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EmAITJournal homepage: <http://publisher.uthm.edu.my/ojs/index.php/emait>

e-ISSN : 2773-5540

Emerging
Advances in
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The Optimization of RHS-polysulfone Membrane towards Operating Condition for Humic Acid Removal

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DOI: <https://doi.org/10.30880/emait.2021.02.01.002>

Received 19 April 2021; Accepted 10 Jun 2021; Available online 30 Jun 2021

Abstract: This study investigates the effects of rice husk silica (RHS) as additive in the polysulfone membrane to enhance antifouling properties in membrane separation process. The performance (of what?) was evaluated in term of pure water flux (PWF), rejection and antifouling properties. The optimized of normalized flux (J_f/J_o) at different parameter in filtration (pH, ionic strength and transmembrane-pressure) was carried out by using the response surface methodology (RSM). The results showed that the addition of 4 wt. % RHS give the highest flux at 300.50 L/m².hour (LMH). The highest rejection was found at 3 wt. % of RHS membrane with value 98% for UV₂₅₄ and 96% for TOC. The optimal value of J_f/J_o was found at 0.62 with the condition of pH: 6.10, ionic strength: 0.05 mol/L and transmembrane-pressure: 2.67 bars. Optimize of RSM analysis from ANOVA also proved that the error of model is less than 0.05% which indicates that the model is significant.

Keywords: Rice husk, silica, polymeric membrane, fouling

1. Introduction

In recent years, water pollution has become a serious problem that needs to be discussed and paid attention to around the world. According to the World Health Organization (WHO) society, the worst situation faced by most of the people in the world is the lack of access to clean water [1]. Since the world population will increase to 1.1 billion in 2015, pollution from all clean water sources (i.e. river water, rain water, sea water, lakes) is turning into a harmful substance like bacteria, debris floating and dust. Serious problems that need to be fixed. Therefore, many studies have recently been

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conducted to treat water media by different methods, such as activated carbon, activated alumina, aeration, ion exchange, neutralization filters and membranes [2, 3]. According to the extensive research reported in the source article [4], membrane technology provides the simplest method.

Basically, membranes are divided into various types of separation, such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis. Ultrafiltration can be used to remove contaminants from contaminated water with strong regulatory activities and little source of high quality water. Due to their pore size range between 2 and 100 nm, these separation stages can be considered very promising drinking water production processes. Ultrafiltration can remove viruses, bacteria, colloids, and larger particles from suspension [5]. According to Yu et al. [6] polysulfone (PSf) is a very popular polymer, due to its good mechanical, thermal and chemical stability, it is widely used in the manufacture of ultrafiltration membranes. However, due to their hydrophobicity, PSf membranes are prone to fouling due to adsorption of proteins and other biomolecules from the feed stream. This fouling mechanism leads to a decrease in flux through concentration polarization, where the membrane is blocked or covered by certain elements in the stream being processed, thus reducing its outflow or flux. Generally speaking, when contamination occurs during the separation process, it will affect membrane performance, such as flow permeability, water permeability, surface porosity, and morphology[7].

Most previous studies have shown that the addition of additives to membrane formulations can play an important role in preventing contamination problems, which can lead to incorrect morphological structures, including pores and surface layers [8]. The addition of different materials or additives has a significant impact on membrane performance by reducing contamination and increasing the rejection rate. The addition of additives in the membrane formation process is expected to increase the performance value of the membrane, make the membrane porous, increase hydrophilicity, induce antibacterial properties, and improve membrane performance [9]. Incorporation of most inorganic fillers (such as polymer additives, silica, aluminum, zeolite, and titanium dioxide) in the doping solution can produce membranes with higher porosity and hydrophilicity [10, 11].

Due to its biodegradable properties and green technology, the potential of organic fibers from natural sources (non-hazardous elements), such as rice husk (high silica content) is being considered. In fact, some inorganic additives can inhibit large voids, enhance the formation of pores, and improve the interconnectivity of pores and the hydrophilicity of the membrane. Inorganic additives can also increase the permeability of the membrane and control the surface properties of the membrane. As reported by Yan et al. [12] alumina (inorganic) additives used as additives in film coating formulations have enhanced antifouling properties. By increasing the hydrophilicity of the membrane, membrane fouling can be reduced to a certain extent. It is expected that the addition of additives to the membrane formulation will change the pore size, pore size distribution, physical and mechanical properties and other characteristics. As reported in many articles, silica can inhibit the formation of amphiphilic and large hollow components, enhance the formation of pores, and improve the interconnectivity of pores and the hydrophilicity of membranes. Consider adding potential organic fiber, namely rice husk, from natural sources (non-hazardous elements) because it contains a compound with a high silica content. In addition, it also has biodegradable characteristics and provides green technology.

