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Modeling and Optimization on the Carbon Dioxide Separation from Natural Gas Using Hydrotalcite-Silica Membrane

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Abstract

The process modeling and optimization of carbon dioxide (CO_2) separation from carbon dioxide-methane (CH_4) binary gas mixture through hydrotalcite (HT)-silica membrane using statistical design of experiments (DoE) is reported in this study. The effect of three important process variables, pressure difference across the membrane (100-500 kPa), temperature (30-190^oC) and CO_2 feed concentration (10-50%) on the CO_2 separation performance of the membrane were investigated. The response surface methodology (RSM) coupled with central composite design (CCD) was used to build up two models to correlate the effect of process conditions to CO2 permeance and CO_2/CH_4 separation selectivity. The analysis of variance (ANOVA) of the quadratic model at 95% confidence interval confirmed that the model was highly significant. The CO_2 feed concentration with 43% showed the best performance with a CO_2 permeance of $6.0x10^{-7}$ $mol.m^{-2}.s^{-1}.Pa^{-1}$ and a CO_2/CH_4 separation selectivity of 109 at 100 kPa pressure difference across the membrane and temperature of 30^oC.

Keywords: Hydrotalcite; porous membrane; carbon dioxide capture; response surface methodology**.**

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1.Introduction

Separation of CO_2 from natural gas is crucial and it has received increasing attention from researchers in energy part of the world [1, 2 & 3]. Separation of CO_2 increases the calorific capacity, yields better transportation conditions and prevents pipeline corrosions [4]. Microporous inorganic membranes have been widely investigated for gas separation in the past few years in comparison with polymeric membranes due to their unique thermal, chemical and mechanical stability [5, 6 & 7].
Hydrotalcite (HT) is class of anionic clays called layered double hydroxide or hydrotalcite-like compounds. HT has been intensively investigated in recent years as good adsorbents for CO_2 [8, 9 & 10]. The HT-silica composite membrane could have the desired features of silica and HT components. Composite membrane could exhibit high CO_2 adsorption capacity due to the HT, while large surface area and small pore size due to the silica component in the membrane. The high CO_2 adsorption capacity, large surface area and small pore size would improve the separation of CO_2 from natural gas [11, 12].
In the previous research studies, a novel microporous composite membrane investigated from HT material modified porous silica membrane to investigate the CO_2 separation from diverse gas mixtures, has been fabricated and characterized successfully [11, 12 & 13]. The modified internal pore surface of silica membrane with HT material, enhanced the adsorbed CO_2 amount that resulted in the increment of the CO_2 permeance, diffusion, and separation, respectively [14, 15]. However, very few researches reported the modeling and optimization of the process variables for gas permeation and selectivity through membrane. Here, the discussion only pertains the factorial design which was conducted by fixing all the process variables with only one variable varied at a time. The drawbacks of this technique are time consuming and difficulty to find the interaction between the process variables. Accordingly, in this paper statistical approach is applied to find the optimum operating conditions for the permeation and separation of CO_2 from CO_2/CH_4 binary gas mixture. Design of Experiment (DoE) is used as statistical tool to determine the optimum conditions, to evaluate the interaction between the variables and to build up an equation that can be represent the CO_2 permeance and CO_2/CH_4 separation selectivity and effects of surface affinity to permeability and selectivity.

2. Modeling and Optimization

2.1 Design of Experiments (DoE)

The design of experiments (DoE) was used for modeling and optimizing the permeation and separation experiments of CO_2/CH_4 binary gas mixture [11, 12 & 13]. Design Expert software version 6.0.6 was used in DoE. The effects of three independent variables (pressure difference across the membrane, temperature and CO_2 feed concentration) on CO_2 permeance and separation selectivity of CO_2/CH_4 were studied in the ranges of 100-500 kPa pressure difference, 30-190 ºC temperature and 10-50% CO_2 feed concentration. In this statistical model, 20 experimental runs were suggested by the response surface methodology (RSM) coupled with central composite design (CCD) available in the Design Expert. The main advantage of RSM is to reduce the required experimental runs required to model the permeation and separation performance of mixed gases. The Design-Expert software enables determination of the functional relationships between independent variables from minimum number of experiments. It also provides empirical model for the desired response as a function of selected variables by applying the multiple regression analysis method on the experimental data [16]. The analysis of variance (ANOVA) was implemented on the empirical model to find its statistical significance. After that, the responses were optimized by numerical optimization approach available in the Design Expert software.

