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Application of the Experimental Design for the Optimization of Microfiltration Membrane

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ABSTRACT

In the present study, the effect of zirconia powder content, polyvinyl alcohol (PVA), Contact time and sintering temperature of zirconia microfiltration membrane elaboration were evaluated using Plackett-Burman design (PBD) and investigated by thickness and pore diameter. The zirconia microfiltration membrane has been prepared by the powder suspension technique. A deflocculated suspension of zirconia was obtained by mixing (5-10% w/w) of zirconia, (30-35% w/w) of PVA (12% w/w aqueous solution) as binder and water. The zirconia layer was deposited on the inner surface of clay support by slip casting with a contact time (2-5 min). After drying at room temperature, the ZrO2 membrane was sintered (800-1000°C) for 2 hours. By using PBD, sintering temperature, PVA and zirconia were recognized and selected as important effectives parameters of pore diameter; On the other hand, PVA and contact time were the main controlling parameters of the thickness of zirconia microfiltration layer. The optimal factors to elaborate the microfiltration membrane by using experimental design include a sintering temperature of 1000°C, zirconia content of 5%, PVA content of 30% and contact time of 2 min predicting a pores diameter of 0,24 µm and thickness of 24 µm.

Keywords: Membrane, Zirconia Microfiltration, Optimization, Experimental Design

1.0 INTRODUCTION

Ceramics membranes have been used in a number of industries due of several performances advantages over its organic counterpart such as thermal and chemical stability, bacteria resistance, possibility of regeneration and better mechanical strength under high pressure [1]. Porous ceramics membrane generally has a composite structure, they consist of a porous support, intermediate layer and the top active membrane layer [2].

The choice of Materials is very important for the membrane preparation as it defines various properties like pore size, porosity, thickness and strength [2-3]. The chemical and physical characteristics of these materials are responsible for these membrane properties. Ceramics membranes are prepared from different material, including Spinel, Silica, Titania, Alumina, Zirconia. Ceramics membranes are generally prepared by using the following steps: (i) powder preparation; (ii) shaping; (iii) Temperature treatment of deposited layer [3].

The slip casting method is the most commonly used for the ceramic microfiltration membrane. In this method, a powder suspension is poured in to a porous mold. The solvent of the well-mixed suspension thus diffuses

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through the pores of the mold forming a gel layer over the porous surface of mold by precipitations of the particles. The capillary suction process of the porous substrate helps in concentrating the suspension particles at the substrate-suspension boundary. The membranes are then dried and sintered. The membrane prepared by this method are highly permeate, but the thickness of the layer is difficult to control [4-5]. The sintering is usually the final step in ceramic membrane production. It’s consisted of three important steps, the initial stage, the intermediate stage and the final stage. The membrane precursor and the particles have different features and movement at each stage, including full densification, grain coarsening and closing of pores. The temperature regulation depends upon the membrane precursor and especially the material type [6].

However, many trials are required for evaluating the effect of these factors on the ceramic microfiltration membrane elaboration. The experimental design is generally used for the reduction of the number of experiments and the determination of a response value. The Plackett Burman is a useful screening tool which helps to identify the significant parameters for further optimization. It provides unbiased estimation for all parameters with high accuracy and increase the efficiency of the process as each variable is screened in the presence of all other variable [7-11].

The objective of this study was to estimate the best elaboration conditions of Zirconia microfiltration membrane based on slip casting route and evaluate the effect of zirconia powder, PVA, contact time and sintering temperature by using Plackett Burman Design as a screening plan.

2.0 MATERIALS AND METHODS

2.1 Preparation of Macroporous Support Clay

The elaboration of ceramic support has been reported in our previous work. The grain size of Moroccan Sahara clay used to prepare the paste is 30 µm. The sample was performed by extrusion of the mixture of clay and organic additives. The extruded pieces were dried at ambient temperature for 24 hours and transferred in an oven at 50°C for 24 hours. The Optimized response to elaborate the ceramic support of porosity 38,79% and Mechanical strength of 12 MPa include a starch of 4% and sintering temperature of 1014,36°C [11-12].

