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An Optimisation Strategy for Enhancing the Product Recovery of a Pressure Swing Adsorption through Pressure Equalisation or Co-current Depressurisation: a Case Study of Recovering Hydrogen from Methane

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Abstract

The step sequence of a PSA (Pressure Swing Adsorption) cycle for gas separation can be as simple as the four steps of Skarstrom cycle, but it can also be made very complex by adding new steps with a view to enhancing the product recovery. In particular, pressure equalisation (PE) or co-current depressurisation (CoD) steps are often introduced to the step sequence of a H₂ purification PSA, as the strategies have been proven successful. However, the PE and CoD steps have been incorporated into the PSA cycle without due consideration

on what is the best way of including and operating the steps. In this study, an equilibrium theory method was taken to optimise the PE and CoD steps for improving the product recovery to the maximum. The theoretical PSA model turned out reliable and insightful when applied to an exemplary H₂ purification PSA system, as the results obtained by simply solving the algebraic equations were so consistent with those of the sophisticated numerical simulation and optimisation. In this study, the equilibrium theory analysis of a PSA elucidated clearly that there exists the optimal number of the PE steps, and the optimal column pressure at the end of CoD step and the results would be affected greatly by the operating conditions, such as feed composition, operating pressure, etc.

Keywords: Pressure Equalisation; Co-current Depressurisation; Equilibrium Theory; Pressure Swing Adsorption; Nonlinear isotherm; Hydrogen; Methane; Optimisation

1. Introduction

Pressure swing adsorption (PSA) is a well-established and highly advanced gas separation process, being widely used for gas separation and purification in industry. A PSA system is capable of separating a gas mixture by means of porous solid materials having different affinities toward each component comprising a gas mixture that lead to differences of adsorption amounts or rates. The first PSA process, coined Skarstrom cycle, consists of two adsorption beds each of which goes through four basic steps of adsorption, blowdown, purge and pressurisation in sequence¹. Thereafter, the design of PSA has evolved to incorporate new steps, such as pressure equalisation (PE), co-current depressurisation (CoD), heavy reflux, etc., into the adsorption cycle, with the aim of enhancing its separation performance. The more complex becomes the step sequence of a cycle, the more adsorption beds does a PSA system need to have in constructing the PSA cycle scheduling table^{2, 3}.

It is well known that the benefit from adding PE step into the basic adsorption cycle is twofold: PE step can improve the light product recovery so effectively and it can also increase the purity

of the heavy component product obtained during the blowdown step. However, adding PE steps involves an adverse effect on the PSA performance, resulting in the bed productivity getting worse^{4, 5}. The poor productivity issue is attributed to the following two reasons: One is that PE steps would reduce the amount of feed that could be processed during one cycle and the other is that PE steps would often require more idle steps to be strewn over the PSA cycle scheduling table to meet the requirement for bed connectivity⁶. Nevertheless, use of PE steps is deemed as a very useful way of improving the light product recovery or the heavy product purity at the sacrifice of the bed productivity⁷.

