technology has contributed to the rise of its application in industries such as separation of pharmaceuticals from fermentation broth [3], metal recovery from wastewater [4], separation of constituents from vegetable oil [5] and virus removal [6]. According to Bowen and Welfoot [6], predicting the performance of such separations is crucial in process design.

However, the performance of nanofiltration membrane is harder to predict with organic solvents as compared to aqueous solutions [7]. In the literature, mathematical models for nanofiltration or reverse osmosis are either based on irreversible thermodynamics or transport models. The transport models include the solution-diffusion model, solution-diffusion imperfection model, Kimura-Sourirajan



model, and Spiegler Kedem model [8]. According to Marchetti et al. [9], solution-diffusion model has been frequently employed in the prediction of performance in various systems whilst Spiegler-Kedem model has been used in the data prediction of single solute and solvent transport in reverse osmosis and nanofiltration system. The partial rejection of various compounds by reverse osmosis and nanofiltration membranes have motivated researchers to develop ways and methods to predict the parameters [10]. Although there are many studies have been published regarding solvent resistant nanofiltration and its membrane transport modeling, the studies on the hydrodynamics and mass transfer characteristics in OSN stirred cell modules (as shown in Figure 1) is scarce [11].

In order to provide the solution to the problem, the incorporation of experimental and model is recommended in computer-aided process engineering, in which, laboratory experiments will be used as a tool to generate experimental data that can be used for modeling and simulation verification [1]. Additionally, although the trend of operational parameters such as stirrer speed and feed concentration against mass transfer coefficient and solute transport parameter can be generally known, the membrane materials can show either positive or negative correlation [12], which depends on the interaction of membrane, solvent or solute [8]. There is currently no discussion in the literature on the estimation of mass transfer coefficient in solvent-diluted palm oil by using MATLAB. Thus, this study investigates the feasibility of custom-made MATLAB codes in estimating the mass transport parameter and solute transport parameter (as shown in Section 2.2.3), as well as its applicability in estimating parameters for solvent-diluted palm oil in SolSep 030306 nanofiltration membrane.

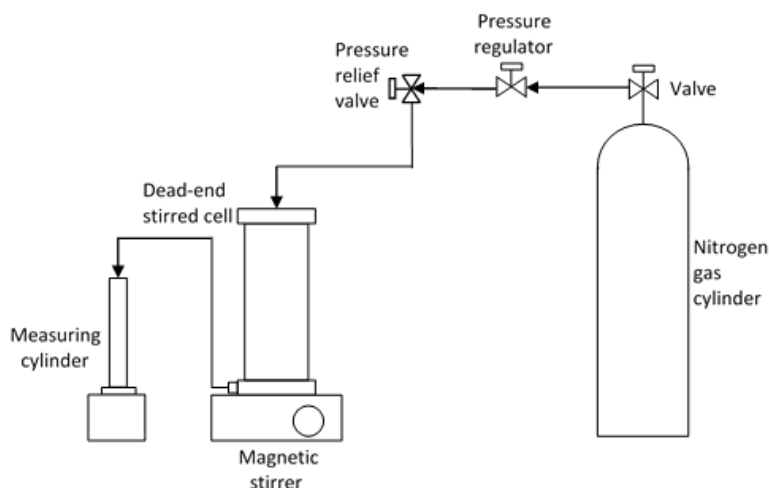


Figure 1. Schematic representation of dead-end stirred cell.

2. Methodology

2.1. Experimental materials and procedure

Food grade palm oil was purchased from a local supermarket (Johor, Malaysia). Acetone was purchased from Sigma-Aldrich, Germany (purity>99%). The experimental set-up can be seen in Figure 1. The membrane housing, which is a stainless-steel cell fastened with high tensile bolts. Porous stainless steel membrane support disc is available to prevent membrane rupture at high operating pressures. The membrane housing has grooves for the placement of 'O' ring, at the top and bottom of the cell, to avoid leakage during high operating pressure experiments. The membrane cell employs a circular type of flat membrane. Membrane NF 030306 from SolSep BV, the Netherlands with molecular weight cut-off of 1000 Da [13] was used throughout the experiments. The type of polymer for 030306 membrane remains unknown [14]. The effective membrane diameter is 51mm after measurement by using a vernier caliper. Before performing the actual experiments for the rejection of triacylglycerides, 030306 membrane was subjected to solvent flux stabilization at 30 bar.