Therefore, in this study, optimization technology is used to properly select the process variables to determine the best parameters. Under this parameter, the effective performance of the humic acid removal process through the RHS-polysulfone membrane can be assumed. The best membrane from previous research [8] is used to check the normalized flux (J_f/J_o) of the separation parameters (pH, ionic strength (IO) and transmembrane-pressure (TMP)) on the performance of the membrane for the scale analysis research. Therefore, the use of process models based on regression methods will show the importance of these parameters, and it is necessary to consider appropriate expressions for these variables. Follow the central compound design method to associate the separation parameters with the measured variables.

2. Experimental

2.1 Factorial Design

The factorial design step consists of an estimation of the parameter effect, designing the model, analysis by using ANOVA, refinement of the model and interpretation of the result. The first step involves the implementation of the first-order model to determine the significant range of parameters and response. The solution of parameters for this operating condition were based on previous study (citation / ref). Parameters such as pH, ionic strength and transmembrane pressure were selected as main parameters. In designing the first-order model, the factorial 2^3 was applied with three replicates at the center point to determine the statistical significance of variable parameter on effecting response of final normalized flux (J_f / J_o). The range of level variables parameters investigated in this study is shown in Table 1. The experimental works were carried out to obtain the measured response within 120 min filtration of HA.

Table 1 - Ranges of parameters used in this study

Parameter	Limit
pH (pH)	3 to 11
Ionic strength (IO)	0.05 Mol/ L to 0.5 Mol / L
Transmembranepressure (TMP)	2 bar to 5 bar

2.2 Response Surface Methodology

The factorial design was furthered augment with the response surface methodology based on central composite design (CCD). CCD is used to generate the prediction of a mathematical correlation model between the significant factors (parameters) with respect to the response (normalized flux, J_f / J_o). CCD consists of 2^n factorial runs, 2 (n) axial runs and center runs (n) where n is the number of factors. The variables parameters labeled as pH (x_1), ionic strength (x_2) and transmembrane-pressure (x_3). The CCD was conducted with 2^3 at six axial points and three replicates at the center point. The rotatable alpha has been applied at the same block with the levels +1.68179, -1, 0, +1 and -1.68179.

The ANOVA approach was applied to investigate the significant effect of the response parameters. In this study, the analysis ANOVA was generated using Design Expert 7. To be noted, ANOVA is a second-order model that enables further optimizing within of parameters studied with curvature response. In this procedure, the response was measured by using a significance value which must less than 0.05. For each variable of pH (x_1), ionic strength (x_2), and transmembrane-pressure (x_3), the statistical significance of the second-order regression models were determined by F-value which is a measurement of data variance, based on the ratio of mean square for each group variance due to error. Good model prediction is presented based on F-value which is the variance ratio and P-value which is the probability. The analysis of ANOVA, correlation of the coefficient of multiple determinations, R^2 must be closed to unity, 1 which represents a good fit model. The significant model must possess high F-value with P-value must 0.05 of F-value.

2.3 Optimization Process

The design Expert 7 with regression analysis to fit the second order degree polynomial equation was applied to analyze the response data obtained from the experiment. All three parameters (pH, ionic strength and transmembrane-pressure) were optimized simultaneously to produce the maximum normalized flux (J_f / J_o) in the range of each parameter. In order to optimize the response, the desirability function (DF) was applied to determine the most desirable model. The DF is represented in the range of 0 to 1 (0 = presenting the completely undesirable and 1 = presenting the completely desirable). The prediction of the optimal point value was obtained from equation (3.10).

$$Y = b_0 + \sum b_i x_i + \sum b_{ii} x_i^2 + \sum b_{ij} x_i x_j \quad (3.10)$$

Where, Y is the response, b is the regression coefficient of the model, x is the coded levels of the independent variables

