

Central Composite Experimental Design Applied to Fabrication of LLDPE Microporous Membrane via Thermally Induced Phase Separation (TIPS) Method

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Abstract—The aim of this study was to apply experimental design in determining the order of magnitude as well as the intersectional interactions between various parameters affecting the microstructure of fabricated polyethylene membranes via thermally induced phase separation method. In this study, among various parameters, the polymer concentration, cooling bath temperature and film thickness were selected as the most effective variables on the structure of membranes. The analysis variance results showed polymer concentration and cooling bath temperature were identically significant. Film thickness effect, however, showed less importance than other parameters. Surface response plots showed that lower polymer concentration and film thickness and higher cooling bath temperature would result in higher porosity. The results of analysis of variance showed that the interactions among individual parameters are significant. However, with an increase in the cooling bath temperature and polymer concentration, the effect of film thickness on the porosity would be negligible.

Key words: Experimental design, central composite design, thermally induced phase separation, microporous membrane, porosity.

I. INTRODUCTION

THE thermally induced phase separation (TIPS) is one of useful methods to produce microporous materials and membranes [1-3]. In TIPS method, homogenous solution is formed by melt blending of a polymer with a high boiling point diluent. Due to the lack of solvent for polyolefins at room temperature, thermally induced phase separation is favorable method which permits to fabricate microporous membranes from polyolefins such as polyethylene and polypropylene. The homogeneous solution is cast or extruded into desired shapes and then cooled down to induce the phase separation. After solidification of polymer by crystallization or glass transition, microporous polymer membrane is formed by extracting the diluent with a volatile solvent [3, 4]. In TIPS process, microporous membrane can be formed via solid-liquid (S-L) or liquid-liquid (L-L) phase separation mechanisms [3, 5]. Polymer-diluent compatibility and polymer concentration are two factors determine the type of phase separation mechanisms. In liquid-liquid phase separation, solution separates into polymer rich and

polymer lean phases, however in solid-liquid phase separation, solution separates into polymer lean phase and pure polymer phase.

Several studies have been conducted to investigate the effect of important variables such as polymer concentration [6], cooling rate [7, 8], diluent properties [2, 8-12], coarsening effect [7] and nucleating agents [13, 14], on membrane structures. Lloyd *et al.* showed that in iPP/C₂₀H₄₂ system, in which solid-liquid phase separation occurs, increasing polymer concentration, increases nucleation density, which in turn decreases the porosity [10]. They also indicated that increasing the cooling rate permits super cooling in which the polymer solution might cool to temperature below its corresponding equilibrium crystallization temperature prior to the actual crystallization of the polymer from solution. Lin *et al.* showed that in PP/DAP system with liquid-liquid phase separation; decreasing polymer concentration increases the interconnectivity of pores [7]. Investigation into the cooling rate indicated that in solid-liquid phase separation, decreasing cooling bath temperature, increases cooling rate and crystallization driving force, which would result in lower porosity [8]. Increasing cooling bath temperature in liquid-liquid phase separation, however, would give more time to polymer lean phase to grow in a specific time interval, which increases the porosity. As mentioned above, the individual effect of influential parameters on the membrane morphology and performance fabricated via TIPS method, have been investigated. However, there are no research on the simultaneous effect of these parameters and their intersectional interaction on the membrane morphology.

Experimental design, a technique commonly used for process analysis, is a potential answer to describe the individual and interaction effects of affecting parameters on the porosity and mechanical strength of membranes which lead us to evaluate the impacts of variables on membrane specifications. This statistical analysis has several important advantageous, such as reducing the number of experiments and possibilities to evaluate interactions among variables [15]. Important experimental design methods are full factorial, partial factorial and central composite design. In our previous work, the intersectional interaction between three parameters such as polymer concentration, bath temperature and membrane thickness was investigated using full factorial experimental

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design [16, 17]. However, full factorial experimental design will never present the optimized fabrication condition, which is a key point in manufacturing industries.