This was done in order to prevent membrane collapse during the course of experiments. The duration of the experiment was according to the volume collected, whereby 10 mL of permeate sample was collected for each experimental run. The measurements of triacylglycerides concentration in the samples were determined gravimetrically by using an electronic weighing scale (BSA24S-BW, Sartorius GmbH, Germany). The experiments were performed at an applied pressure of 10 bar, 20 bar, and 30 bar, whilst the feed concentration was varied at 5 wt%, 10 wt% and 20 wt% of palm oil with respect to acetone and stirrer speed at 100 RPM, 170 RPM, and 400 RPM. All experiments were performed in batch mode. 1 mL of the total feed was kept in the oven to eliminate solvent residues and was weighed to obtain feed concentration. The corresponding rejection and permeate flux were also measured.

Observed rejection, R_0 can be calculated as follows:

$$R_0 = \left(1 - \frac{C_P}{C_R}\right) \times 100\% \quad (1)$$

where C_P and C_R are the final concentration of solute in permeate and retentate respectively [15].

2.2. Membrane Transport Model

2.3. Film theory

During the process of separation, solute will accumulate at the membrane interface, and this phenomenon is known as concentration polarization. According to film theory, the material balance of the solute using an appropriate boundary condition can be written as follows [16,17]:

$$\left(\frac{C_{A2} - C_{A3}}{C_{A1} - C_{A3}}\right) = \exp\left(\frac{J_v}{k}\right) \quad (2)$$

where the mass transfer coefficient is equal to the diffusivity of solute A in solvent B across the concentration boundary layer (D_{AB}/l).

Observed rejection, R_0 can be written as

$$R_0 = \frac{C_{A1} - C_{A3}}{C_{A1}} \quad (3)$$

and true rejection, R can be written as

$$R = \frac{C_{A2} - C_{A3}}{C_{A2}} \quad (4)$$

By substituting equation (3) and equation (4) into equation (2), the following expression is obtained

$$\frac{R_0}{1 - R_0} = \left(\frac{R}{1 - R}\right) \left[\exp\left(\frac{J_v}{k}\right)\right] \quad (5)$$

2.4. Combined film theory/ solution-diffusion model

A widely known transport model for the permeation in the polymeric membrane is known as the solution-diffusion model, which is written as

$$J_v = A(\Delta p - \Delta\pi) \quad (6)$$

$$J_A = \left(\frac{D_{AM} K}{\delta} \right) (C_{A2} - C_{A3}) \quad (7)$$

where A is the permeability parameter of the solvent and $(D_{AM} K / \delta)$, solute transport parameter, can be considered as a single parameter.

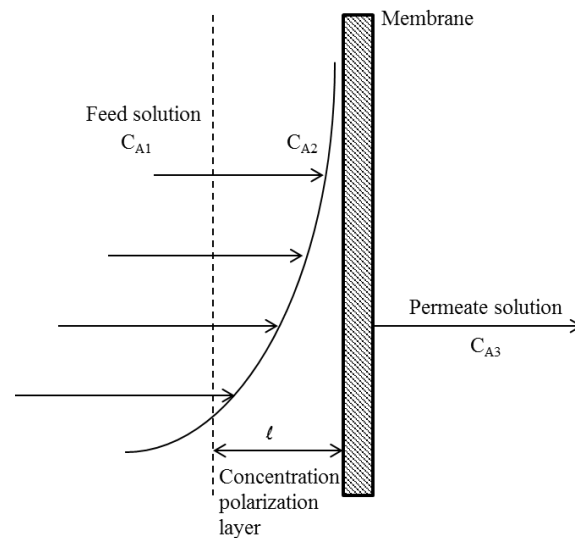


Figure 2. Schematic showing the concentration profile of solute in the feed and permeate solution.

In order to consider the effect of concentration polarization, equation (5) can be combined with equation (6) and equation (7), to give [17]:

$$\frac{R_0}{1 - R_0} = \left[\frac{J_V}{\left(\frac{D_{AM} K}{\delta} \right)} \right] \left[\exp \left(\frac{-J_V}{k} \right) \right] \quad (8)$$

where R_0 is the observed rejection, k is the mass transfer coefficient at the boundary layer, J_V is the volumetric flux, and $\frac{D_{AM} K}{\delta}$ is the solute transport parameter. The data of observed rejection and permeate flux taken at different pressures can be used to estimate solute transport parameter $(D_{AM} K / \delta)$ and mass transfer coefficient, k .

2.5. Parameter estimation of the mass transfer coefficient and solute transport parameter

In the literature, the estimation of model parameters is depending on the mathematical model which has been validated with experimental data [18]. Mass transfer coefficient, k , is the model parameter that can be used to characterize the concentration polarization in membrane transport [19]. In this work, the mass transfer coefficient and solute transport parameters that were estimated by using MATLAB were compared with the experimental and numerical results obtained by Murthy and Gupta [17] in order to verify the feasibility of MATLAB in estimating parameters. Combined film theory/solution diffusion (CFSD) model was used in this study. MATLAB nonlinear parameter estimation was performed for 3 sets of data by using Sourirajan's data as shown in Table 1.