The experimental design matrix of 2^3 full factorial with CCD for the permeation and separation of CO_2/CH_4 binary mixture using HT-silica membrane is shown in Table 1. Three factors full factorial design requires a total 20 experimental runs where it consists of 8 factorial points, 6 axial points and 6 replicates at center points. The replicate at center point (experimental runs of 15-20) was used to check the reproducibility of experimental data. The experimental runs were conducted randomly so as to minimize bias from the systematic trends in the variables. Two responses of CO_2 permeance and CO_2/CH_4 separation selectivity were considered to study the effect of process variables. The empirical model is shown below:

$$Y = \beta_0 + \beta_1 A + \beta_2 B + \beta_3 C + \beta_{12} AB + \beta_{13} AC + \beta_{23} BC + \beta_{11} A^2 + \beta_{22} B^2 + \beta_{33} C^2 \quad (1)$$

Where: Y is the predicted response. A, B and C are the actual forms of pressure difference, temperature and CO_2 feed concentration, respectively. The term β_0 is the offset, β_1 , β_2 and β_3 are the linear terms, β_{11} , β_{22} and β_{33} are the quadratic terms, and β_{12} , β_{13} and β_{23} are the interaction terms. The analysis of variance (ANOVA) was implemented on the empirical model to find its statistical significance. After that, the responses were optimized by numerical optimization approach available in the Design Expert software.

Table 1 Experiment matrix and responses for the permeation and separation of CO_2/CH_4 across micro-porous HT-silica membrane.

Run	Point Type	Variables			Responses (Y)	
		Pressure difference, kPa (A)	Temperature, °C (B)	CO_2 Feed Conc., % (C)	CO_2 permeance $\times 10^7$, mol/m².s.Pa (Y_1)	CO_2/CH_4 Selectivity (Y_2)
1	Fact	180	62	42	3.71	74.25
2	Fact	180	158	42	1.86	32.63
3	Fact	420	62	18	2.08	23.54
4	Fact	420	158	42	0.76	16.97
5	Fact	180	62	18	3.75	48.44
6	Fact	420	158	18	1.67	10.52
7	Fact	180	158	18	2.77	19.61
8	Fact	420	62	42	1.37	39.22
9	Axial	300	110	10	2.78	14.36
10	Axial	300	190	30	1.33	19.12
11	Axial	300	30	30	3.82	52.56
12	Axial	500	110	30	1.31	23.26
13	Axial	300	110	50	0.87	23.81
14	Axial	100	110	30	4.23	55.55
Repeated runs for reproducibility						
15	Center	300	110	30	1.85	29.76
16	Center	300	110	30	2.19	31.96
17	Center	300	110	30	2.16	30.54
18	Center	300	110	30	1.82	25.18
19	Center	300	110	30	1.78	27.72
20	Center	300	110	30	1.75	26.56

2.2 Gas permeation and separation selectivity

The measured permeance of gas species *i* in the binary gas mixture, K_i was obtained by taking the ratio of mole flux of the gas species *i*, N_i, in the permeate to the log-mean pressure difference, $\Delta P_{ln,i}$, across the membrane,

$$K_i = N_i/\Delta P_{ln,i} \quad (2)$$

Where,

$$\Delta P_{ln,i} = \frac{(p_{F,i}-p_{P,i})-(p_{R,i}-p_{P,i})}{\ln[(p_{F,i}-p_{P,i})/(p_{R,i}-p_{P,i})]} \quad (3)$$