In comparison with full factorial experimental design, the central composite design gives almost as much information as a multilevel factorial, requires much fewer experiments than a full factorial [18]. In addition, the central composite design is mainly used for improving and optimization of the processes and mainly used to evaluate the relative significance of several factors affecting the system even in the presence of complex interactions [19]. Since it is essential that an experimental design methodology be very economical for extracting the maximum amount of complex information, saving significant experimental time as well as material used for analysis, response surface methodology (RSM) is used in this work.

The main objective of the present work is application of RSM method to study the impact of some important parameters on membranes specification. Among several affecting parameters, polymer concentration, cooling bath temperature and film thickness were selected and their impacts as well as their order of magnitudes and the level of importance on membrane porosity were evaluated using RSM technique. RSM was also used to describe the individual and interactional effects of three variables at three levels on membrane porosity.

II. EXPERIMENTAL

A. Materials

Linear low density polyethylene (LLDPE) was provided by Tabriz Petrochemical Company (MFI=0.9 g/10min; 190°C, 2.16 kg). Mineral oil (extra pure) as a diluent was purchased from ACROS, acetone (extra pure) as a solvent and isobutanol were purchased from MERCK. All chemicals used in this work were of the highest purity commercially available and were not further purified.

B. Membrane preparation

The LLDPE and mineral oil were mixed in screw capped test tube in which the portion of polymer to diluents was about 20 wt%. The test tube was sealed to prevent evaporation of diluent during melt blending. The test tube was heated in an oil bath at 180 °C temperature for 2 hours for dissolution of polymer-diluent mixture. The polymer-diluent mixture was dissolved at 180 °C. In order to prevent diluent evaporation and phase separation, homogenous solution was poured onto a preheated sealed glass plate. The cast film thickness was adjusted using an adjustable casting knife. The membrane accompanying with glassy plate was rapidly exposed into cooling water bath with a specific temperature. After peeling off the plates, membranes were immersed into acetone to extract diluents out and dried at room temperature.

C. Phase diagram

In order to measure the cloud point of binary polymer-

diluent, the obtained homogeneous sample was cut into small pieces and placed between a pair of microscope slides, while a 100 µm thick PTFE film with a square opening in the center was inserted between the slides. The sample was heated on a hot stage (LEICA DMRX) up to 180 °C for 3min and then cooled to 25 °C with a controlled rate of 1 °C/min. During the cooling step, the temperature in which clear binary solution was changed to a turbid mixture was reported as cloud point. DSC (Shimadzu DSC-60) was used to determine the dynamic crystallization temperature, T_c . The solid polymer-diluent samples were sealed in an aluminum differential scanning calorimetry pan, melted at 180°C for 3min, then cooled with 1 °C/min to 25°C. The onset of the exothermic peak during the cooling was taken as the dynamic crystallization temperature

D. Porosity measurements

The resulted membranes were cut into small pieces. The porosity is determined by impregnating membranes with isobutanol for 24 hours. The impregnated samples were blotted with tissue papers to remove the residual isobutanol on the surface of membranes. The mass of the samples before and after immersing in isobutanol was measured using a digital microbalance. The porosity of the membrane (ϵ) is defined as the pore volume divided by the total volume of the membrane as follows:

$$\epsilon = \frac{(w_2 - w_1)\rho_1}{\rho_1 w_2 + (\rho_2 - \rho_1)w_1} \times 100 \quad (1)$$

where w_1 is the initial membrane weight, w_2 is the immersed membrane weight, and ρ_1 and ρ_2 are the density of LLDPE (0.92 g/cm³) and i-butanol (0.802 g/cm³), respectively.

E. Water permeability

The membranes were initially pressurized with distilled water at 2 kPa for 30min. The effective membrane area was 3.14 cm². Experiments were carried out in a dead-end water permeation set up at room temperature. After compaction at 2.5 kPa, the pure water permeability of the membranes was measured at 2 kPa. The water permeability was calculated using the following equation:

$$J_w = \frac{Q}{A \cdot \Delta P} \quad (2)$$

where, J_w is the permeability of membranes (l/m².bar.hr), Q is the quantity of permeate (l/h). A is the effective membrane surface area (m²) and ΔP is the trans membrane pressure. The pressure was averaged from the readings of the inlet pressure gauge and outlet one. The permeability of each type of flat sheet membrane was the averaged value of two batches, and each batch was measured for 3 times.