Table 1. Experimental Data (NaCl-Water)[20].

Feed concentration 0.2 M (=1.2262%)					
$\frac{D_{AM}K}{\delta} = 7.5 \times 10^{-4}$, cm/s					
$k = 30 \times 10^{-4}$, cm/s		$k = 57 \times 10^{-4}$, cm/s		$k = 100 \times 10^{-4}$, cm/s	
R ₀	$J_V \times 10^4$, cm/s	R ₀	$J_V \times 10^4$, cm/s	R ₀	$J_V \times 10^4$, cm/s
Set 1		Set 2		Set 3	
0.597	33.553	0.718	34.357	0.769	34.382
0.599	30.446	0.709	31.104	0.757	31.469
0.598	27.266	0.698	27.778	0.741	27.997
0.593	24.050	0.683	24.452	0.723	24.635
0.584	20.797	0.663	21.089	0.698	21.126
0.569	17.471	0.636	17.580	0.667	17.654
0.544	14.072	0.599	14.145	0.625	14.181
0.503	10.673	0.546	10.709	0.566	10.673

Table 2. Parameters estimated by using CFSD model by Murthy and Gupta [17] and method from this study (MATLAB) at 95% confidence interval (CI).

Model parameter	Estimation run 1			Estimation run 2			Estimation run 3		
	Murthy and Gupta (1997)	This Study	95% CI	Murthy and Gupta (1997)	This Study	95% CI	Murthy and Gupta (1997)	This Study	95% CI
$\frac{D_{AM}K}{\delta} \times 10^4$, cm/s	7.386	7.386	7.355-7.417	7.392	7.393	7.364-7.420	7.383	7.432	7.339-7.524
$k \times 10^4$, cm/s	29.960	29.960	29.807-30.113	57.034	57.036	56.578-57.492	99.920	103.20	98.368-108.029

2.6. Concentration polarization modulus and Peclet number

According to Murthy and Chaudhari [21], concentration polarization must be taken into consideration in reverse osmosis, nanofiltration or ultrafiltration membrane model. The inclusion of concentration polarization will enable the determination of true rejection at the membrane surface. In order to measure the magnitude of concentration polarization, the concentration polarization modulus (C_{A2} / C_{A1}) [22], must be determined. When the concentration polarization modulus is unity, the effect of concentration polarization is negligible, however, as the concentration polarization modulus deviates from unity, the membrane selectivity and flux will be greatly impacted by concentration polarization [22]. The modulus can be written as follows [21,22]:

$$\frac{E_0}{E} = \frac{C_{A2}}{C_{A1}} = \frac{\exp(J_V/k)}{1 + E_0[\exp(J_V/k) - 1]} \quad (9)$$

whereby E_0 is enrichment factor, which can be also be written as C_{A3} / C_{A1} ; E is true enrichment factor, can also be written as C_{A3} / C_{A2} . Peclet number is written as the convective transport, J_V , divided by diffusive transport, k [21].

3. Results and Discussion

3.1. Membrane transport parameters and mass transfer coefficient estimation

Observed rejections and permeate flux data obtained from the experiment were supplied to the MATLAB R2013a to estimate the parameters, based on the Levenberg-Marquardt method. The observed rejections and permeate flux data were obtained by keeping the concentration of feed constant, whilst varying the operating pressures for each set of data. The membrane transport parameters and mass transfer coefficient were estimated and fitted to the membrane transport models. The mass transfer and solute transport parameters in this study were estimated by using the CFSD model. The results were tabulated in Table 3.

Table 3. Parameters obtained in this study at different concentrations and stirrer speeds (applied pressure 10 bar).

Set No.	Feed concentration (wt %)	Stirrer rotational speed (RPM)	$k \times 10^5$ (cm/s)	$\frac{D_{AM}K}{\delta} \times 10^5$ (cm/s)	Permeate flux $\times 10^5$ (cm/s)	Concentration polarization modulus (C_{A2}/C_{A1})	Peclet number (J_V/k)
1	5	100	88.95	30.41	52.22	1.5891	0.5871
2	5	170	140.3	39.62	66.39	1.4900	0.4732
3	5	400	143.9	37.96	80.56	1.6330	0.5597
4	10	100	52.07	17.65	34.44	1.7093	0.6615
5	10	170	80.10	22.39	38.89	1.5406	0.4855
6	10	400	80.25	20.56	40.56	1.5823	0.5054
7	20	100	23.27	6.190	17.50	2.0194	0.7519
8	20	170	31.39	8.145	24.44	2.0858	0.7787
9	20	400	37.45	9.589	31.67	2.2554	0.8455