2.2 Preparation of ZrO_2 Microfiltration Membrane

The powder suspension technique was used to prepare the zirconia microfiltration membrane layer. A deflocculated suspension of zirconia was obtained by mixing zirconia powder, PVA (12% w/w aqueous solution) as binder and water (with Dispersant 0,2% w/w) like dispersing agent. The zirconia membrane layer was deposited on the inner surface of Moroccan clay support by slip casting (Figure 1). After drying at room temperature, the ZrO_2 membrane was sintered for 2 hours [1, 13].

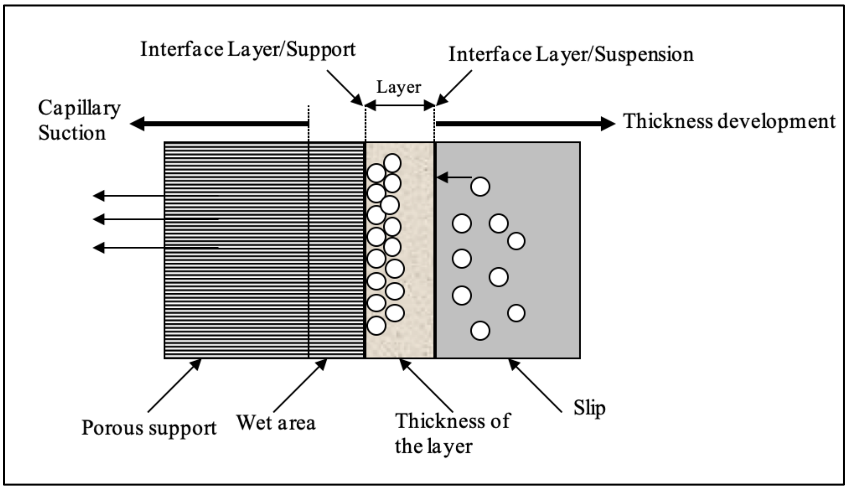


Figure 1 Mechanism of layer formation by slip casting technique

2.3 Methods of Characterization

Different techniques were used to investigate the properties of zirconia microfiltration ceramic membrane.The porosity was determined by a mercury intrusion porosimetry method (Micrometrics, Model Autopore 9220) for sintered specimens support at different temperature.The Thickness and morphology of Zirconia membrane was examined by scanning electron microscopy (Hitachi, S-4500).

2.4 Plackett-Burman Design (PBD)

The most well-known non-regular designs are the PB designs introduced by Plackett and Burman (1946) [7]. They have become known for their ability to investigate a large number of factors in a relatively low number of experimental runs. Due to this property the PB designs are often used for screening. The number of runs n in a PB design is equal to a multiple of four. Plackett and Burman only included the designs with n ≤ 100, and they also omitted the design where n = 92. For PB designs where the number of runs is equal to a power of two the designs coincide with the regular ones, and the rest of the PB designs are non-regular.

The elaboration of microfiltration ceramic membrane depends on some factors. There are chemicals mineralogical, Organic additives, processing operation (drying and sintering) and so forth. Only one factor is varied by time and the others are fixed when any factor is optimized; subsequently, the best value obtained by this procedure is fixed and other factors will be varied by the time; thus, using the unvaried procedure to optimize all variables is time consuming.

The interaction among all factors are neglected in invariable procedure, so the best conditions could be achieved [14]. The PBD is an effective method used for screening the significant factors from a large number of factors affecting the process. The following first-order polynomial equation (Equation (1) was used to perform mathematical modeling) [15].

$$Y = b_0 + \sum_{i=1}^{K} b_i X_i \quad (1)$$

The following codified equation was used to explain the Plackett-Burman designs with 4 factors.