3. Results and Discussion

3.1 First Order Model - Factorial Design

The effect of various parameters to the fouling performance using response surface methodology (RHS) was successfully conducted. The 4 wt. % RHS membrane was used to examine the fouling performance under the three different of transmembrane-pressure (TMP), pH and ionic strength (IO) condition. The 4 wt. % RHS membrane was selected based on its good performance in pure PWF, rejection, antifouling performance and low membrane resistance. Table 2 presented the result experimental layout with 2^3 full factorial central composite designs (CCD) and the screening process result by investigating the effect and interaction of TMP, pH and IO within 120 min with humic acid filtration. The range of filtration condition was 3-11 for pH, 0.05-0.5 mol/L of KCl for IO and 2-5 bars for TMP.

3.2 ANOVA Analysis and Analysis Normal Probability

Table 3 shows the ANOVA result obtained from the optimization analysis. Result showed that the 2nd order model in this study is significant with F-value and P-value was 49.10 and 0.0001 (less than 0.05) respectively. In fact, for a significant model with coefficients of individuals, the larger the magnitude of the F value, the smaller the corresponding "Prob value > F". From the ANOVA analysis, the main effect source of variables x_1 , x_2 , and x_3 showed that the values were significant. At the same time, the curvature of this model also gives significant value. The curvature analysis indicated that the experiment data is located in the significant region. In addition, the coefficient of determination R^2 of the model was 96.1% that shows the variability of the response of model while remaining 3.9% was not explained. This higher value of R^2 shows that there is a good agreement between experimental and predicted value. Furthermore, the "Lack of fit" value for ANOVA analysis was not significant and therefore the model is considered as desirably fit model [13].

Figure 1 depicts the normal probability of the residual for the variables x_1 , x_2 , and x_3 towards normalized flux of fouling analysis. Herein, Figure 1 indicates that normal probability for residual distribution is nearly in straight line. It demonstrated that the denoting errors are evenly distributed. Therefore, the model proposed adequate and reasonable for free form at any violation of the independence or constant variance assumption [14].

Table 2 - The result experimental layout of 2³ full factorial CCD

Standard	Run	pH	IO	TMP	Final Normalized Ratio (J _f /J _o)
1	1	-1	-1	-1	0.568421
2	7	1	-1	-1	0.543158
3	6	-1	1	-1	0.530526
4	9	1	1	-1	0.505263
5	8	-1	-1	1	0.517895
6	3	1	-1	1	0.48
7	11	-1	1	1	0.48
8	2	1	1	1	0.454737
9	4	0	0	0	0.593684
10	10	0	0	0	0.581053
11	5	0	0	0	0.606316

Table 3 - ANOVA for 2³ full factorial design: response:

Source	Sum of square	DF	Mean square	F Value	p-value prob>F	
Model	609.48	3	203.16	49.1	0.0001	significant
A-A (pH)	100.55	1	100.55	24.3	0.0026	significant
B-B (IO)	150.2	1	150.2	36.3	0.0009	significant
C-C (TMP)	358.74	1	358.74	86.7	< 0.0001	significant
Curvature	950.95	1	950.95	229.83	< 0.0001	significant
Residual	24.83	6	4.14			
Lack of Fit	4.97	4	1.24	0.12	0.96	not significant
Pure Error	19.86	2	9.93			
Cor Total	1585.26	10				

Value of “Prob>F” less than 0.05 indicate model term are significant.

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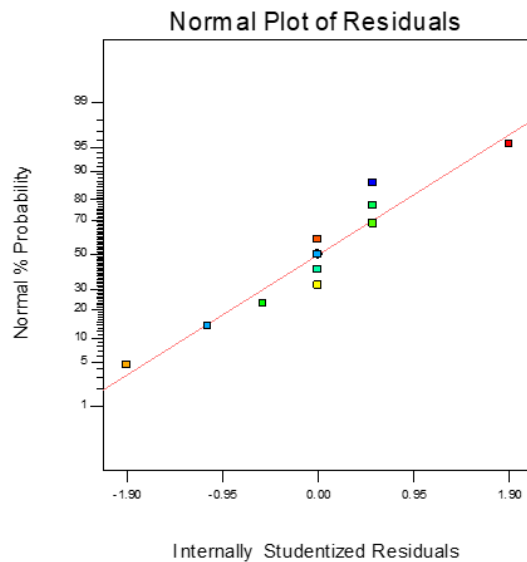