Table 1. Overview of PSA processes with various PE steps

Ref.	Adsorbent	Number of beds	Number of steps	Number of PE steps	pressure before blowdown (bar)	Origin of purge gas	P_H (bar)	P_L (bar)	Feed composition (%)	Purity (%)	Recovery (%)
Moon et al., 2018 ⁸	°AC+ Zeolite LiX	8	11	3	~6	^d pv	35	1.1	H ₂ = 88.75 CO = 2.66 N ₂ = 5.44 CO ₂ = 2.12 Ar = 1.03	99.99	~89
		8	11	3	~7.5	pv				99.99	~85.5
		8	12	4	~7	pd				99.99	~87
Nikolic et al., 2008 ⁹	AC	1	4	0	-	pd	26.3	1	H ₂ = 75.5 CO ₂ = 17 CO = 4 CH ₄ = 3.5	96.87	34.40
		4	6	1	-	pd				99.50	58.06
		8	8	2	-	pd				99.58	68.87
		12	10	3	-	pd				99.36	72.50
Subraveti et al., 2019 ¹⁰	AC	Non-scheduled	6	1	21.8	steam purge	34.5	10.13	H ₂ = 60 CO ₂ = 40	99.9	77.7
			8	2	17.5					99.9	83.6
			10	3	15.6					99.9	86.3
Luberti et al., 2014 ³	Zeolite 5A	4	9	2	~7.5	pv	34	1	H ₂ = 88.75 CO ₂ = 2.12 CO = 2.66 N ₂ = 5.44 Ar = 1.03	99.999	70.56
		6	9	2	~4	pv				99.999	78.75
		6	11	3	~4	pv				99.999	76.11
		9	11	3	~3.5	pv				99.998	87.05
		12	13	4	~2.5	pv				99.999	89.11
Lopes et al., 2011 ¹¹	AC	Non-scheduled	6	1	2.75	pd	5	0.5	H ₂ = 79 CO ₂ = 17 CH ₄ = 2.1 CO = 1.2 N ₂ = 0.7	99.992	70.96
			10	3	1.63	pd				99.993	79.98
			6	1	5.5	pd	10	1		100	47.1
			10	3	3.25	pd				100	67.50
Nikolic et al., 2009 ¹²	AC	1	4	0	-	pd	25	1.5	H ₂ = 75.5 CO ₂ = 17 CO = 4 CH ₄ = 3.5	99.99+	~33
		2	6	1	-	pd				99.99+	~59
		5	8	2	-	pd				99.99+	~71
		8	10	3	-	pd				99.99+	~72
Waldron and Sircar, 2000 ¹³	AC	4	7	1	-	pv	14.8	1.36	CH ₄ = 20 H ₂ = 80	99.999	85.0
		4	9	2	-	pv				99.999	86.5
		5	11	3	-	pv				99.999	87.5
Azpiri Solares and Wood, 2020 ^{14, 15}	AC	4	7	1	-	pv	36.7	1	H ₂ = 60 CO ₂ = 40	99.994	94.35
		5	9	2	-	pv				99.994	95.61
		6	11	3	-	pv				99.994	96.87
Delgado et al., 2011 ¹⁵	Silicate	1	4	0	-	pd	7	0.1	CH ₄ =50 N ₂ =50	96.5	38.5
		2	6	1	-	pd				96	75.2
		3	8	2	1	pd				96.3	96.7

Note: a: value with '~' means data read from figures; b: pd: product purge; c: AC: activated carbon; d: pv: providing purge.

The effects of PE steps on the PSA performance have been extensively studied so far and the results of the past researches are summarised in Table 1. It can be deduced by reflecting on Table 1 that the product recovery is increased by adding a PE step into the basic four step, and it can be increased further by adding more PE steps. Another point that is worth noting is that providing purge step was added to the adsorption cycle by either placing the step between two DPE (depressurising pressure equalisation) steps or after the final DPE step. During the providing purge step, a column is depressurised by releasing the gas co-currently to the gas flow direction of the feed step and the effluent gas is used as a purge gas. As a result, the light component product obtained could be saved from its use as a purge gas. The providing purge step is also called co-current depressurisation (CoD) step, as the purge gas stream is generated by depressurising the column in the direction of gas flow in the feed step. In principle, both DPE and CoD steps are similar to each other in that a high purity of light component can be produced.

As can be seen in Table 1, various PSA cycles have been studied in an attempt to see the effect of the number of PE steps on the PSA performance. While it is obvious that the light product recovery was increased significantly by having PE steps, it is noticeable that the extents of the improvement were diminished with more PE steps added. In other words, the product recovery would increase to a lesser extent when the number of PE steps increases to 2 from 1 than when a PE step is added to the basic four step. Likewise, adding another PE to the existing 2 PE cycle would not be able to enhance the product recovery as much as what would be achievable by increasing the number of PE steps to 2 from 1. There may be the number of PE steps where adding another PE step would not be able to increase the product recovery any more.

This work is to clarify what would happen to the product recovery in case of PE steps being added to the adsorption cycle using an equilibrium theory analysis method. The theoretical model of a PSA system containing pressure equalisation or forward depressurisation (co-

current depressurisation in this study) was originally developed by Chiang¹⁶. The equilibrium theory model was solved in this study to evaluate and optimise the cycle performance of a PSA process with multiple PE steps or CoD step. The equilibrium theory model was solved and applied to a PSA system for separating methane from hydrogen to see the effect of PE steps or CoD step on the PSA performance.

2. Equilibrium Theory model of a PSA System with Co-current Depressurisation (CoD) or Pressure Equalisation (PE)

2.1. Governing equations

In this work, the governing equations are expressed in reference to the past works¹⁶⁻¹⁸. The feed is a binary gas mixture of the strongly adsorbing component *A* and the weakly adsorbing component *B*. The gas is assumed to be ideal gas. The two adsorption isotherms are uncoupled and they are both linear. In principle, the energy and momentum balances around an adsorption column are expressed in such a rigorous way that it considers kinetic energy as well as convective energy and the heat of adsorption^{19, 20}. The energy and momentum balances are not required in this case, as the adsorption system is assumed to be isothermal with no pressure drop. Under the general assumptions of equilibrium theory, the mass balance equation of component *A* can be written as:

$$\frac{\partial Py}{\partial t} + \beta_i \frac{\partial uPy}{\partial z} = 0 \quad \beta_i = \frac{1}{1 + \left(\frac{1-\varepsilon}{\varepsilon}\right) k_i} \quad (1)$$