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_4 \quad (2)$$

The results of the experiments design were analyzed using statistical software to evaluate the effect as well as the statistical parameters, the statistical plots (Pareto, normal probability of the standardized effects and main effects). PBD having 12 experiments for zirconia microfiltration ceramic membrane elaboration was studied and a matrix was established according to their high and low levels, represented by +1 and -1 respectively. The coded value of variables with the response (Pore diameter and Thickness) were illustrated in Table 1. The main effect of the microfiltration ceramic membrane elaboration was identified based on the P value with 95% of confidence level.

Table 1 Factor levels tested using Plackett-Burman Design

Parameters	Experimental value	
	Low(-1)	High(+1)
Zirconia powder (%)	5	10
PVA (%)	30	35
Contact time (min)	2	5
Sintering temperature (ST) (°C)	800	1000

3.0 RESULTS AND DISCUSSION

3.1 Effects of the Processing Factors on the Physical Properties of ZrO_2 Microfiltration Membrane Using Plackett-Burman

The measured value of the technological properties (Thickness and Pore Diameter) of the sintered test specimens as a function of zirconia powder content, Polyvinyl Alcohol (PVA), Contact time and sintering temperature are given in Table 2.

A regression equation was obtained for each zirconia membrane using factorial design at a 5% level of significance. Analysis of ANOVA and plots of observed values versus predicted one were used to confirm the validity and precision of model.

Table 2 Experiment design matrix and measured values of the considered responses using Plackett-Burman (Y1: Pore Diameter (µm); Y2: Thickness(µm))

Run	Coded Values				Experimental Values				Experimental Response		Predicted Response	
	A	B	C	D	Zirconia Content (%) (A)	PVA Content % (B)	Contact Time (min) (C)	Sintering Temperature (°C) (D)	Y_1 (µm) Pore Diameter	Y_2 (µm) Thickness	Y_1 (µm) Pore Diameter	Y_2 (µm) Thickness
1	1	-1	-1	-1	10	30	2	800	0,19	24	0,191	24,167
2	1	-1	1	1	10	30	5	1000	0,25	25	0,248	25,833
3	-1	-1	1	1	5	30	5	1000	0,24	26	0,243	25,500
4	1	1	-1	1	10	35	2	1000	0,27	26	0,271	25,833
5	1	-1	1	-1	10	30	5	800	0,19	26	0,193	25,833
6	-1	-1	-1	-1	5	30	2	800	0,19	24	0,186	23,833
7	-1	-1	-1	1	5	30	2	1000	0,24	24	0,241	23,833

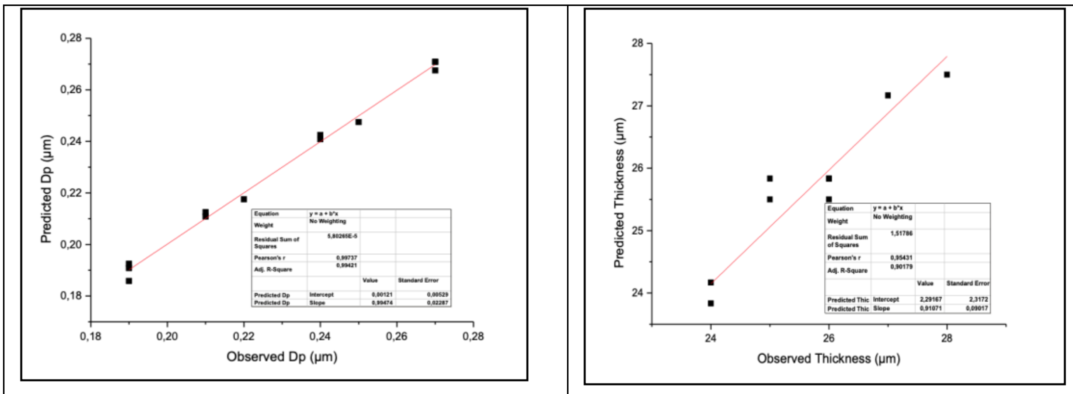
Run	Coded Values				Experimental Values				Experimental Response		Predicted Response	
	A	B	C	D	Zirconia Content (%) (A)	PVA Content % (B)	Contact Time (min) (C)	Sintering Temperature (°C) (D)	Y_1 (µm) Pore Diameter	Y_2 (µm) Thickness	Y_1 (µm) Pore Diameter	Y_2 (µm) Thickness
8	-1	1	1	-1	5	35	5	800	0,21	27	0,213	27,167
9	-1	1	1	1	5	35	5	1000	0,27	27	0,268	27,167
10	1	1	1	-1	10	35	5	800	0,22	28	0,218	27,500
11	-1	1	-1	-1	5	35	2	800	0,21	25	0,211	25,500
12	1	1	-1	1	10	35	2	1000	0,27	26	0,271	25,833