Fig. 1 - Normal plot of residuals

3.3 ANOVA Analysis

The second order model of RSM method was applied to further optimize the filtration condition. Table 4 showed the responses of full factorial CCD which is extension analysis from the first order model. The J_f / J_o ratio was analysed from 20 set data experiment consisting of eight factorial points, six axial points and six center points. Meanwhile, Table 5 presented the ANOVA analysis result that shows the quadratic effect of factors studied and interaction of the response parameters. Table 5 showed that the model “prob > F” was less than 0.05 which indicated that the model is significant. It noticed that the Model F-value of 10.62 implies that the model is significant. There is only a 0.05% chance that a "Model F-value" this large could occur due to error. The ANOVA analysis showed that the “Lack of fit” was not significant and this described a desirably fit model.

Table 4 - Full Factorial design (Second Order Model)

Standard	Run	pH	IO	TMP	Final Normalized Ratio (J_f / J_o)
1	6	3	0.05	2	0.568
2	4	11	0.05	2	0.543
3	2	3	0.5	2	0.531
4	9	11	0.5	2	0.505
5	1	3	0.05	5	0.518
6	7	11	0.05	5	0.480
7	10	3	0.5	5	0.480
8	11	11	0.5	5	0.455
9	8	7	0.28	3.5	0.594
10	3	7	0.28	3.5	0.581
11	5	7	0.28	3.5	0.606
12	12	0.27	0.28	3.5	0.531
13	13	13.73	0.28	3.5	0.455
14	14	7	0	3.5	0.632
15	15	7	0.65	3.5	0.518
16	16	7	0.28	0.98	0.581
17	17	7	0.28	6.02	0.493
18	18	7	0.28	3.5	0.556
19	19	7	0.28	3.5	0.619
20	20	7	0.28	3.5	0.618

Table 5 - ANOVA analysis for RSM Quadratic Model

Source	Sum of square	DF	Mean Square	F-Value	Prob > F	
Model	0.053	9	5.91E-03	10.62	0.0005	significant
A-A (pH)	4.26E-03	1	4.26E-03	7.65	0.0199	
B-B (IO)	7.98E-03	1	7.98E-03	14.35	0.0036	
C-C (TMP)	9.67E-03	1	9.67E-03	17.39	0.0019	
AB	1.99E-05	1	1.99E-05	0.036	0.8536	
AC	1.99E-05	1	1.99E-05	0.036	0.8536	

BC	1.99E-05	1	1.99E-05	0.036	0.8536	
A ²	0.024	1	0.024	43.36	< 0.0001	
B ²	2.04E-03	1	2.04E-03	3.66	0.0849	
C ²	9.21E-03	1	9.21E-03	16.56	0.0023	
Residual	5.56E-03	10	5.56E-04			
Lack of Fit	2.56E-03	5	5.12E-04	0.85	0.568	Not significant
Pure Error	3.01E-03	5	6.01E-04			
Cor Total	0.059	19				
Value of "Prob>F" less than 0.05 indicate model term are significant.						

The main effect of variables x_1 , x_2 , and x_3 shows a significant value (Prob > F less than 0.05) and give a major effect on the J_f / J_o . In addition, the second order effect of x_1^2 , x_2^2 , and x_3^2 were also found to be responsible for the secondary on the response. Based on the ANOVA analysis, the interaction among the parameters shows insignificant, this low or more interaction is commonly found in almost water separation since the effect of the main parameter is highly dominant than their interaction (A-B-C). As explained by Zularisam *et al.*, (2009) and Razali *et al.*, (2013) that this insignificant interaction cannot simply neglect because any individual had an influence on the interaction with each other.

Meanwhile, the R^2 indicates that the model as fitted explains 91% of the variability in the correlation that existed between the experimental and predicted values. The prediction for optimizing the normalized flux in the focus parameters range was obtained via empirical model equation are mathematical correlation. The multiple regression equation of final normalized flux in the coded and actual value as equations (1) and (2).