F. Scanning Electron Microscopy (SEM)

Dried membranes were fractured in liquid nitrogen and crosses sections of membranes were examined using a scanning electron microscope (SEM, LEO440-I) with an accelerating voltage of 15 kV.

G. Experimental design

The central point for each factor was assumed 0 and the design is symmetric around this. For three variables, the central composite design consists of 20 experiments, including 8 factor points, 6 axial points and 6 center points with six replications. The central composite design was constructed in Table I.

TABLE I
CENTRAL COMPOSITE EXPERIMENT DESIGN

Experiment number	Blocks	X ₁	X ₂	X ₃	Porosity (%)
9	1	-1	0	0	70
12	1	0	1	0	65
20	1	0	0	0	52
17	1	0	0	0	53
3	1	-1	1	-1	78
18	1	0	0	0	53
13	1	0	0	-1	57
5	1	-1	-1	1	50
11	1	0	-1	0	43
4	1	1	1	-1	62
6	1	1	-1	1	35
2	1	1	-1	-1	40
8	1	1	1	1	57
19	1	0	0	0	52
16	1	0	0	0	52
7	1	-1	1	1	72
15	1	0	0	0	53
14	1	0	0	1	50
1	1	-1	-1	-1	69
10	1	1	0	0	48

Factors in this experiment were polymer concentration (X₁), cooling bath temperature (X₂) and film thickness (X₃). The mathematical relationship of the response Y on the three significant independent variables X₁, X₂ and X₃ can be approximated by a nonlinear polynomial model. Eq. (3) including three squared terms, three factors expressing interaction terms, three linear terms and one intercept term as shown below:

$$Y = b_0 + b_1 X_1 + b_2 X_2 + b_3 X_3 + b_{11} X_1^2 + b_{22} X_2^2 + b_{33} X_3^2 + b_{12} X_1 X_2 + b_{13} X_1 X_3 + b_{23} X_2 X_3 \quad (3)$$

Y is membrane porosity, b₀ is the average of the results of the replicated center points; b₁, b₂ and b₃ are the main half-effects of the coded variables X₁, X₂ and X₃, respectively; b₁₁, b₂₂ and b₃₃ are the squared effects; b₁₂, b₁₃ and b₂₃ are double factor interaction half-effects. Design and analysis of the central composite experiments were carried out with MINITAB14. The low, middle and high levels of each variable were designated as -1, 0 and +1, respectively. The corresponding actual values for each variable are listed in Table II.

TABLE II
EXPERIMENTAL FACTORS AND LEVELS INVESTIGATED IN MEMBRANE POROSITY

Factor	Parameter values		
	-1	0	1
X ₁ : Polymer concentration (wt %)	20	25	30
X ₂ : Cooling bath temperature(°C)	0	30	60
X ₃ : Film thickness(μm)	250	375	500

III. RESULTS AND DISCUSSION

A. Phase diagram

The phase diagram of polyethylene-mineral oil is shown in Figure 1. Since polyethylene and mineral oil solubility parameters are close to each other, the compatibility of polymer and diluents directly affects on phase diagram [20]. This binary system is quite compatible and it caused only solid-liquid phase separation to be occurred. In this system, only crystallization curve was observed. It seems that binodal curve was shifted below of crystallization curve.

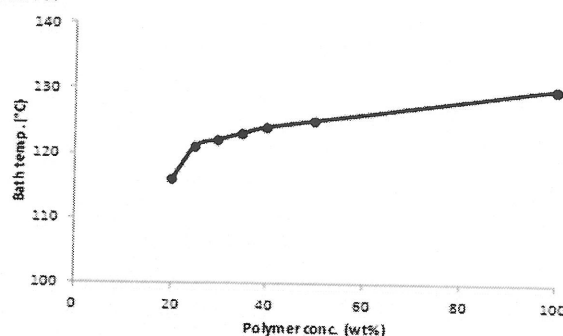


Fig. 1. Phase diagram of linear low density polyethylene (LLDPE) and mineral oil.