$p_{P,i}$ and $p_{R,i}$ are the partial pressures for gas species *i* in feed, permeate and retentate, respectively. The separation selectivity of binary mixture, α_{ij}, between 2 gases can be computed based on the ratio of the perm$p_{F,i}$eance of gas species-*i* to that of species-*j* in the binary mixture containing of species *i*,*j*.

$$\alpha_{ij} = K_i/K_j \quad (4)$$

3. Results & Discussion

3.1 Response surface modeling of CO_2 permeance

Table 2 presents the statistical results of the analysis using ANOVA for the CO_2 permeance in the CO_2/CH_4 binary mixture. The ANOVA results show that the CO_2 permeance is best described with a polynomial model. This quadratic model is proposed by RSM software in terms of actual factors, as shown in Equation 5. The positive sign (+) in front of model terms designates synergetic effect, increase the CO_2 permeance, whereas the negative sign (-) designates antagonistic effect, decrease the CO_2 permeance.

$$\begin{aligned} \boldsymbol{K}_{CO_2} &= +8.6698 - 0.0206\,\boldsymbol{A} - 0.0361\,\boldsymbol{B} \\ &+ 3.8567\,\boldsymbol{C} + 1.8559 \times 10^{-5}\boldsymbol{A}^2 + 8.5526 \\ &\times 10^{-5}\,\boldsymbol{B}^2 - 5.0659\,\boldsymbol{C}^2 + 3.9996 \\ &\times 10^{-5}\,\boldsymbol{AB} - 5.9220 \times 10^{-3}\boldsymbol{AC} \\ &- 0.0236\,\boldsymbol{BC} \end{aligned} \quad (5)$$

ANOVA is applied for estimation of the main variable effects and their potential interaction on the CO_2 permeance. The most important outputs from ANOVA results are the Fisher value (F-value) and associated probability value (Prob>F). The (Prob>F) value indicates the probability equals the proportion of the area under the curve of the F-distribution that lies beyond the observed F-value. In other words, when the (Prob>F) value is small, the particular term is considered to significantly affect the CO_2 permeance. The model terms with (Prob>F) less than 0.05 indicate that the terms are significant for the model of CO_2 permeance. The model F-value is 25.72 and Prob>F value is less than 0.05 implying that the developed model is significant at 95% confident level. In present study, terms A, B, C, A^2, B^2 and AB are significant for CO_2 permeance at 95% confident level. Although the terms C^2, AC and BC are not significant to the model because of their values of probability (Prob>F) are greater than 0.05, they are involved in Equation 5 to get a hierarchy model [16]. The lack of fit is the ratio between the residuals and pure error. Lack of fit F-value of 3.11 with (Prob>F) of 0.1194 denotes that it is not significant comparative to the pure error due to noise, consequently the suggested model for CO_2 permeance in Equation 5 is valid for the present study.