The results equations of Pore diameter (µm) (Y1) and thickness (µm) (Y2) are reported after:

$Y_1=0.229+0,0025A+0,0125B+0,027D$ (3)

$Y_2=25.667+0,167A+0.833B+0,833C$ (4)

Based on the experimental data, regression models were fitted for Y1 and Y2, as shown in Equations (3) and (4), respectively. The adequacy of the initial model was tested via parity plot for observed versus predicted values, as demonstrated in Figure 2. As seen in Figure 2, the high values of the correlation coefficient (R2 =0,99) and (R2 =0,901) for Pore diameter and thickness respectively demonstrates good correlation between the observed and the predicted responses by initial models.



Equation	y = a + b*x		
Weight	No Weighting		
Residual Sum of Squares	5,80265E-5		
Pearson's r	0,99737		
Adj. R-Square	0,99421		
		Value	Standard Error
Predicted Dp	Intercept	0,00121	0,00529
Predicted Dp	Slope	0,99474	0,02287

(a)

Equation	y = a + b*x		
Weight	No Weighting		
Residual Sum of Squares	1,51786		
Pearson's r	0,95431		
Adj. R-Square	0,90179		
		Value	Standard Error
Predicted Thic	Intercept	2,29167	2,3172
Predicted Thic	Slope	0,91071	0,09017

(b)

Figure 2 Parity Plot of predicted versus observed responses for (a) Pore Diameter (Dp) and (b) Thickness

Analysis of variance (ANOVA) was employed to investigate the adequacy and significance of the model. The effect of a factor is defined as the change in response produced by a change in the level of the factor. This is frequently called the main effect because it refers to the primary factors of interest in the experiments. The ANOVA results showed that the equation adequately represented the actual relationship between each response and the significant variables. The F-value implies that the models are significant and value of Prob>F less than 0,05 indicate that the models terms are significant. Especially larger F-value with the associated P value (smaller than 0,05, confidence intervals) means that the experimental system can be modelled effectively with less error [16].

According to the ANOVA results (Table 3 and 4), the values of F_{cal} (331,00 and 17,85 for Pore diameter (Y_1) and thickness (Y_2) respectively were higher and P values were lower than 0,05 which shows the significance and suitability of PBD model. Moreover, the normal probability of the residuals almost indicated no departures from the normality (Figure 4).

As shown in the Table 5, High coefficient of determination (R^2: 0,994 and 0,910 for Y_1 and Y_2 respectively) and adjusted coefficient of determination (R^2adj: 0,991 and 0,859 for Y_1 and Y_2 respectively) indicate the good agreement of experimental response values with model predicted values. The predicted R-squared (R^2pred: 0,984 and 0,737 for Y_1 and Y_2 respectively) was also in reasonable agreement with adjusted R-squared and showed a good prediction of model.