Starting from Eq.1, the equations for estimating the gas velocity along the column can be derived. During constant-pressure steps, the gas velocities at two arbitrary locations 1 and 2 along the column are related by:

$$\frac{u_1}{u_2} = \frac{1 + (\beta - 1)y_2}{1 + (\beta - 1)y_1} \quad \beta = \frac{\beta_A}{\beta_B} \quad (2)$$

During pressure-varying steps, the gas velocity at either of the two column ends is zero, i.e. $u = 0$ at $z = 0$ or L . The gas velocity equation is expressed differently depending on the boundary condition as follows:

$$u = \frac{-z}{\beta_B[1 + (\beta - 1)y]} \frac{1}{P} \frac{dP}{dt} \quad u = 0 \text{ at } z = 0 \quad (3a)$$

$$u = \frac{L - z}{\beta_B[1 + (\beta - 1)y]} \frac{1}{P} \frac{dP}{dt} \quad u = 0 \text{ at } z = L \quad (3b)$$

The component mass balance, Eq. 1, can be rearranged into a form of quasilinear partial differential equation.

$$\frac{\partial y}{\partial t} + \frac{\beta_A u}{1 + (\beta - 1)y} \frac{\partial y}{\partial z} = \frac{y(y - 1)(1 - \beta)}{1 + (\beta - 1)y} \frac{d \ln P}{dt} \quad (4)$$

Eq. 4 can be solved by the method of characteristics. As a result, the following two ordinary differential equations are obtained:

$$\frac{dz}{dt} = \frac{\beta_A u}{1 + (\beta - 1)y} \quad (5)$$

$$\frac{dy}{dP} = \frac{(\beta - 1)(1 - y)y}{[1 + (\beta - 1)y]P} \quad (6)$$

In case of adsorption breakthrough of an adsorption system with linear isotherms, a shock wave is expected to evolve by self-sharpening effect if the column is sufficiently long. The shock wave progresses along the column at the shock wave velocity that is estimated by the weak solution of the component mass balance as follows.

$$u_s = \left. \frac{dz}{dt} \right|_s = \beta_A \frac{u_2 y_2 - u_1 y_1}{y_2 - y_1} \quad (7)$$

where 1 and 2 indicate positions ahead of and behind the shock wave, respectively.