Based on the ANOVA result in Table 2, the F-values show the pressure difference across the membrane gives the greater impact on CO_2 permeance followed by temperature and CO_2 feed concentration. The interaction between pressure difference and temperature on CO_2 permeance at central level of CO_2 feed concentration (30%) is shown in the three-dimensional response surface plot in Figure 1. As it can be observed from the figure that the CO_2 permeance decreases with the increase of pressure difference from 100-500 kPa for separation temperature ranging from 30 to 190 ^oC. This result is consistent with those reported in literature for the membranes that cosidered surface diffussion mechanism for CO_2 separation [17]. The CO_2 permeance declined because the coverage-gradient driving force increase is less than the CO_2 partial pressure difference. As the pressure difference across the membrane is increased, the adsorbed concentration gradient of CO_2 increases, and therefore, CO_2 flux increases. The increase in CO_2 flux with increase in the adsorbed concentration gradient is lower than the increase in CO_2 partial pressure difference [18, 19]. Then, the CO_2 permeance (CO_2 flux/ The log-mean partial pressure difference) declines as the pressure difference is increased from 100 to 500 kPa. At low temperature (30 ^oC) the developed model predicts that the CO_2 permeance declined from 6.22×10^{-7} to 2.20×10^{-7} $mol.m^{-2}.s^{-1}.Pa^{-1}$ as the pressure difference is increased from 100-500 kPa, while at high temperature from 2.96×10^{-7} to 1.50×10^{-7} $mol.m^{-2}.s^{-1}.Pa^{-1}$. At high temperature the CO_2 adsorption coverage on HT is low. Thus, the CO_2 permeance decreases with increase in the pressure difference at high temperature less than at low temperature. The response surface plot of CO_2 permeance at 110 ^oC with a different CO_2 feed concentration and pressure differance is shown in Figure 2. Generally the trend of CO_2 permeance is rather similar to that of Figure 3. The increase in the separation temperature leads to reduction of the surface coverage and at the same time increases the CO_2 micropore diffusivity. Hence, the CO_2 permeance decreases with increase in separation temperature, which means the surface coverage promotes the micropore diffusivity [19]. The CO_2 permeance decreaces with increase in CO_2 feed concentration from 10 to 50% because the coverage-gradient driving force increase is less than the CO_2 partial pressure difference [20].

Table 2 ANOVA results for CO_2 permeance in the CO_2/CH_4 binary mixture.

Source	Sum of squares	DF*	Mean square	F-value	Prob > F	
Model	18.43	9	2.05	25.72	<0.0001	Significant
A	9.06	1	9.06	113.75	<0.0001	
B	4.73	1	4.73	59.42	<0.0001	
C	2.45	1	2.45	30.75	0.0002	
A^2	0.99	1	0.99	12.47	0.0054	
B^2	0.54	1	0.54	6.78	0.0263	
C^2	0.074	1	0.074	0.93	0.3578	
AB	0.41	1	0.41	5.14	0.0467	
AC	0.056	1	0.056	0.70	0.4208	
BC	0.14	1	0.14	1.80	0.2097	
Residual	0.80	10	0.08	-	-	
Lack of Fit	0.60	5	0.12	3.11	0.1194	Not significant
Pure Error	0.19	5	0.039	-	-	
Cor Total	19.23	19	-	-	-	
Dev**	0.28					
R^2	0.9586					

DF* = Degree of Freedom, Dev** = Standard Deviation

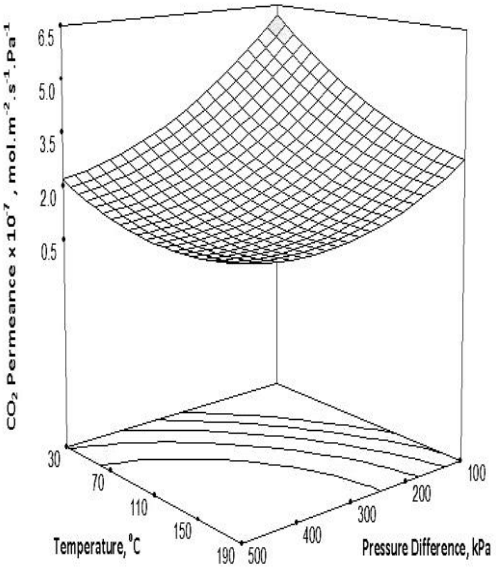

Figure 1: Effect of pressure difference across the HT-silica membrane and temperature on CO_2 permeance at 30% CO_2 feed concentration