Table 3 Analysis of Variance (ANOVA) for response surface for the prediction of Pore Diameter)

Source	**Sum of Square (SS)**	**Df**	**Mean Squares (MSS)**	**F Value**	**P-Value Probability (P)>F**
Model	0,011033	4	0,002758	331,00	0,000
Linearity	0,011033	2	0,002758	331,00	0,000
Zirconia content -A	0,000075	1	0,000075	9,00	0,020
PVA-B	0,001875	1	0,001875	225,00	0,000
Contact Time-C	0,00008	1	0,00008	1,00	0,351
Sintering Temperature (°C) - D	0,009075	1	0,009075	1089,00	0,000
Error	0,000058	7	0,000058		
Lack-of-Fit	0,000058	6	0,000010		
Pure Error	0,000000	1	0,00000		
Total	0,011092	11			

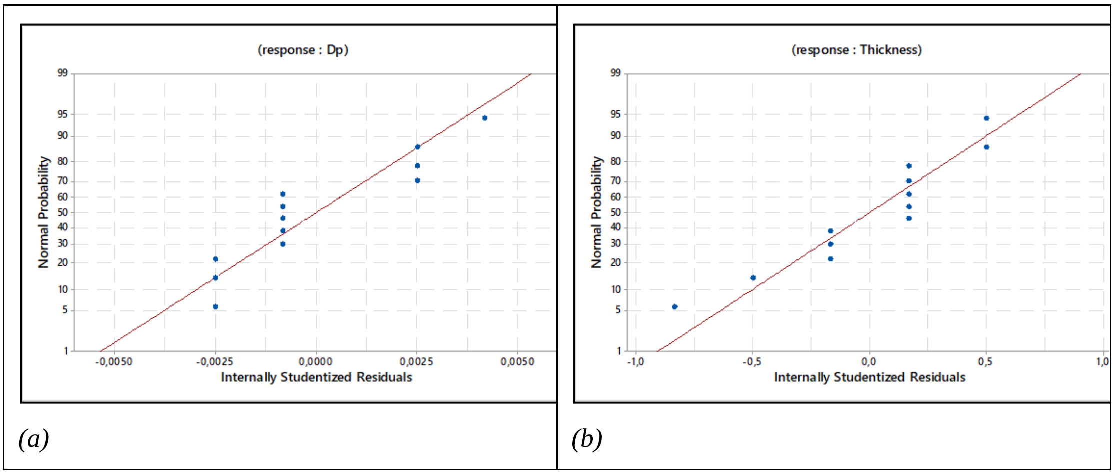
Table 4 Analysis of Variance (ANOVA) for response surface for the prediction of Thickness

Source	**Sum of Square (SS)**	**Df**	**Mean Squares (MSS)**	**F Value**	**P-Value Probability (P)>F**
Model	17,0000	4	4,25000	17,85	0,001
Linearity	17,0000	4	4,25000	17,85	0,001
Zirconia content -A	0,3333	1	0,3333	1,40	0,275

Source	**Sum of Square (SS)**	**Df**	**Mean Squares (MSS)**	**F Value**	**P-Value** **Probability (P)>F**
PVA - B	8,3333	1	8,3333	35,00	0,001
Contact Time - C	8,3333	1	8,3333	35,00	0,001
Sintering Temperature (°C) - D	0,0000	1	0,0000	0,00	1,000
Error	1,6667	7	0,23810		
Lack-of-Fit	1,6667	6	0,27778		
Pure Error	0,000000	1	0,00000		
Total	18,6667	11			

Table 5 Values of correlation coefficient (R2) related to the adopted models

Response	**R^2 Coefficient of Determination**	**R^2 Adjusted**	**R^2 Predicted**
Y_1: Pore Diameter (μm)	0,994	0,991	0,984
Y_2: Thickness (μm)	0,910	0,859	0,737



(a) *(b)*

Figure 2 Normal Probability of the Residuals of (a) Pore Diameter and (b) Thickness

Factors that influence the pore diameter ((μm) and thickness (μm) were evaluated by using factorials plots: main effect, Pareto and normal probability plots [11, 17].

Taking into consideration the value of linear coefficient shown in the above equations, the weight effect of the considered parameters followed the order: D>B>C for the pore diameter and B=C for the thickness.

