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Parameters Optimisation of Isopropanol Purification by Hybrid Distillation-Vapour Permeation Process Using Response Surface Methodology

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A hybrid distillation-vapour permeation (D-VP) system was selected to get Isopropanol (IPA) with high purity. In this study, a technical and economic analysis was performed comparing the hybrid D-VP systems with polymeric (PVA/PVDF) and ceramic (NaA type zeolite) membranes. Response surface methodology (RSM) was used to optimise the hybrid process parameters. First of all, the simulation of hybrid D-VP process for IPA purification was conducted with UniSim Design. Then Plackett-Burman design was employed to screen the significant parameters affecting total annual cost (TAC) from 11 variables. After that, steepest ascent method was undertaken to determine the optimal regions of the selected significant parameters. Finally, Box-Behnken design was adopted to further analyse the mutual interactions between these parameters and to identify their optimal values that would generate a minimum TAC. Optimal parameters were the vapour flow-rate to VP of 580 kg/h, VP operating temperature of 140 °C, permeate pressure of 1.5 kPa. And the TAC of optimal HDCM system is 49.06 €/t, about 19.80 % less than that of pri-optimised system.

1. Introduction

Isopropanol (IPA) is widely used as one of the important solvents and cleaners in modern chemical, semiconductor, pharmaceutical and electronic industries. Recycling of IPA from IPA-water mixtures is necessary from environmental and economic point of view. However, IPA forms an azeotrope with water at 87.4 wt.% of IPA at atmospheric condition, which makes the separation of these mixtures difficult and uneconomical by conventional methods such as distillation.

Vapour permeation (VP) offer a more promising and energy-efficient alternatives for azeotropic separation. Ceramic membranes are solvent and temperature stable, and zeolite NaA membranes offered significant potential for a dehydration agent with high permeation flux and separation factor as well as high chemical and thermal stability (Pina et al., 2004). However, VP itself still encounters some challenges such as the membrane productivity, the sensitivity to friction losses in the feed stream and the possibility of condensation (Cen and Lichtenthaler, 1995). Then application of VP alone is not an optimal choice.

In order to reduce energy consumption, hybrid distillation-vapour permeation (D-VP) system has attracted much attention in recent years (Naidu and Malik, 2011). Szitkai et al. (2002) modelled and optimised hybrid ethanol dehydration systems using MINLP, differential equations were employed for modelling the membrane modules, computational experiences used GAMS/DICOPT. Koczka et al. (2007) considered different configurations with rigorous modelling tools based on the comparison of total annual cost (TAC), three hybrid separation processes were rigorously modelled with CHEMCAD, and optimised with the dynamic programming optimisation method for the case of the separation of ethanol-water mixture. However, they did not consider structural and operational parameter optimisation of hybrid process.

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Most of the studies on hybrid distillation-membrane processes involved changing one of the independent parameters at a time while maintaining the others at a fixed level. Such studies ignored the interaction effects among the important parameters affecting the separation. One possible solution is to use the response surface methodology (RSM) which is widely used to analyse the effects of multiple factors and their interactions (Conto et al., 2015). It overcomes disadvantages of conventional methods and is proved to be an effective way for process parameters analysis that uses mathematical and statistical techniques to analyse the influence of independent variables on a specific response (Dahmoune et al., 2014).
In this paper, hybrid D-VP processes with ceramic membrane (HDCM) for IPA purification were simulated with UniSim Design based on the improved membrane module. Plackett-Burman (PB) design was used to screen the factors essential for TAC. Then steepest ascent method was employed to approach the vicinity of the optimal conditions. Subsequently, Box-Behnken design (BBD) for RSM was used to estimate the relationship between a response and optimal parameters for minimum TAC, and then the economic analysis was performed for the dehydration of IPA by HDCM.

2. Simulation and Experimental designs

2.1 Validation of D-VP model with UniSim Design

The simulation of HDCM process for IPA purification was conducted with UniSim Design. To verify the modelling results, the simulation output was compared with the corresponding data given in literature (Hoof et al., 2004) for the same conditions. The basic parameters were given in Table 1 (Hoof et al., 2004).