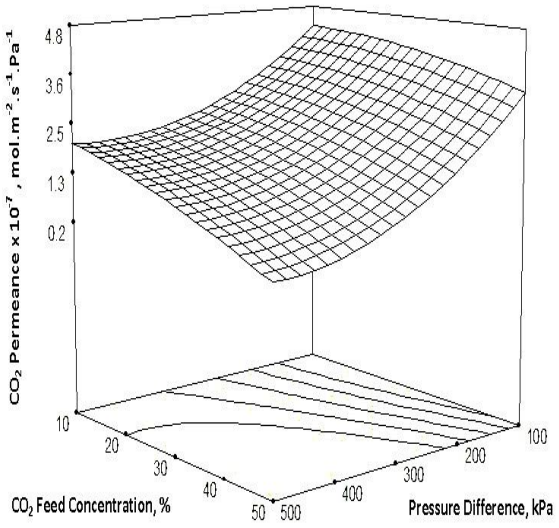

Figure 2: Effect of pressure difference across the HT-silica membrane and CO_2 feed concentration on CO_2 permeance at temperature of 110 ºC

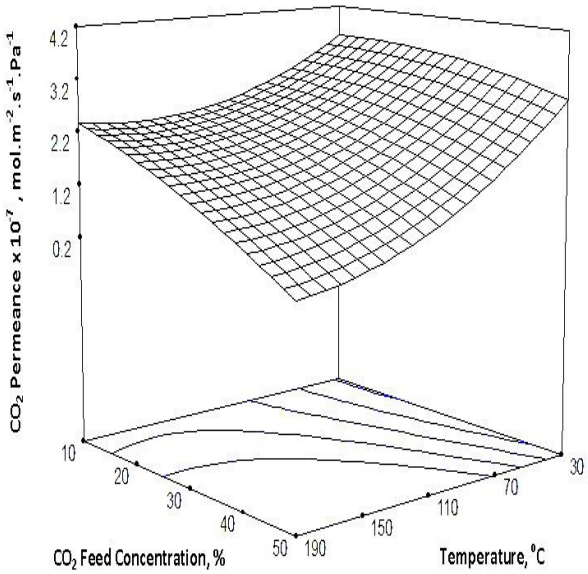

Figure 3: Effect of temperature and CO_2 feed concentration on CO_2 permeance at pressure difference across the HT-silica membrane of 300 kPa

3.2 Response surface modeling of CO_2/CH_4 separation selectivity

The analysis of variance ANOVA for CO_2/CH_4 separation selectivity is presented in Table 3. The ANOVA results show that the CO_2/CH_4 separation selectivity is best presented with a quadratic model. This model is suggested by RSM software in terms of actual factors, as shown in Equation 6.

$$\alpha_{CO_2/CH_4} = +86.0929 - 0.3023\,\boldsymbol{A} - 0.6162\,\boldsymbol{B} + 274.4279\,\boldsymbol{C} + 2.9250 \times 10^{-4}\boldsymbol{A}^2 + 1.2711 \times 10^{-3}\boldsymbol{B}^2 - 215.5016\,\boldsymbol{C}^2 + 7.7738 \times 10^{-4}\boldsymbol{AB} - 0.1476\,\boldsymbol{AC} - 0.4866\,\boldsymbol{BC} \quad (6)$$

As shown in ANOVA Table 3, F-value of the developed model gives a value of 32.11 with Prob>F value of <0.05 implying that the model of CO_2/CH_4 separation selectivity is significant at 95% confidence level. In this study, A, B, C, A^2, B^2, C^2 and AB are significant terms for CO_2/CH_4 separation selectivity. Whereas AC and BC are not significant to the CO_2/CH_4 separation selectivity because of their values of probability (Prob>F) are higher than 0.05. However, they are included in Equation 6 to get a hierarchy model [16]. Lack of fit F-value of 3.77 with (Prob>F) of 0.0858 implies that it is not significant relative to the pure error due to noise. As a result, the proposed model for CO_2/CH_4 separation selectivity in Equation 6 is valid for the present study.