The main effect which are helpful in visualizing which factors most affects the response of each parameters represent deviations of the average between high and low levels of each one of them as shown in Figure 6.

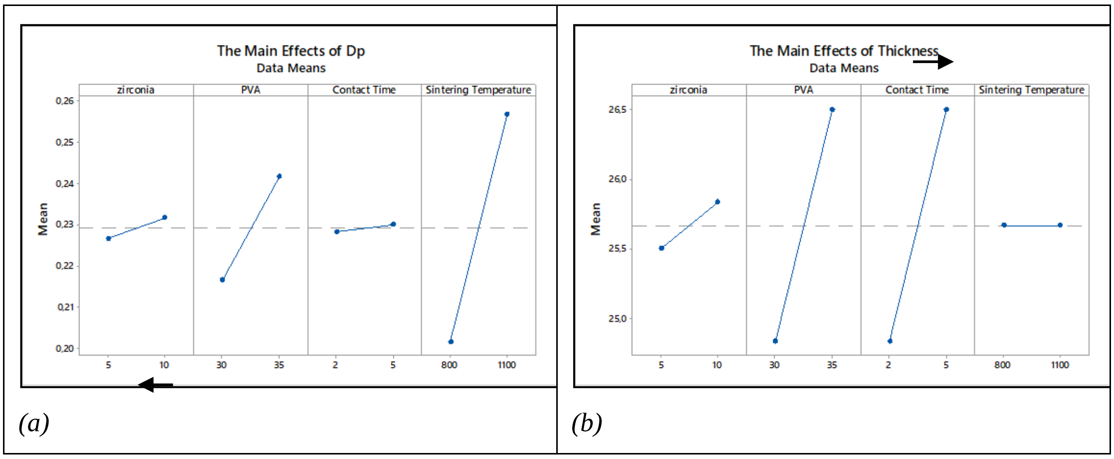
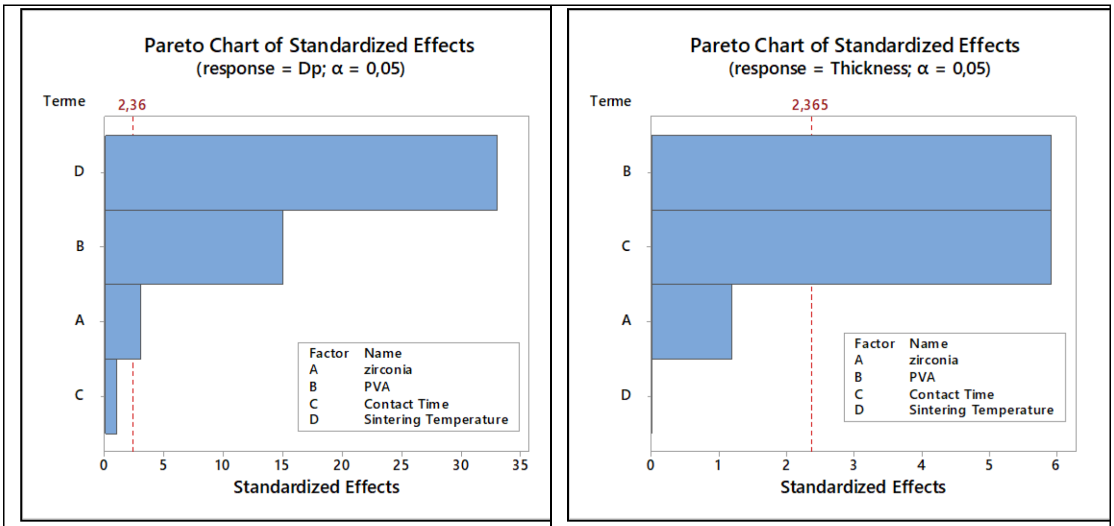


Figure 6 Main Effects for (a) Pore Diameter and (b) Thickness

Each level of factor effects the response differently; if the slop is close zero, then the magnitude of the main effects will be small. As The results show, the sintering temperature appears to have a great effect on the response on Pore diameter as indicated by steeply slope due the great surface followed by PVA and zirconia. On the other hand, the main effect of thickness is PVA and contact time, the sintering temperature don’t have any influence.