B. Analysis of variance (ANOVA) and estimated regression coefficient for response Y

Experiments were carried out by considering a central composite design. As shown in Table III, effects of linear terms, squared terms and two factor interactions for Y₁ was determined. The coefficients of the nonlinear polynomial model; Eq. (3) and p values were shown in Table IV. With a 95% confidence, the polynomial model was postulated as shown in Eq. (4).

$$Y = 52.9091 - 9.7X_1 + 9.7X_2 - 4.2X_3 + 5.2273X_1^2 + 1.625X_1 X_2 + 0.023X_1 X_3 + 0.042X_2 X_3 \quad (4)$$

The P-value in the analysis of variance (ANOVA) for all parameters and their interactions are less than 0.05 to the computed F-values obtained for membrane porosity.

TABLE III
ANALYSIS OF VARIANCE FOR Y (MEMBRANE POROSITY)

Source	DF ^a	AdjSS ^b	AdjMS ^c	F ^d	P ^e
Block	9	2264.03	251.559	64.55	0.000
Linear	3	2058.20	686.067	176.05	0.000
Square	3	135.45	45.152	11.59	0.001
Interaction	3	70.37	23.458	6.02	0.013
Residual error	10	38.97	3.897	-----	-----
Pure error	5	1.33	0.267	-----	-----
Total	19	-----	-----	-----	-----

^a Degree of freedom.

^b Adjusted sum of squares.

^c Adjusted mean of squares.

^d F test.

^e p value.

Computed F-values which are greater than the F-value in the statistical table, indicating that response surface model is significant at a 95% confidence. P-values of linear, square and interaction terms shown in Table IV are less than 0.05.

High F-value of linear term means that, porosity curves versus independent variables would be linear.

TABLE IV

ESTIMATED REGRESSION COEFFICIENTS FOR Y (MEMBRANE POROSITY)

Term	Coef. ^a	SE Coef. ^b	T ^c	P ^d
Constant	52.9091	0.6786	77.963	0.000
X ₁	-9.7000	0.6243	-15.538	0.000
X ₂	9.7000	0.6243	15.538	0.000
X ₃	-4.2000	0.6243	-6.728	0.000
X ₁ *X ₁	5.2273	1.1904	4.391	0.001
X ₂ *X ₂	0.2273	1.1904	0.191	0.852
X ₃ *X ₃	-0.2727	1.1904	-0.229	0.823
X ₁ *X ₂	1.6250	0.6979	2.328	0.042
X ₁ *X ₃	1.8750	0.6979	2.686	0.023
X ₂ *X ₃	1.6250	0.6979	2.328	0.042

^a Coefficient.^b Standard error of the coefficient.^c t test.^d p value.

C. Effect of variables on membrane porosity by response surface plots

Response surfaces can be plotted as three-dimensional plots that display the response as a function of two factors while keeping a third factor at center level. Effect of each variable was shown as the three-dimensional (3D) and contour (2D) plots for the membrane.

Figure 2 shows the impact of the concentration of polymer and the bath temperature in an constant thickness (375 μm) on the porosity of membrane. As the results indicate, decreasing the polymer concentration and increasing the bath temperature increase the porosity of the membrane. In the two-dimensional diagrams, the maximum porosity (marked in dark in Figure 2) is achieved in low polymer concentrations and high bath temperatures.

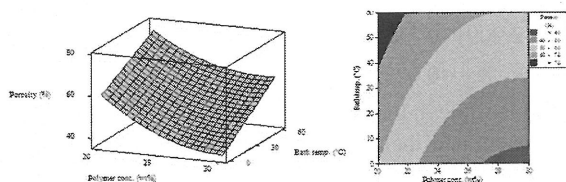


Fig. 2. The three-dimensional (3D) and contour (2D) plots of LLDPE membrane porosity (Y) versus polymer concentration (X₁) and water bath temperature (X₂).