According to the F-values of the ANOVA result in Table 3, the temperature shows the highest effect on CO_2/CH_4 separation selectivity followed by the pressure difference across the HT-silica membrane and CO_2 feed concentration. The quadratic effect of pressure difference (A^2), temperature (B^2), CO_2 feed concentration (C^2) and the interaction pressure differance and temperature (AB) are relatively significant, with the F-value of 15.56, 7.57, 8.5 and 9.82, respectively. The dimensional response surface plots for the CO_2/CH_4 separation selectivity at center level with its interaction between pressure difference, temperature and CO_2 feed concentration are shown in Figure 4, Figure 5 and Figure 6. It can be seen from Figure 6 that the CO_2/CH_4 separation selectivity decreased as the pressure difference was increased from 100-500 kPa. The CO_2 flow mechanism is surface diffution and micropore diffusion due to the high CO_2 adsorption capacity (47.48 mg CO_2/g sorbent) and small pore diameter (8Å) of the HT-silica membrane [12], while CH_4 flow mechanism is micropore diffusion. The weakly adsorbed molecule CH_4 was hindered from penetrating through the micro-porous HT-silica membrane due to mouth narrowing by adsorbed CO_2 molecules [19-21]. The CO_2 permeance decreases proportionally more than CH_4 permeance, and thus the CO_2/CH_4 selectivity is decreased. The movement of the gases molecules through membrane pores was affected by the intermolecular collusion, the interaction between gas molecules and membrane pore wall, and the interplay between the movements of different gas molecules and its relation with the kinetic diameter of the gas molecule [17, 19]. The effect of CO_2 feed concentration on CO_2/CH_4 separation selectivity at different pressure difference and temperature are shown in Fig 5 and Figure 6. It can be observed that the CO_2/CH_4 separation selectivity increases with increase in CO_2 feed concentration due to the increase of CO_2 loading and more CO_2 molecules available in the gas mixture leads to increase the blocking effect and decrease the CH_4 permeance. However, at CO_2 feed concentration near saturation, increasing the CO_2 feed concentration caused a small increase in CO_2 loading along the membrane wall, thus the CH_4 permeance remained constant while the CO_2 permeance continued to decrease with increase in CO_2 feed concentration [17, 20].

Table 3 ANOVA results for separation selectivity in the CO_2/CH_4 binary mixture.

Source	Sum of Squares	DF*	Mean Square	F-Value	Prob > F	
Model	4551.71	9	505.75	32.11	<0.0001	Significant
A	1414.44	1	1414.44	89.80	<0.0001	
B	1920.70	1	1920.70	121.93	<0.0001	
C	432.48	1	432.48	27.46	0.0004	
A^2	246.59	1	246.59	15.65	0.0027	
B^2	119.21	1	119.21	7.57	0.0204	
C^2	133.85	1	133.85	8.50	0.0154	
AB	154.70	1	154.70	9.82	0.0106	
AC	34.86	1	34.86	2.21	0.1677	
BC	60.61	1	60.61	3.85	0.0782	
Residual	157.52	10	15.75	-	-	
Lack of Fit	124.49	5	24.90	3.77	0.0858	Not significant
Pure Error	33.03	5	6.61	-	-	
Cor Total	4709.23	19	-	-	-	
Dev**	3.97					
R^2	0.9666					

DF* = Degree of Freedom, Dev** = Standard Deviation

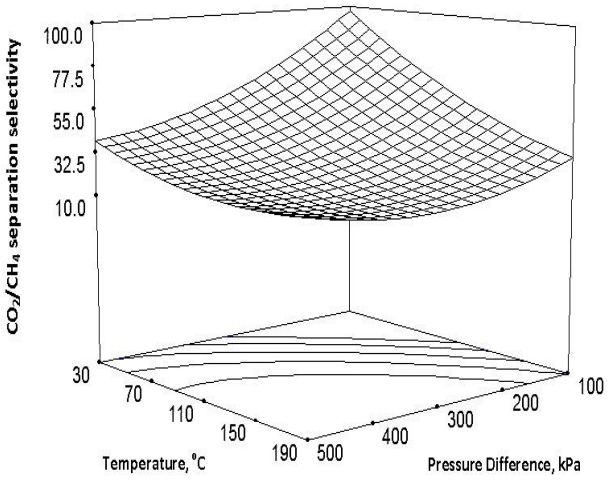

Figure 4: Effect of pressure difference across the HT-silica membrane and temperature on CO_2/CH_4 separation selectivity at 30% CO_2 feed concentration