2.2. Equilibrium theory analysis of CoD and PE steps

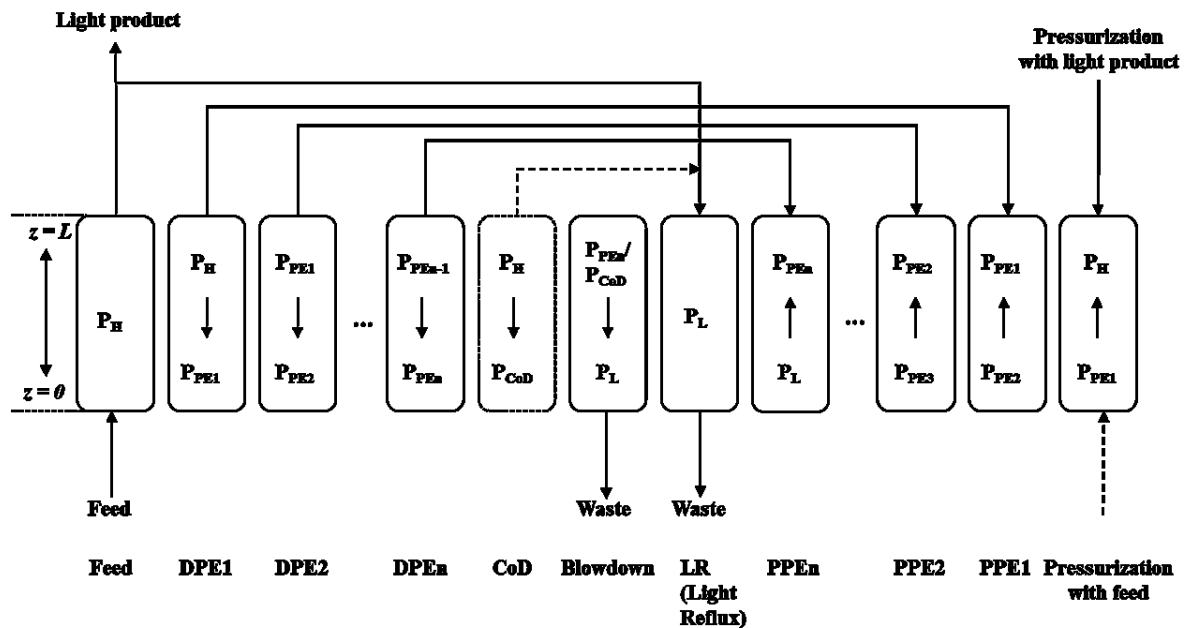


Figure 1. A schematic to describe the adsorption column of a general PSA system undergoing the step sequence of an adsorption cycle and depict the gas flow direction and the bed connectivity at each step. Note that the CoD (Co-current Depressurisation) step can also be positioned between two DPE (Depressurising Pressure Equalisation) steps rather than after the final DPE step.

The basic four step sequence of a Skarstrom PSA cycle can be expanded by incorporating CoD or PE steps to improve the product recovery. A general PSA cycle designed to produce the light component of a very high purity is presented in Figure 1. In this study, two different cohorts of the step sequences were studied.

- PSA with PE (Pressure Equalisation): Feed – (multiple) DPE – blowdown – Light Reflux – (multiple) PPE – LPP (or FP)
- PSA with CoD (Co-current depressurisation): Feed – CoD – blowdown – Light Reflux – LPP (or FP)

The two cases share common features in that the column is depressurised by opening the valve near the column end at $z=L$ and accordingly the light product is released out of the column in the direction co-current to the gas flow at the adsorption step. They are different from each other with respect to what column the depressurising column is connected to. While the CoD step often acts to provide a purge stream required to regenerate another column, the DPE step is to discharge the light component stream with which to pressurise another column at a lower pressure. Considering the functions of the two steps, it is essential to run a PSA system in such a way that the gas effluents of the column in CoD or DPE steps contain the heavy component as little as possible.

In case of a Skarstrom cycle, the column has to be made long enough to ensure the formation of a perfect shock wave, and the adsorption step has to be designed so that the shock wave reaches the product end at the end of adsorption step. By doing so, the column is entirely saturated with the feed with the adsorbents fully utilised during the adsorption step. During the subsequent blowdown step, the gas mole fraction may well vary with time due to desorption occurring with depressurisation, but it does not vary spatially.

The similar strategy of PSA design and operation also applies to a PSA system with pressure equalisation or co-current depressurisation. At the start of the blowdown step, the shock wave must reach the column end. However, the adsorption step has to be terminated earlier, considering the distance that the shock wave would travel further toward the product end during the PE or CoD steps. Also the shock wave has to be fully developed during the adsorption step.

We should be able to estimate the distance that a shock wave would travel while the column is depressurised. Obviously the distance depends highly on the extent of depressurisation. While the column pressure at the end of CoD step can be chosen arbitrarily by operators, the column pressure at each PE step cannot but change stepwisely, determined by the equilibrated pressure that the column reaches after connecting two columns that were initially at high and low pressures respectively. The equilibrated pressure that the pressures

of two connected columns converge to at the end of the i th DPE step of n PE steps is calculated by:

$$P_{E_i} = \frac{iP_L + (n + 1 - i)P_H}{n + 1} \quad (8)$$

The gas mole fraction at the end of the final PE step is calculated from the feed gas mole fraction and the pressure ratio as follows:

$$\left(\frac{y_{E_n}}{y_f}\right)^{\frac{1}{(\beta-1)}} = \left(\frac{1 - y_{E_n}}{1 - y_f}\right)^{\frac{\beta}{(\beta-1)}} \left(\frac{P_H}{P_{E_n}}\right)^{-1} \quad (9)$$

The number of moles of the light component leaving the column during DPE steps or CoD step, N_{DPE} , can be estimated by:

$$N_{DPE} = \phi \beta (\psi_H - \psi_{E_n}) = \beta \phi \psi_H \left[(1 - Y_{ads} y_f) - (1 - y_f) Y_{ads}^{\frac{1}{\beta}} \right] \quad (10)$$

where ϕ is the number of moles of the light component consumed for complete regeneration, i.e. $\phi = \frac{\varepsilon A_{cs} L P_L}{\beta_A R T}$.

The distance that the shock wave travels further during the PE steps (or CoD step) can be estimated simply by the mass balance around the column. Accordingly, the dimensionless position of the shock wave at the end of adsorption step, Y_{ads} , can be found by solving the following equation¹⁷:

$$Y_{ads} y_f + (1 - y_f) Y_{ads}^{\frac{1}{\beta}} - \frac{\psi_{E_n}}{\psi_H} = 0 \quad (11)$$

In case of CoD case, Y_{ads} can also be found by replacing ψ_{E_n} with ψ_{CoD} , the ratio of the column pressure at the end of CoD step to the desorption pressure.