The increase of the sintering temperature causes an enlargement of the pore diameter but at high value of sintering the pores tend to decrease [18]. Such effects are related to the strong influence of temperature on the melt formation and consequently on the sintering process [19-20]. However, the PVA affects the properties of the membrane support, the increase of PVA content provokes an increase of pore diameter. This effect is due of pore forming during burning out around 500°C of PVA [21-22]. The PVA is used as binder in casting process to provide sufficient strength to the body so that the green bodies can be modeled and retained in the desired shape without breaking or damage, before and during sintering process. Also, they cause to achieve a higher thickness in function of time contact with desired support [1, 23].

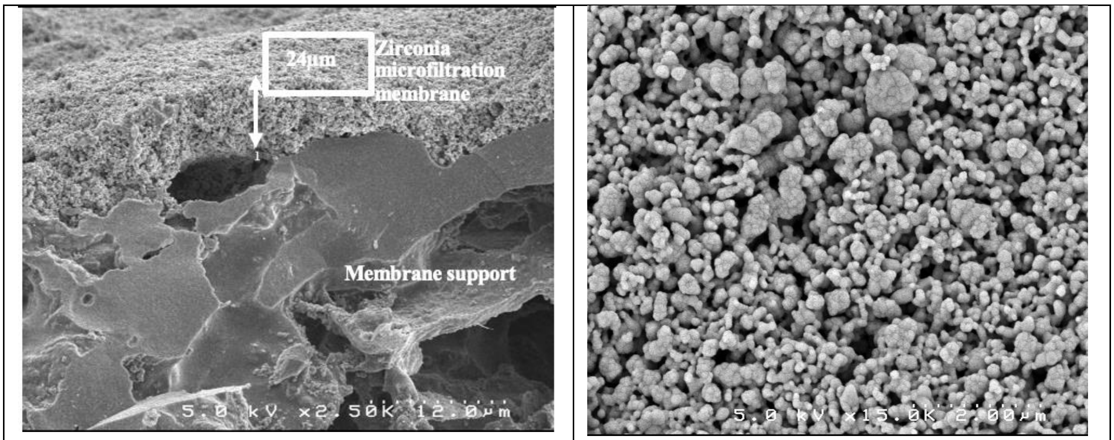
The relative importance of the main effects was also observed on the Pareto Chart as shown in Figure 8. The value that exceed the reference line are considered significant values and those which do not are considered insignificant [8, 11]. According to this figure, for Pores Diameter the sintering temperature, PVA and Zirconia are the main effect, otherwise for the thickness, the PVA and contact time are significant.



(a) (b)

Figure 8 Pareto Chart of Individual Factors effects of (a) Pores Diameter and (b) Thickness

Response optimization for the current model was performed with the target set as high Diameter of pores and less thickness (less hydrodynamic resistance). The optimized predicting response (pores diameter of 0,24 µm and thickness of 24 µm.) to elaborate the zirconia microfiltration membrane include a sintering temperature of 1000°C, zirconia content of 5%, PVA content of 30% and contact time of 2 min.



(a) (b)

4.0 CONCLUSION

The application of Plackett Burman design is an effective tool to evaluate the important significant factors influencing the elaboration of zirconia microfiltration ceramic membrane. 3 factors namely the Sintering temperature, PVA and zirconia content were found to exert a significant effect on the process of pores diameter. The two factors namely PVA and contact time were considered to be potentially influential on the thickness. A detailed analysis of the results was conducted using a multilinear regression method based on the analysis of variance using Fisher test and the validation by the coefficient of determination R2 were found to be statically significant and presented low variability.

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