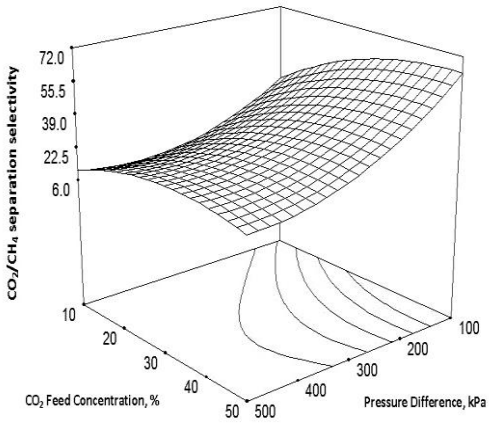

Figure 5: Effect of pressure difference across the HT silica membrane and CO_2 feed concentration on CO_2/CH_4 separation selectivity at temperature of 110 ºC

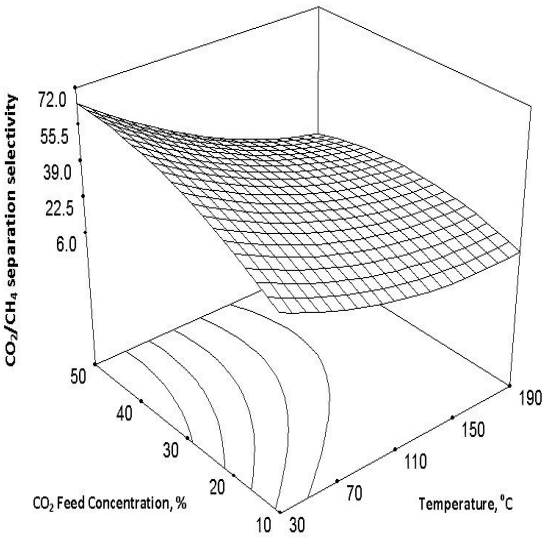

Figure 6: Effect of temperature and CO_2 feed concentration on CO_2/CH_4 separation selectivity at pressure difference across the HT-silica membrane of 300 kPa

3.3 Process optimization using response surface methodology (RSM)

The eventual objective of RSM is to find out the optimum conditions as compromise between higher CO_2 permeance and CO_2/CH_4 separation selectivity. The optimum condition preset at high and low level ranges of the three independent variables: pressure difference, temperature and CO_2 feed concentration for separation studies of CO_2/CH_4 binary gas mixture over HT-silica membrane is obtained using numerical optimization feature of Design Expert 6.0.6 Software. The optimization module in Design Expert searches for a combination of variable levels that simultaneously satisfy the requirements placed for all the responses and variables. Table 4 summarizes the optimization criteria used to seek the optimum values for the two responses. Table 5 presents the four optimum solutions generated by Design Expert software. By default, the solutions are sorted from best to worst depending on total desirability. In the present work, the desirability function approach is used in the response surface methodology to optimize the operating conditions. The experimental conditions that provide the highest desirability response value are determined using this method. The optimum conditions in Solution 1 with the highest total desirability of 1.000 are chosen for further process studies. The optimum conditions for CO_2 permeance and CO_2/CH_4 separation selectivity are found to be 100.94 kPa pressure difference, 30.09 ºC separation temperature and 43.05% CO_2 feed concentration. The developed model predicted that an optimum CO_2 permeance of 6.0×10^{-7} $mol.m^{-2}.s^{-1}.Pa^{-1}$ and CO_2/CH_4 separation selectivity of 109 can be obtained.