2.3. Product Recovery of a PSA system with CoD or PE steps¹⁶

After the last DPE step (or CoD step), the column is depressurised from P_{E_n} (or P_{CoD}) to the desorption pressure, P_L , with the mole fraction in the gas phase increasing from y_{E_n} (or y_{CoD}) to y_b accordingly. Afterwards the adsorbents are regenerated by flowing a purge gas through the column. During the purge step, the purge gas is sourced from either a part of the light component product or the CoD step's effluent, being admitted to the column at $z = L$. In case of the column being not fully regenerated, the column can be divided into two sections with respect to whether or not the heavy component is present inside the section. The length ratio of the fully-regenerated section to the entire column is defined as extent of purging, X . The moles of the purge gas consumed for regeneration is:

$$N_{pg} = \phi X \quad (12)$$

The minimum extent of purging, X_{min} , is the ratio of the purge gas amount required to push the concentration front of y_b out of the column to ϕ . X_{min} is calculated by:

$$X_{min} = [1 + (\beta - 1)y_b]^2 \quad (13)$$

After regeneration, the column is repressurised by light component or feed during PPE steps followed by either LPP (Light Product Pressurisation)²¹ or FP (Feed Pressurisation) steps. In case of LPP, the adsorption column is pressurised from desorption pressure to adsorption pressure solely by light component. In contrast, in case of FP, the column is pressurised up to P_{E_1} by light component and it follows pressurising the column further to P_H with feed. The total number of moles of the light component consumed for pressurising the column is estimated by:

$$N_{BF} = \phi\beta(Z\psi_H - 1) \quad (14)$$

where the extent of backfill, Z , is defined as the ratio of the column pressure raised by light component only to the adsorption pressure. $Z < 1$ for feed pressurisation and $Z = 1$ for light product pressurisation.

At the end of the purge step, the location of the heavy component front at its mole fraction of zero is distanced from the feed end by $L(1 - X)$. The position moves back and forth along the column in the process of pressurising the column with either light component or feed that are admitted to the column at $z = L$ or $z = 0$, respectively. At the end of LPP or FP steps when the column is fully pressurised, the position of the heavy component front at its mole fraction of zero is estimated by:

$$z_f = L[1 - Z^\beta + (1 - X)\psi_H^{-\beta}] \quad (15)$$

During the adsorption step, the concentration front evolves into a perfect shock wave by self-sharpening effect. It is essential to find the position where the perfect shock wave emerges firstly during the adsorption step, Y_s . To find Y_s , we need to construct the mass balance equation of heavy component around the column and simultaneously estimate how far the heavy component front at its mole fraction of zero travels. Y_s can be estimated by:

$$Y_s = 1 - \left[\frac{Z^\beta - \beta Z - (1 - y_f)(1 - \beta)}{(1 - \beta)y_f} + \frac{[1 - (1 + \Lambda)(1 - \sqrt{X})]^2 - 1}{(1 + \Lambda)(1 - \beta)y_f\psi_H^\beta} \right] \quad (16a)$$

where

$$\Lambda = \frac{1 + (\beta - 1)y_f}{(1 - \beta)y_f\psi_H^{1-\beta}} \quad (16b)$$

To enable an adsorption column to be fully utilised in a PSA system, the shock wave must develop fully, so that it has the mole fraction range of 0 to y_f before the adsorption step ends.

In other words, Y_s has to be less than 1 in case of Skarstrom cycle. As for a PSA cycle with PE or CoD steps, Y_{ads} is determined by Eq. 11 and Y_s must satisfy the following condition:

$$Y_s \leq Y_{ads} \quad (17)$$

Eq. 16 implies that there exists a critical extent of purging, X_{cr} , to make Y_s equal to Y_{ads} . In operating a PSA system, the purge gas consumption has to be determined so as to ensure that the extent of purging, X , is equal to or larger than X_{cr} . The critical extent of purging is also affected by the extent of backfill, Z . Given the fact that feed pressurisation leads the heavy component's front to move toward the product end, the critical extent of purging for a PSA with feed pressurisation ($Z < 1$) will always be larger than the critical extent of purging for a PSA with light product pressurisation ($Z = 1$).