Table 1: Original data of literature

Description	Parameters	Value	Description	Parameters	Value
	Flow rate (kg/h)	1,000		Thermodynamic model	NRTL
	Fraction (IPA,wt%)	0.50		Number of trays	10
Feed	Temperature (°C)	20	Distillation	Pressure (kPa)	100
	Pressure (kPa)	110		Reflux ratio	1.35
	Location	9		Top flow rate (kg/h)	601
	Temperature (°C)	95			
Membrane	Permeate pressure (kPa)	2			
	Selectivity	200	Target	Retentate fraction (IPA,wt%)	0.995

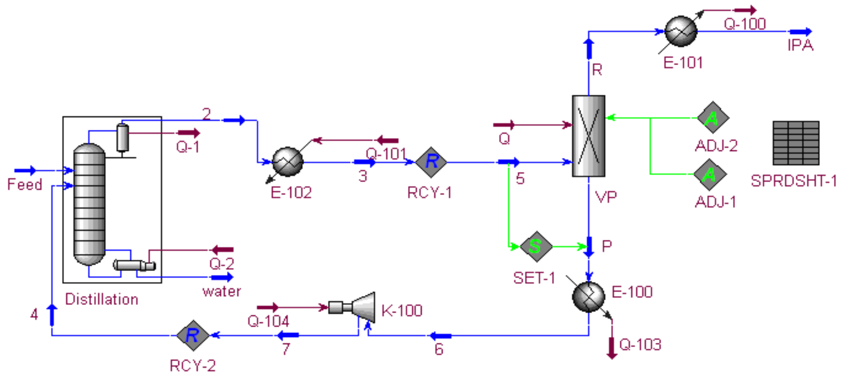

Figure 1: Simulation Model of IPA-water separation process in Unisim Design

Model of D-VP process for IPA-water separation in UniSim Design was shown in Figure 1. The feed enters the distillation column, where the aqueous isopropanol mixture is concentrated up to nearly close to the azeotropic concentration. The top vapour stream heated up to a specific temperature enters into the vapour permeation section, where the rest of the water is removed and recycled back to the distillation column. HDCM process for IPA purification model with UniSim Design was run. For distillation process, the flow rate and the composition of the top stream were 610 kg/h and 81.92 wt.% IPA. For membrane separation process, the retentate flow rate and the target IPA fraction were 500.8 kg/h and 99.79 wt.%, respectively. Compared with the literature results (Hoof et al., 2004), the percentage errors of the flow rate and the composition of the top stream were 1.47 % and 1.30 %, and the percentage errors of the retentate flow rate and the target IPA fraction were 0.16 % and 0.14 %. Therefore, the simulation results of D-VP model with UniSim Design were available.

2.2 PB Experimental designs

PB design is a powerful and efficient mathematical approach when rapidly searching for key factors in a multivariable system. It offers a good and fast screening procedure and mathematically computes the significance of a large number of variables with relatively few experiments, which is time saving and gives the effect of change in more than one factors in single experiment (Singh et al., 2011). In the studies, as showed in Table 2, PB design was employed to screen the significant parameters from 11 variables (including 3 dummy variables). TAC was considered as the response in the design. These variables were investigated and 12 tests were carried out. Each independent variable was examined at two levels: -1 for low level and +1 for high level. Table 2 illustrated the variables and their corresponding levels used in the experimental design. The design matrix selected for the screening of significant parameters for TAC and the corresponding responses were shown in Table 3.

Table 2: Levels of the variables tested in PB design

Variables	Symbol	Variation Levels	
		Low (–1)	High (+1)
Feed tray location	A	3	6
Permeate recycle location	B	3	6
Reflux ratio	C	1	2
Vapour flow-rate to VP (kg/h)	D	600	700
VP operating temperature (°C)	E	100	140
Permeate pressure (kPa)	F	1	3
Feed temperature (°C)	G	20	80
Column pressure (kPa)	H	101.3	110
Dummy	I,J,K	-	-

Table 3: PB design matrix and response values

Run order	A	B	C	D	E	F	G	H	I	J	K	TAC (€/t)
1	1	1	-1	1	1	1	-1	1	1	-1	-1	56.53
2	-1	1	1	-1	1	1	1	-1	-1	1	-1	50.06
3	1	-1	1	1	-1	1	1	-1	-1	-1	-1	77.61
4	-1	1	-1	1	1	-1	1	-1	-1	-1	1	50.25
5	-1	-1	1	-1	1	1	-1	1	1	-1	1	51.34
6	-1	-1	-1	1	-1	1	1	1	1	1	1	77.79
7	1	-1	-1	-1	1	-1	1	1	1	1	-1	55.15
8	1	1	-1	-1	-1	1	-1	-1	-1	1	1	72.32
9	1	1	1	-1	-1	-1	1	1	1	-1	1	54.36
10	-1	1	1	1	-1	-1	-1	1	1	1	-1	61.32
11	1	-1	1	1	1	-1	-1	-1	-1	1	1	59.15
12	-1	-1	-1	-1	-1	-1	-1	-1	-1	-1	-1	59.60

A-K are symbols shown in Table 2.