Figure 3 shows the variation in the membrane porosity versus the polymer concentration and thickness of membrane films. In this figure, the bath temperature is maintained at 30°C. It is concluded that higher thickness of polymer film would result in lower membrane porosity.

The articles have made no mention of the impact of the membrane thickness on its porosity. However, one can assume that the thicker the membrane, the longer the way would be for the diluent to move towards the bath to flow out of the porous membrane. Since the diluent's thrust depends on mass transfer and is intrinsically slow process in comparison with other transport phenomena, longer periods of time are needed. In higher thicknesses, before the diluent flows out of the system, phase separation occurs mainly due to heat transfer. The point is that due to the higher driving force for heat transfer in thermally

induced phase separation method, heat transfer is faster than mass transfer and the diluent is trapped between polymer crystals in high thicknesses. When the diluent molecules are entrapped, crystals would not grow sufficiently and subsequently they grow individually and in smaller proportions. The result is that the membrane porosity declines.

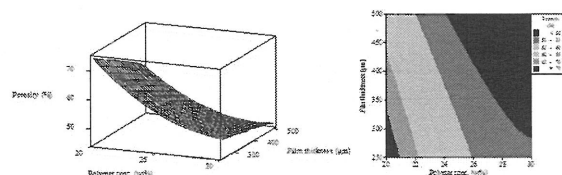


Fig. 3. The three-dimensional (3D) and contour (2D) plots of LLDPE membrane porosity (Y) versus polymer concentration (X₁) and film thickness (X₂).

The important point in Figure 3 is that the gradient in the slope of porosity-thickness diagram is far lower than the slope of porosity-concentration diagram. It means that the polymer's concentration is more influential than its thickness on the porosity.

Figure 4 shows the variation in the membrane porosity versus the bath temperature and thickness of membrane films. As shown in this figure, increasing the bath temperature and decreasing the membrane thickness increases the membrane porosity. These results are agreed with our proposed speculation, described in Figures 2 and 3.

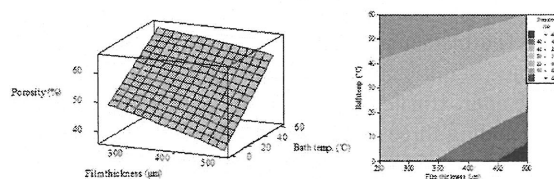


Fig. 4. The three-dimensional (3D) and contour (2D) plots of LLDPE membrane porosity (Y) versus film thickness (X₁) and water bath temperature (X₂).

D. Intersection effect of variables on membrane porosity

Another important point regarding the impact of effective parameters on the membrane porosity pertains to the interactions between the factors affecting the porosity changes in the membrane's structure. As Table III indicates, the p-values are less than 0.005 for mutual effects, proving the interaction of parameters examined. Therefore, it is necessary to precisely assess these mutual effects.

1) Interaction of polymer concentration and membrane thickness

Figure 5 shows the permeation of pure water (PWP) for three polymer concentrations in two thicknesses 250 and 500 μm in which the cooling bath temperature is kept at 30°C. As the results show, the PWP is higher in 250 μm samples than that of 500 μm ones. However, the difference in the rate of PWP reaches its minimum in 30 wt% of polymer concentration. The proximity of PWP in the membranes with different thicknesses is indicative of the significance of mutual impact of concentration and

thickness on the porosity. The reason is that the interaction between the polymer concentration and the thickness of membrane film is of high significance in high polymer concentrations and therefore the impact of the membrane's thickness on the porosity is negligible. In high polymer concentrations, the density of crystals is so high that the diluent could not flow out in both higher and lower thicknesses. Figure 3 confirms this point as the porosity-concentration slope is lower than the porosity-thickness slope.

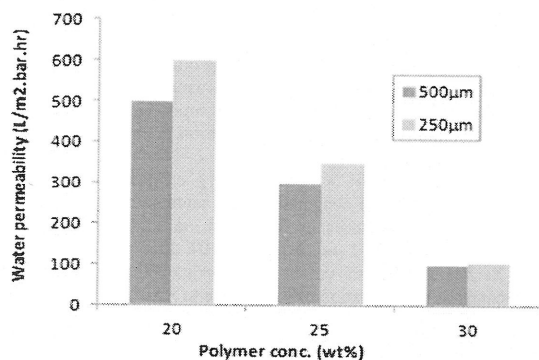


Fig. 5. Evaluation of pure water permeation test for LLDPE membranes with two film thicknesses fabricated in three different polymer/diluent ratios in which the water bath temperature was fixed at 30°C.