The total number of moles of the feed entering the column during both feed pressurisation and adsorption steps are:

$$N_H = \phi\psi_H Y_{ads} - \frac{\phi(1 - \sqrt{X})^2}{(1 - \beta)y_f} \quad (18)$$

During the adsorption step, a pure light component is produced as the effluent at the product end. The number of moles of light component product is:

$$N_{HP} = N_H[1 + (\beta - 1)y_f] - \phi\psi_H\beta(1 - Z) \quad (19)$$

The recovery of the process can be calculated by:

$$\begin{aligned} R &= \frac{N_{HP} - (N_{BF} - N_{DPE}) - N_{pg}}{(1 - y_f)N_H} \\ &= 1 - \frac{y_f}{1 - y_f} \frac{\beta(1 - \sqrt{X})^2 + \beta(1 - \beta)\psi_H(1 - y_f)Y_{ads}^{\frac{1}{\beta}} + (1 - \beta)(X - \beta)}{(1 - \beta)y_f\psi_H Y_{ads} - (1 - \sqrt{X})^2} \end{aligned} \quad (20)$$

The cycle throughput, defined by the number of moles of the net light component product per one cycle, is

$$T = N_{HP} - (N_{BF} - N_{DPE}) - N_{pg} \quad (21)$$

At the very initial stage of purge step, the purge gas effluent has the heavy component mole fraction of y_b , thus the stream must be directed to the bottom product pool. After the minimum extent of purging, the heavy component mole fraction of the effluent would start decreasing but is still so high that its addition to the bottom product can increase the heavy component's mole fraction. At some point of the purging step there exists an extent of purging where the gas effluent's composition is exactly the same as the average mole fraction of the heavy product pool. Beyond the threshold, the gas effluent would end up diluting the heavy product pool, so the purge step must end to maintain the bottom product purity as high as possible. The optimum extent of purging where the heavy component purity of the bottom product reaches the maximum, X_{opt} , is found by solving Eq.22.

$$\bar{y} = \frac{y_f}{1 - (1 - y_f)R} = \frac{1 - \sqrt{X_{opt}}}{1 - \beta} \quad (22)$$

By substituting Eq. 20 into Eq.22, it is simplified into Eq.23.

$$\frac{Y_{ads}\psi_H y_f}{(1 - \sqrt{X_{opt}})} - \frac{\beta\psi_H(1 - y_f)Y_{ads}^{\frac{1}{\beta}}}{(\sqrt{X_{opt}} - \beta)} = 1 \quad (23)$$

By solving Eq.23, we can find the optimum extent of purging, X_{opt} , for a general PSA system that is depicted in Figure 1.

To optimise the PSA operation with respect to the light component recovery, it is important to choose the proper extent of purging between the critical and optimum extents of purging.

Obviously, the actual extent of purging must be equal to or larger than X_{cr} in any case, as it is crucial to ensure that a perfect shock wave be formed during the feed step. If X_{opt} is greater than X_{cr} , we must choose X_{opt} to achieve the maximum product recovery. Otherwise X_{cr} should be taken in order to guarantee the formation of a perfect shock wave. In case of a PSA system with the column regenerated as much as X_{cr} during the purge step, the perfect shock wave would be formed firstly when the adsorption step ends.