2.3 Steepest Ascent Method

The initial estimate of the optimal conditions was far from the actual optimum. Steepest ascent method is a simple and efficient procedure for moving the region of a response in the direction of the maximum change toward the optimum. The zero level of PB design was identified as the basic point of steepest ascent path. Experiments were conducted along with the steepest ascent path until the response showed no further increase. This point would be near the optimal point and could be selected as the center point of BBD (Zhou et al., 2011).

2.4 Response surface methodology

The optimal levels of the significant factors and the interactions of these variables on TAC were analysed by BBD. In this study, a three-factor, three-level BBD with 15 runs was employed. The second-order model used to fit the response to the independent variables was shown in Eq(1):

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_i \sum_j \beta_{ij} X_i X_j \qquad (1)$$

where Y is the predicted response; X_i and X_j are input variables that influence the response Y ; k is the number of variables; β_0 is the constant term; β_i is the linear coefficient; β_{ii} is the quadratic coefficient and β_{ij} is the interactive coefficient. Design-Expert 7.1.3 was used for the experimental designs as well as for regression and graphical analysis of the experimental data obtained.

3. Results and Discussions

3.1 Screening of significant parameters by PB design

The data listed in Table 3 indicated that for HDCM process there was a wide variation in the TAC from 50.06 €/t to 77.79 €/t in the 12 trials. This variation suggested that medium and culture condition optimisation were important for reducing TAC. Partial regression coefficients and analyses of their significance were evaluated for determination of the effects on TAC shown in Table 4. The analysis of the regression coefficients and contribution of 11 factors showed that A, D, F, G, J, K and L had positive effects on TAC, whereas B, C, E and H had negative effects on TAC for both HDCM process. Variables with significant effect were E > F > D > A > B > G > C > L > J> K> H for HDCM. The first three variables with significant effect E(VP operating temperature), F (permeate pressure) and D(vapour flow-rate to VP) were selected for further optimisation to obtain a minimum response for HDCM process.

Table 4: Partial regression coefficients and analyses of their significance

Factors	Stdized Effects	Sum of squares	Contri-bution	Signifi-cance	Factors	Stdized Effects	Sum of squares	Contri-bution	Signifi-cance
A	4.42	58.65	4.86	4	G (°C)	3.35	33.63	2.78	6
B	-3.59	38.56	3.19	5	H (kPa)	-0.038	0.0044	0.00037	11
C	-2.63	20.67	1.71	7	J	1.18	4.16	0.34	9
D (kg/h)	7.28	159.07	13.17	3	K	0.31	0.27	0.02	10
E (°C)	-12.79	490.63	40.63	1	L	2.74	22.52	0.93	8
F (kPa)	9.67	280.43	23.22	2					

3.2 Optimisation by steepest ascent path experiment

Based on the above regression analysis, the path of steepest ascent was started from the zero level of the PB design and moved along the direction in which E increased and D, F decreased to reduce TAC. Table 5 illustrated the changing directions of the three variables. The minimum TAC was at the third order and no further improvement could be achieved in the response. The parameters were vapour flow-rate to VP of 620 kg/h, VP's operating temperature of 120 °C, permeate pressure of 1.5 kPa. It suggested that these points were near the optimal points and then were chosen for further optimisation, taken as the 0 level in RSM.

Table 5: Experimental design of steepest ascent and experimental data

Run order	D (kg/h)	F (kPa)	E (°C)	TAC (€/t)	Run order	D (kg/h)	F (kPa)	E (°C)	TAC (€/t)
1	580	2.5	100	65.39	4	640	1	130	53.86
2	600	2	110	55.85	5	660	0.5	140	55.04
3	620	1.5	120	53.79					

3.3 Optimisation by Box-Behnken design

Based on the PB design and the method of the steepest ascent, three variables (vapour flow-rate to VP, VP operating temperature and permeate pressure) were used to determine the optimum levels of these parameters. The design matrix of the variables was given in Table 6, along with the experimental values of the response.

The mathematical model for HDCM process representing the TAC as a function of the independent variables within the region under investigation was expressed by the following Eq(2) based on the simulation data of Table 6:

$$TAC = 53.32 + 0.18D - 1.33E + 16.94F + 0.00032DE - 0.0063DF - 0.13EF - 0.00012D^2 + 0.0047E^2 + 1.35F^2 \qquad (2)$$

The statistical significance of Eq(2) was confirmed by an F-test, and the analysis of variance (ANOVA) for response surface quadratic models were summarized in Table 7.

As can be seen from Table 7, the F-value of 24.63 implied that the model was significant. There is only a 0.13 % chance that a "Model F-value" could occur due to noise. The P-values were used to check the significance of each variable, and the smaller P-value is, the bigger the significance of the corresponding variable is. Values of "Prob>F" less than 0.05 indicated that the model fitness was significant. According to the P-values, the model terms D, E, F, EF and E^2 were found significant. The model determination coefficient R^2 suggested that the fitted model could explain 97.79 % of the total variation. Those implied the regression model was very reliable for IPA purification in the present study from the above analysis.