Coded value:

$$\text{Final Normalized Ratio } (J_f / J_o) = +0.60 - 0.018 * x_1 - 0.02 * x_2 - 0.027 * x_3 + 1.579E-003 * x_1 * x_2 - 1.579E-003 * x_1 * x_3 + 1.579E-003 * x_2 * x_3 - 0.041 * x_1^2 - 0.012 * x_2^2 - 0.025 * x_3^2 \tag{1}$$

Actual value:

$$\text{Final Normalized Ratio } (J_f / J_o) = +0.43977 + 0.031821 * x_1 - 7.00894E-003 * x_2 + 0.061462 * x_3 + 1.75439E-003 * x_1 * x_2 - 2.63158E-004 * x_1 * x_3 + 4.67836E-003 * x_2 * x_3 - 2.55690E-003 * x_1^2 - 0.23470 * x_2^2 - 0.011235 * x_3^2. \tag{2}$$

Figure 2 showed that the analysis of the normal probability with the residual. It was noticed that the residual generally fall on the straight line residual distribution. It demonstrated that the implying errors are distributed normally. Hence, this implies model proposed adequate and reasonable for constant variance assumption.

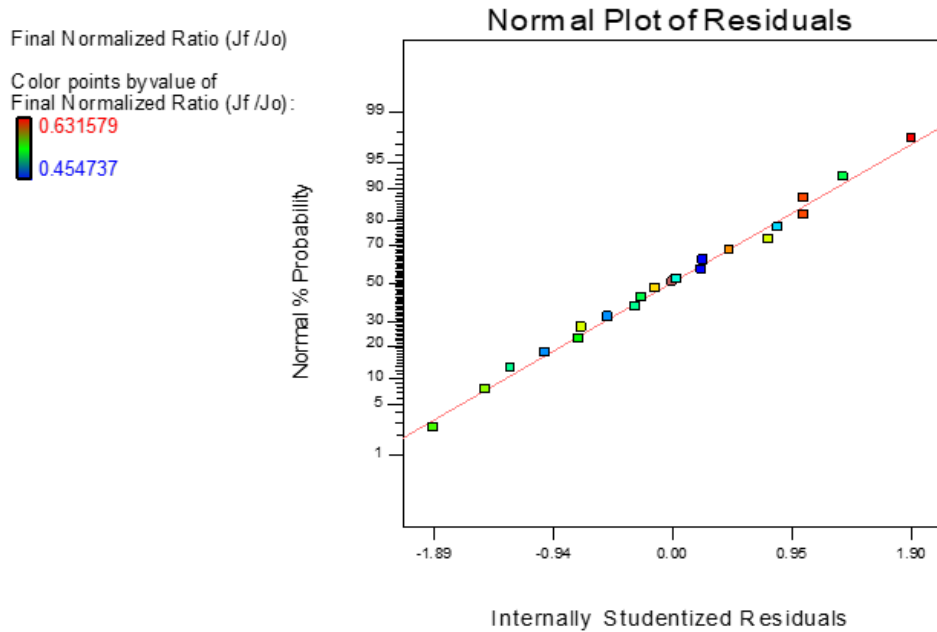


Fig. 2 - The normal plot residuals for second order model

3.4 Effect of pH, Ionic strength (IO) and Transmembrane-pressure (TMP) on normalized flux (J_f/J_o)

The three-dimensional response surface was presented to analyse the effect between independent process responses. Figure 3 shows the three-dimensional response surface and contour plot for the relative effect of normalized flux (J_f/J_o). It was observed that the J_f/J_o was dependent on the interaction of pH, IO and TMP.

The change of J_f/J_o was further analysed as a function of pH and IO. Figure 3 (a) shows the response surface graph of the effect of pH and IO on the J_f/J_o . Figure 3 (a) indicated that the J_f/J_o increased with pH from 3 to 7 and decreased from 8 to 11. Meanwhile, the J_f/J_o was decreased with increasing the IO from 0.05 mol/L until 0.5 mol/L. The result demonstrated that the IO effect on J_f/J_o in a linear way and the maximum of 1 J_f/J_o at high level factor (+1.682). The contour slope was depended on the effect of pH and IO and demonstrated that appears to be gradually steep at higher level condition. The contour indicated that the highest J_f/J_o was 0.59 at pH 7 and 0.28 mol/l of KCl. Figure 3 (a) shows the interaction plot of pH and IO.