3. Case study: Recovering hydrogen from methane

3.1. Adsorption isotherms

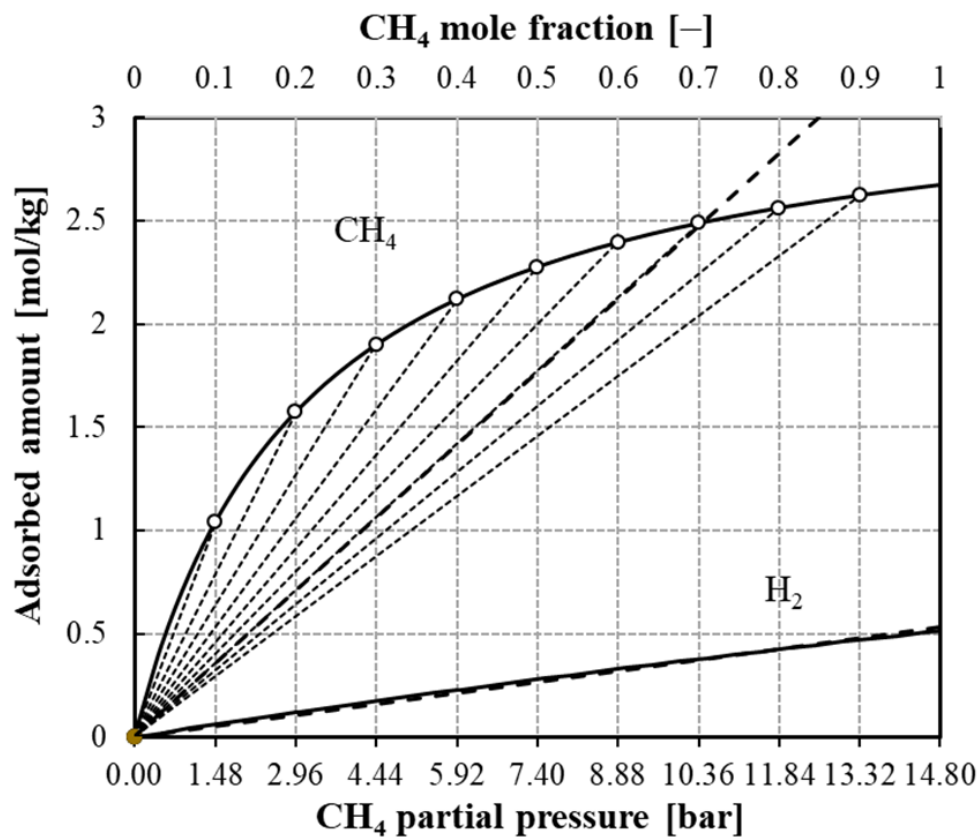


Figure 2. Langmuir isotherms of CH₄ and H₂ on activated carbons at 303.15K¹³ with the secant lines of the CH₄ isotherm and the linear fit of the H₂ isotherm.

It was aimed to make use of the equilibrium theory model in order to show the effect of pressure equalisation or co-current depressurisation on the PSA's light component product recovery. In this study, an exemplary PSA system designed for separating hydrogen from methane was chosen. The design and operating conditions of the H₂ purification PSA system were based on the Waldron and Sircar's past work¹³. The equilibrium theory model of a PSA system in this study was developed under the assumptions of both adsorption isotherms being linear, while the numerical simulations of the Waldron and Sircar's work were conducted with

the nonlinear Langmuir isotherms for both components as shown in Figure 2. The Langmuir isotherm parameters of each gas are presented in Table 2.

Table 2. Langmuir isotherm parameters on activated carbon (particle density = 776 kg/m³) at 303.15K¹³ and slopes of the linear fits of the isotherms.

Gas	q_0 [mol/kg]	b_i [bar ⁻¹]	Slope of the linear fit [mol/kg/bar]	R ²	k_i [-]
CH ₄	3.24	0.319	0.2385	0.9181	4.666
H ₂	3.24	0.013	0.03619	0.9988	0.708

In this case, methane is the strongly adsorbing component and hydrogen is the weakly adsorbing component. Accordingly, the methane isotherm on activated carbon is highly favourable while the hydrogen isotherm can be fitted well to a straight line, as shown in Figure 2. In this study, the effect of the CoD or PE steps on the PSA performance was studied with the blending ratio of methane and hydrogen in the feed varying from 0 to 1 with the 0.1 interval. Given the highly favourable isotherm of methane, two approximation methods were proposed to reconcile the present equilibrium theory model based on linear isotherms with the highly non-linear isotherm of the system concerned. One is the fixed beta case where the methane isotherm is approximated by a straight line over the entire range of zero to 1 of the mole fraction and the β_i is computed based on the slope of the linear fit that is presented in Table 2. The other is the secant beta case where the secant lines of the isotherm are found at each mole fraction, thus the slopes of the secant lines vary with the mole fraction. Refer to Figure 2 to see the differences of the two approximation methods.