In order to check the validity of the DoE model predication, the predicted optimum for all the responses are then verified by performing five repeated experiments at the optimum conditions (solution 1). Table 6 summarizes the separation results for the CO_2/CH_4 binary gas mixture of the repeated experimental runs. The experimental values of the CO_2 permeance and CO_2/CH_4 separation selectivity at the optimum conditions are compared with the predicted values by the DoE software. The mean% error is 3.27 for CO_2 permeance and 3.80 for CO_2/CH_4 separation selectivity. The mean% error for both responses is less than 5% means that the experimental value is close to the predicted value. It can be concluded that the developed model by DoE software with RSM in this study shows good predictability and sufficient reliability for the modeling and predicting the HT-silica membrane performance for the permeation and separation of CO_2/CH_4 binary gas mixture.

Table 4 Constraint used for optimization of CO_2 permeance and separation selectivity of the CO_2/CH_4 binary gas mixture.

Criteria	Goal	Lower limit	Upper limit
Pressure Difference, kPa	In the range	100	500
Temperature, °C	In the range	30	190
CO_2 Feed Concentration, %	In the range	10	50
Permeance $\times10^{-7}$, $mol.m^{-2}.s^{-1}.Pa^{-1}$	Maximize	0.76	5.23
CO_2/CH_4 Separation Selectivity	Maximize	5	106

Table 5 Optimum condition of CO_2 permeance and separation selectivity of the CO_2/CH_4 binary gas mixture.

Solution	Pressure difference, kPa (A)	Temperature, °C (B)	CO_2 Feed Conc., % (C)	CO_2 Permeance $\times 10^{-7}$, $mol.m^{-2}.s^{-1}.Pa^{-1}$ (Y_1)	CO_2/CH_4 Separation selectivity (Y_2)	Total desirability
1	100.94	30.09	43.05	6.0	109.0	1.000
2	103.14	37.04	49.20	5.6	106.4	1.000
3	100.00	41.08	50.00	5.5	104.8	0.994
4	100.00	30.00	28.97	6.2	96.7	0.953

Table 6 Experiments at optimum conditions simulated by DoE for the CO_2 permeance and separation selectivity of CO_2/CH_4 binary gas mixture.

Run	CO_2 Permeance $\times 10^{-7}$, $mol.m^{-2}.s^{-1}.Pa^{-1}$		% Error	Separation Selectivity		% Error
	Experimental	DoE		Experimental	DoE	
1	6.25	6.0	4.17	114.2	109.0	4.77
2	6.21	6.0	3.50	112.6	109.0	3.30
3	6.29	6.0	4.83	115.5	109.0	5.96
4	5.94	6.0	1.00	108.4	109.0	0.55
5	6.17	6.0	2.83	113.8	109.0	4.40
Mean error			3.27			3.80

% Error = ((Exp.-DoE)/DoE)*100

4. Conclusions

Process modeling and optimization of mixed gas permeation and separation of CO_2/CH_4 through HT-silica membrane were performed using RSM coupled with CCD available in statistical method of DoE. The analysis of the RSM showed that the process variables (pressure difference, operating temperature and CO_2 feed concentration) had significant effects on CO_2 permeance and CO_2/CH_4 separation selectivity. From the study, it was evident that optimal conditions that maximize CO_2 permeance and favor higher CO_2/CH_4 separation selectivity were unfavorable at higher temperature and pressure difference. The developed models by DoE showed good predictability and sufficient reliability for the modeling and predicting the CO_2 permeance and separation selectivity of CO_2/CH_4 mixed gas through HT-silica membrane with the values of correction coefficient (R^2) higher than 0.95.

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List of Symbols

A	Factor code of pressure difference	-
B	Factor code of temperature	-
C	Factor code of CO2 feed concentration	-
K	Gas permeance	mol/m^2.s.Pa
N	Mole flux	mol/m^2.s
P	Pressure	Pa
ΔP	Pressure difference	Pa
R^2	Regression coefficient	-

Greek symbols

α	selectivity	-
β	Regression coefficient	m

Subscripts

F	Feed	-
p	Permeate	-
R	Retentate	-
i, j	Component gas CO_2 and CH_4	-