Figure 3 (b) shows the effect of TMP and pH on the J_f/J_o . The figure depicts the highest of J_f/J_o in the range of 2.5 bar to 3.5 bars for TMP and 6 to 8 for pH value. In addition, figure showed that the contour plot indicated that the highest J_f/J_o at 0.585 [15]. Both contour effects showed the increment value move toward to maximum region. The result showed that the TMP influence toward final J_f/J_o increased with the increase of IO. This is due to the cake layer formation on the membrane surface affected by pH and TMP in filtration. Figure 3 (c) shows the highest J_f/J_o at 0.62 with the lowest level factor (-1.682) of IO and at the middle (0) of TMP. Meanwhile, the contour plot showed that the maximum of J_f/J_o effect by TMP and IO was 0.60.

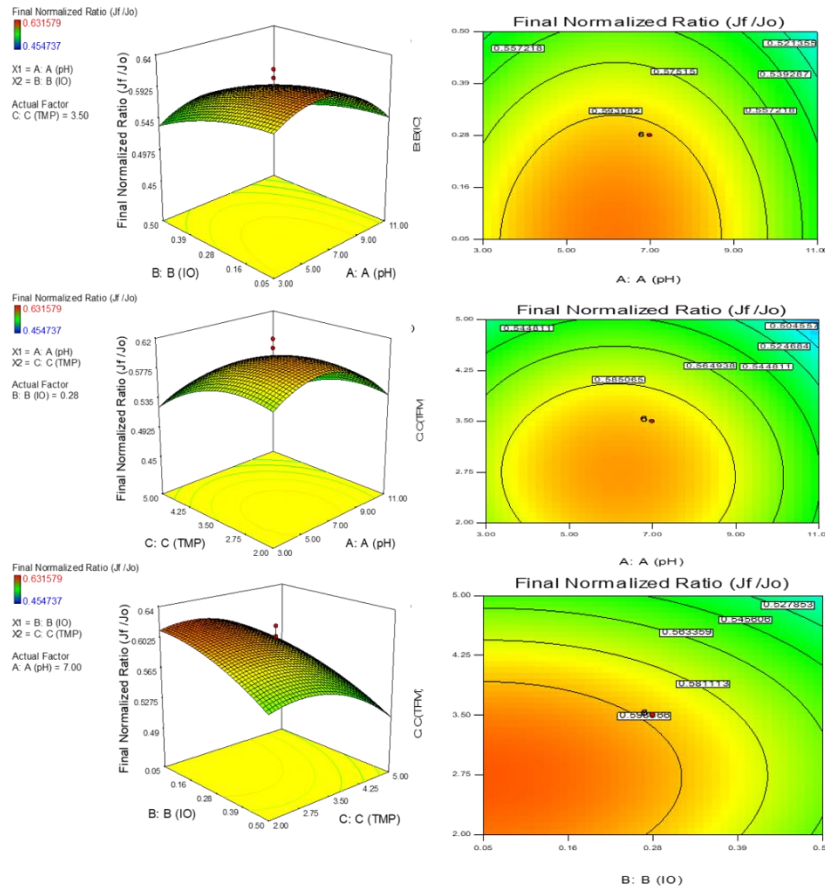


Fig. 3 - The three-dimensional response surface and contour plot for relative effect of final normalized flux (J_f/J_0) at (a) $x_1: x_1 - x_2: x_2$ (b) $x_3: x_3 - A: A$ (d) $x_2: x_2 - x_3: x_3$

3.5 Effect of Transmembrane-pressure

Figure 4 presents the normalized flux at different condition at TMP 1, 6, and 3.5 bars, respectively. The result shows at 6 bars the lowest reduction of normalized flux after 120 min HA filtration at 51% compared to 1 and 3.5 bars at 41% and 44% respectively. This demonstrated that the normalized flux significantly reduced as the TMP increases in filtration.

Commonly, the high pressure will lead to higher penetration of HA particles through the membrane surface and resulting in foulant compaction on the membrane pore. In addition, the high pressure also leads to the increase of cake layer formation due to the lifting boundary layer on the membrane surface and higher bulk concentration. A similar result was observed by Arthanareeswaran et al [16], in which proved that the increased pressure during filtration resulted in increased membrane fouling.