By solving the Eq(2), the optimal values of the parameters were listed in Table 8. It was found that the minimum TAC was 48.54 €/t IPA for HDCM.

Table 6: BBD and simulation values

Run order	D (kg/h)	F (kPa)	E (°C)	TAC (€/t)	Run order	D (kg/h)	F (kPa)	E (°C)	TAC (€/t)
1	580	1.5	100	56.65	9	620	0.5	100	56.39
2	660	1.5	100	61.37	10	620	0.5	140	52.50
3	580	1.5	140	49.06	11	620	2.5	100	66.60
4	660	1.5	140	54.80	12	620	2.5	140	52.54
5	580	0.5	120	50.67	13	620	1.5	120	53.78
6	660	0.5	120	56.92	14	620	1.5	120	53.78
7	580	2.5	120	53.47	15	620	1.5	120	53.78
8	660	2.5	120	58.72					

Table 7: ANOVA for the regression quadratic model equation of BBD

Source	Sum of Squares	df	Mean Square	F Value	P Value (Prob>F)	Comments
Model	262.12	9	29.12	24.63	0.0013	significant
D (kg/h)	60.34	1	60.34	51.02	0.0008	significant
E (°C)	128.87	1	128.87	108.96	0.0001	significant
F (kPa)	27.56	1	27.56	23.3	0.0048	significant
DE (°C·kg/h)	0.26	1	0.26	0.22	0.6598	not significant
DF (kPa·kg/h)	0.25	1	0.25	0.21	0.6638	not significant
EF (°C·kPa)	25.83	1	25.83	21.84	0.0055	significant
D^2 (kg^2/h^2)	0.13	1	0.13	0.11	0.7561	not significant
E^2 (°C^2)	12.97	1	12.97	10.96	0.0212	significant
F^2 (kPa^2)	6.73	1	6.73	5.69	0.0627	not significant
Residual	5.91	5	1.18			
Lack of Fit	5.91	3	1.97			
Pure Error	0	2	0			
Cor Total	268.03	14		R^2=97.79%		

Table 8: Optimal values for D-VP process with different membranes

Item	vapour flow-rate to VP (kg/h)	VP operating temperature (°C)	permeate pressure (kPa)	TAC (€/t IPA)
Data	580	140	1.5	48.54

3.4 Economic evaluations

The TAC of the HDCM system was divided up in operation cost, investment cost and maintenance cost. Table 9 showed the costs for the process investigated. When the operation costs of pre-optimised and optimised D-VP processes with different membranes are compared, the systems optimised have the higher operation costs for HDCM. This may be because the VP process of pre-optimised was performed at operationg temperature of 95 °C and permeate pressure of 2 kPa while the system optimised was performed at operationg temperature of 140 °C and permeate pressure of 1.5 kPa. Due to the increased requirement of heating steam as the desired temperature to VP increased, and more requirement of cooling energy as permeate pressure decreased, the operation costs increased. For the HDCM pre-optimised, membrane area of 25 m^2 was needed, and for the system optimised, membrane area of 12 m^2 was needed. The different investment and maintenance costs between HDCM pre-optimised and optimised is due to the fact that more membrane area is required with VP systems pre-optimised than that optimised.

4. Conclusions

In the present study, RSM was used to optimise the hybrid D-VP process parameters for IPA purification with HDCM process, TAC as the response value. Optimal parameters were the vapour flow-rate to VP of 580 kg/h, VP operating temperature of 140 °C, permeate pressure of 1.5 kPa, the minimum TAC predicted was 48.54 €/t. The HDCM pre-optimised and optimised were simulated in Unisim Design, it can be concluded that the TAC of optimal HDCM system is 49.06 €/t, about 19.80 % less than that of pri-optimised system.

Table 9: Economic evaluation for D-VP process

Costs	pre-optimised	optimised
Cooling water costs (€/y)	6,017	7,267
Steam costs (€/y)	45,867	50,844
Permeate condensing costs (€/y)	255	404
Total operation costs (€/y)	52,139	58,515
Specific operation costs (€/t IPA)	14.47	16.23
Distillation columns (€/y)	60,851	60,851
VP unit (€/y)	15,924	7,292
Total investment costs (€/y)	76,775	68,087
Specific investment costs (€/t IPA)	21.31	18.88
Membrane replacement (€/y)	71,238	32,368
Maintenance installations (€/y)	20,204	17,917
Total maintenance costs (€/y)	91,442	50,285
Specific maintenance costs (€/t IPA)	25.39	13.95
Specific TAC (€/t IPA)	61.17	49.06

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