2) Interaction of cooling bath temperature and membrane thickness

Figures 6 and 7 show the SEM images of cross section of microporous membranes with 20 wt% of polymer concentration prepared in different cooling bath temperatures for two membrane film thicknesses. Comparing Figures 6 and 7 which differ only in cooling bath temperature, one can conclude that higher bath temperature reduces to nothing the impact of thickness on porosity. The reason is that in higher cooling bath temperatures, the diluent moves much more rapidly and higher thickness is no important obstacle to the outflow of diluent through crystals. Therefore, the two membranes have almost identical porosities in high cooling bath temperature.

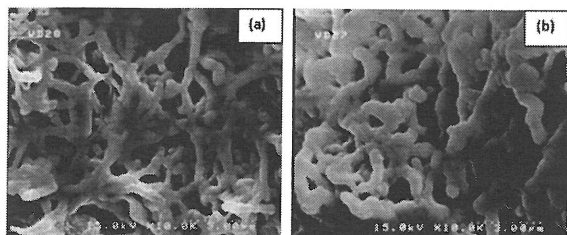


Fig. 6. Cross section SEM images of LLDPE membranes fabricated with a polymer concentration of 20 wt%, cooling bath temperature of 0 °C in different film thicknesses, (a) 250 μm and (b) 500 μm.

This point is confirmed when the pure water permeation through membrane is studied. Figure 8 shows 20 wt% polymer membrane's PWP in different cooling bath temperatures. Identical flows are seen in high temperatures although thicknesses differ.

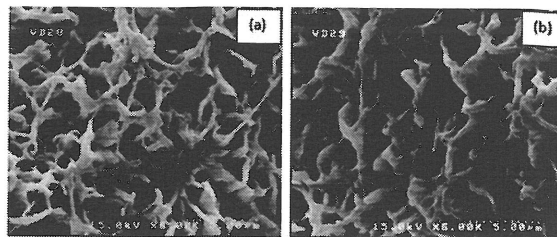


Fig. 7. Cross-sections SEM images of LLDPE membranes fabricated with a polymer concentration of 20 wt%, water bath temperature of 60 °C in different film thicknesses, (a) 250 μm and (b) 500 μm.

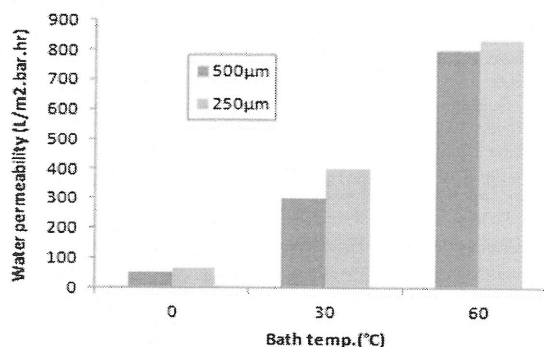


Fig. 8. Evaluation of pure water permeation test for LLDPE membranes with two film thicknesses cooled in water bath with adjusted temperature fixed at 0°C to 30°C and 60°C, respectively, in which the polymer concentration was adjusted at 25% wt of polymer/diluent.

IV. CONCLUSION

Polymer concentration, cooling bath temperature and film thicknesses were selected as main affecting parameters on membrane porosity. The central composite design was used to evaluate the impact as well as the order of magnitude of above mentioned parameters on membrane porosity. It was shown that central composite design can be nominated as an efficient and economical technique to obtain the maximum amount of information with the fewest number of experiments. The analysis of variance revealed that the impact of membrane film thickness can be hindered by the cooling bath temperature as well as polymer concentration. The obtained results were confirmed by pure water permeation tests and SEM images.

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