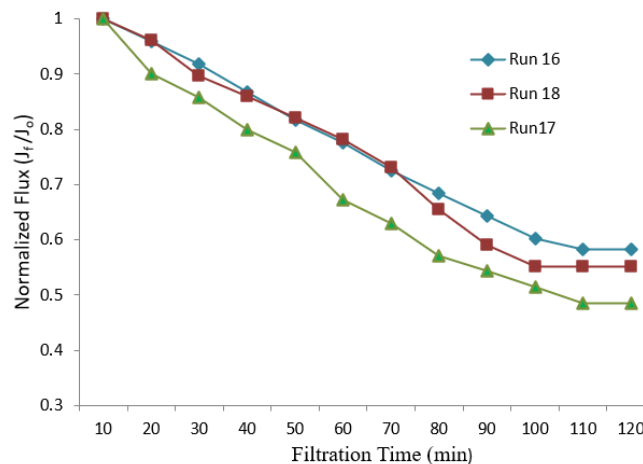


Fig. 4 - Normalized flux at different TMP

3.6 Effect of pH

Figure 5 showed the normalized flux (J_f / J_o) with 120 min filtration obtained at different pH and demonstrated that all plot was reduced with time. As can be clearly seen from the figure, run 19 with pH 7 showed a greater normalized flux ratio compared to the pH 0.27 (Run 12) and 13.37 (Run 13). At pH 7 the flux was reduced only 38% after 120 min HA filtration compared to pH 0.27 and 13.73 at 49% and 55% respectively. This phenomenon could be due to the HA behavior that was affected by pH value.

The result indicates that at low pH, the net charge on HA decreased by a reduction in intramolecular and intermolecular electrostatic repulsion due to the protonation of the carboxylic group ($-COOH$) in HA. Therefore the fouling was increased with cake resistance layer on the membrane surface due to the particles aggregation. The reduction in normalized flux at low pH was caused by a greater difference charge between HA and membrane surface [17]. Hence, both charged attract each other to create the cake resistance layer on the membrane surface as illustrated in Figure 6 (a).

In contrast with the higher pH value that shows the net charges on both HA solution membrane increases causing the repulsion between HA and membrane increased which indirectly reducing fouling. The increase of repulsion energy could be due to the increase of hydrophobic HA caused by the ionization of the $-COOH$ group that brings to the reduction of particles size [18]. The increase of repulsion mechanism among HA particles that avoid agglomeration can create smaller and colloid of HA particles. These smaller particles formed to create pore blocking and acceleration more accumulation of foulant inside the pore as well as at surface especially at higher pressure. As demonstrated in this work, increasing TMP at fixed pH and IO, the normalized flux is reducing indicating the increase fouling. This intense effect of fouling in alkaline compared to acidic is obviously shown in Table 5 where all data for pH with higher significant reduction of normalized flux that showing a major fouling effect. This contradicts effect as shown in Figure 5 revealed that fouling with pore blocking mechanism led to the worst fouling effect as compared to surface foulant that possesses by the acidic medium.

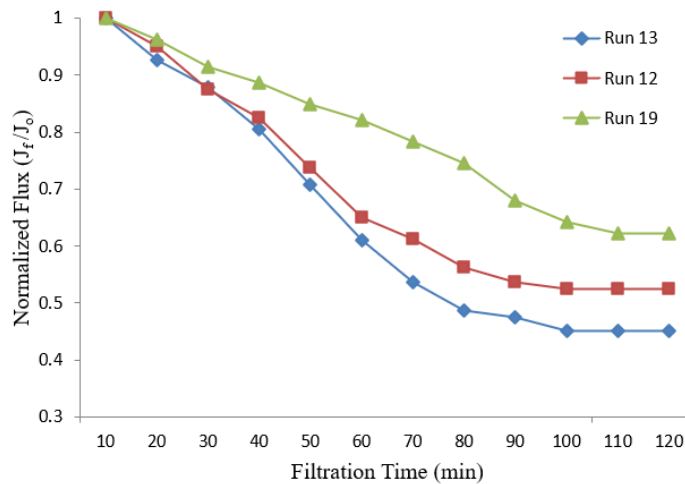


Fig. 5 - The normalized flux at different pH

- a) Low pH- the HA particles attract to membrane surface.
- b) High pH- small hydrophobic particles block the pore.