3.2. Effect of feed concentration

Experiments were performed to investigate the effect of feed concentration from 5 wt% to 20 wt%. As the feed concentration increases, the boundary layer thickness increases due to the increase in retained components at the membrane surface [23]. The boundary layer thickness can also be characterized quantitatively by concentration polarization modulus [21], as can be seen in Table 3. The increase in boundary layer thickness causes the permeation flux to decrease. Furthermore, according to the mass transfer correlation by Bowen *et al.* [24], the mass transfer coefficient is inversely proportional to viscosity. As the feed concentration increases, the viscosity increases, and thus the mass transfer coefficient decreases, which can be seen in Figure 3.

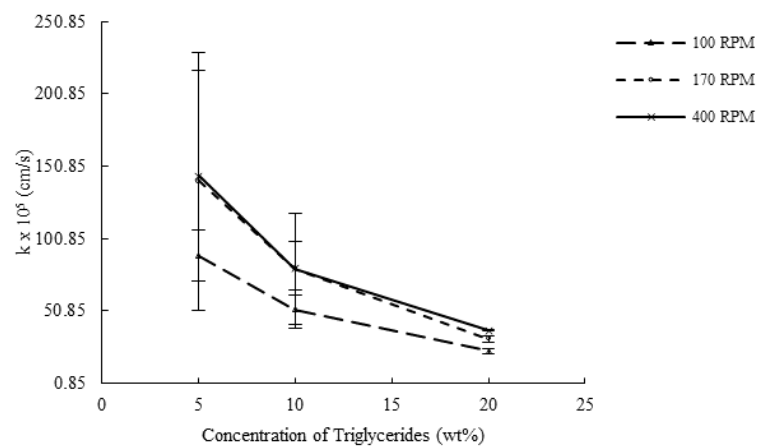


Figure 3. Graph of mass transfer coefficient (k) versus concentration of triglyceride.

3.3. Effect of stirrer rotational speed

Figure 4 shows the trend of mass transfer coefficient obtained from estimation by varying rotational speed (100 to 400 RPM) for 5 wt% to 20 wt% feed concentration. According to Strathmann [23] the increase in stirrer rotational speed decreases the boundary layer thickness and subsequently increases the membrane flux. However, Figure 4 shows that the increase in rotational speed does not significantly affect the mass transfer coefficient at 20 wt% feed concentration. According to the mass transfer correlation by Bowen [24], as the feed concentration increases, the viscosity increases, and thus, resulted in the decrease in mass transfer coefficient.

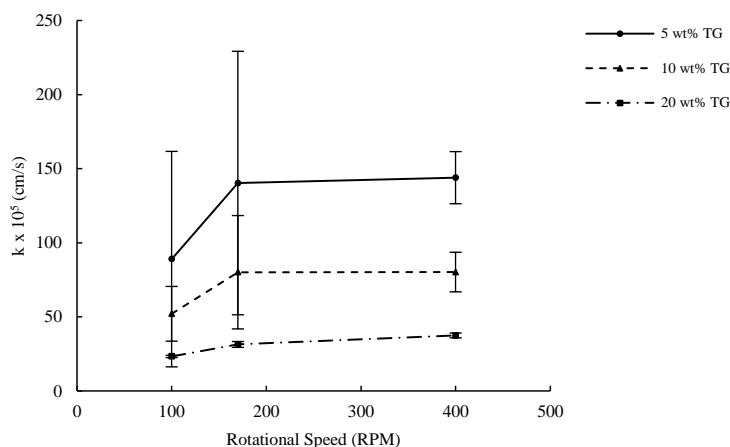


Figure 4. Graph of mass transfer coefficient (k) versus stirrer rotational speed.

4. Conclusion

The mass transfer and solute transport parameters were successfully estimated by using MATLAB software with Levenberg-Marquardt least squares method. The data comparison between literature and this study shows that MATLAB is capable of estimating the parameters at a confidence level of 95%. The experimental study on oil feed concentration and stirrer speed depicted a theoretically acceptable correlation. The results from this work suggested that the mass transfer coefficient decreases with feed concentration when acetone is used as a solvent in 030306 membrane. However, the stirrer speed does not significantly impact the mass transfer coefficient at stirrer speed above 200 RPM. This study successfully incorporates the use of MATLAB in the estimation of mass transfer coefficient in nanofiltration for solvent-diluted palm oil which is useful in the process understanding during membrane scale-up and evaluation.

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