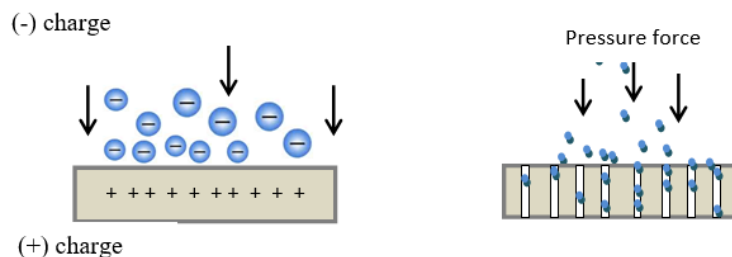


Fig. 6 - a) solution at low pH; b) solution at high pH

3.7 Effect of Ionic Strength (IO)

The normalized flux for the run 14, 15 and 18 is shown in Figure 7. Figure 7 shows plotted for difference of IO condition (Run 14: 0 mol/L, Run 18: 0.28 mol/L and Run 15: 0.65 mol/l). As can be observed from the plot the flux

significantly reduced as the IO increased in the solution. The normalized flux after the 120 min filtration times was reduced from 0.63 to 0.44 for 0.00 mol/L KCl to 0.65 mol/L KCl. This result indicates that with increased of IO the membrane fouling was increased.

This phenomenon can be related to the increased of cake polarization and pore blocking on the membrane surface that can be explained by the decrease of electrostatic repulsion due to the counter of ion (K^+) in the HA solution. Basically, the electrostatic shielding can be explained by the negative charge of the carboxyl functional group of HA molecules that are attracted to the membrane surface. The molecules of HA become small in size and coiled and spherical organic matter that can enter the pores of the membrane via absorbed on the surface due to the difference charged of surface and HA particles. It has been reported that the high IO leads to a partial plugging of the pore affect membrane filtration and increased fouling [19].

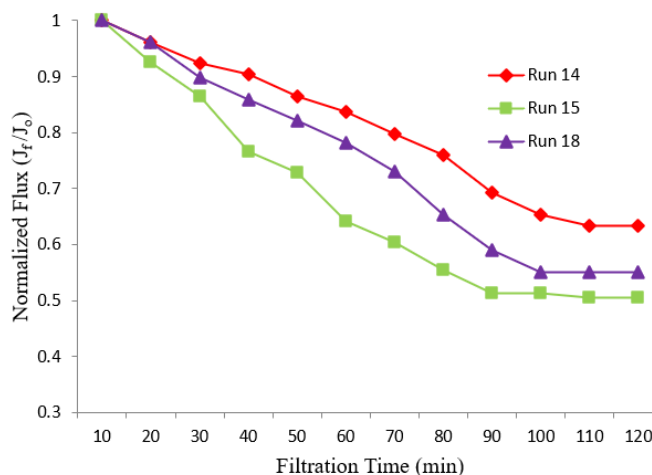


Fig. 7 - Normalized Flux at different IO

4. Conclusion

In this work, flat sheet PSf/PEG/RHS membranes were prepared via phase inversion technique. The effect of RHS loading in membrane formulation for humic acid separation was investigated. The membrane fouling characteristic and the optimal normalized flux of the modified membrane were carried out by humic acid separation. The significant conclusion generated from the conducted experiment are highlighted as follows:

1. The membrane optimization was done by using the Response surface methodology. Results showed that the prediction optimal of normalized flux (J_f / J_0) was 0.632 at 0.92 of the desirability model. In addition, the model adequacy is reasonably at 95% of prediction after experimental verification.
2. It can be concluded that the presence of hydrophilic RHS particles has improved the PWF, rejection and antifouling performance of PSf membrane.

Toward the end of this project, there are some extension needs to be considered to enhance the modification membrane. The following recommendations to extend the study are:

1. Combination of two or more inorganic additives such as silica/silver/eugenol as an antifouling and antibacterial agent to the prepared membrane.
2. Improve the surface roughness by using 2, 4, 6-triaminopyrimidine (TAP) as compatibilizer which then led to an increase in the antifouling performance.
3. Energy binder studies on silica and membrane should be carried out to determine the interaction between particles and polymer.
4. In measuring the permeability value, the pressure should be stabilized before data collecting to get more accurate reading.
5. Make sure all the bubbles were released from the solution by using electrostatic machine before casting to avoid a hole at the membrane surface.

Acknowledgement

The authors would like to thank to financial support by Ministry of Higher Education with Research Grants Scheme vot No 9003-00759 is gratefully acknowledged.

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