3.2. Optimising Co-current Depressurisation (CoD) step

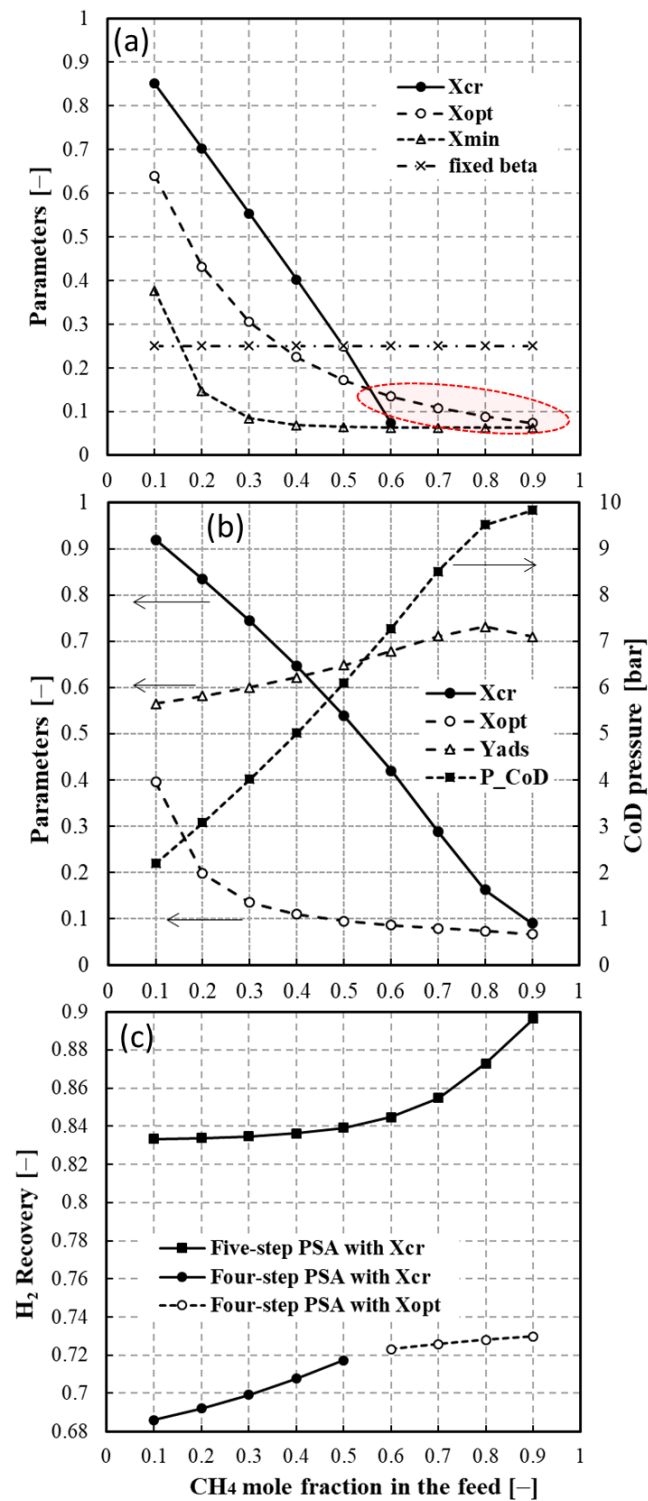


Figure 3. Critical, optimum and minimum extents of purging for (a) a four-step PSA, (b) a five-step PSA with co-current depressurisation step and (c) comparison of the H₂ product recovery between the 4-step and 5-step PSAs at varying feed composition (fixed beta case). P_H = 14.8 bar and P_L = 1.36 bar for all cases.

By solving the equilibrium theory model, we estimated firstly the maximum H₂ recovery achievable in the 4-step PSA consisting of feed, blowdown, purge and light product pressurisation. Then it was examined how much the H₂ recovery could be improved by adding the co-current depressurisation step to the step sequence. In the reference study¹³, the H₂ purification PSA system was operated between the adsorption pressure of 14.8 bar and the desorption pressure of 1.36 bar. With the operating pressures fixed for all the cases, the binary gas composition of the PSA feed was varied from 0.1 to 0.9 with the 0.1 interval. In the first place, the equilibrium theory model was solved with the fixed beta regardless of the feed composition and the calculation results are presented in Figure 3. As shown in Figure 3(a), in case of the four-step PSA, the critical extents of purging are greater than the optimal extents of purging in the methane mole fraction range of 0.1 to 0.5, thus the critical extent of purging were chosen to estimate the hydrogen recovery. At $y_f = 0.6$, the optimum extent of purging was chosen because it is greater than the critical extent of purging. At y_f being equal to or greater than 0.7, a perfect shock wave would be formed regardless of the extent of purging, thus the optimum extents of purging were chosen to estimate the recovery. The resulting hydrogen recovery lies in the range of 0.68 to 0.73 as shown in Figure 3(c).

In designing a PSA with co-current depressurisation (CoD), the pressure that the column reaches at the end of the CoD step can be determined freely in the range of adsorption pressure to desorption pressure at the operator's discretion. The PSA's performance is highly affected by the choice of the CoD step's final pressure. Accordingly, it is so important to find out the optimum CoD's final pressure that enables us to maximise the light component product recovery. As can be seen in Figure 3(b), the optimum CoD end pressure varies greatly depending on the feed composition. The larger methane mole fraction a feed gas has, the higher does the CoD's final pressure have to be set. This is because depressurisation of the column would not increase much the methane mole fraction in the gas phase if the methane mole fraction in the feed was already so high. In the 5-step PSA, the critical extent of purging were chosen for maximum recovery over the entire range of feed composition. The location of

a shock wave at the end of adsorption step was adjusted to a value below the column length, considering the distance it would travel during the subsequent CoD step. The estimated hydrogen product recovery of the 5-step PSA ranged from 0.83 to 0.90. As a result, the hydrogen product recovery is expected to increase drastically simply by adding the CoD step between adsorption and blowdown steps as shown in Figure 3c.

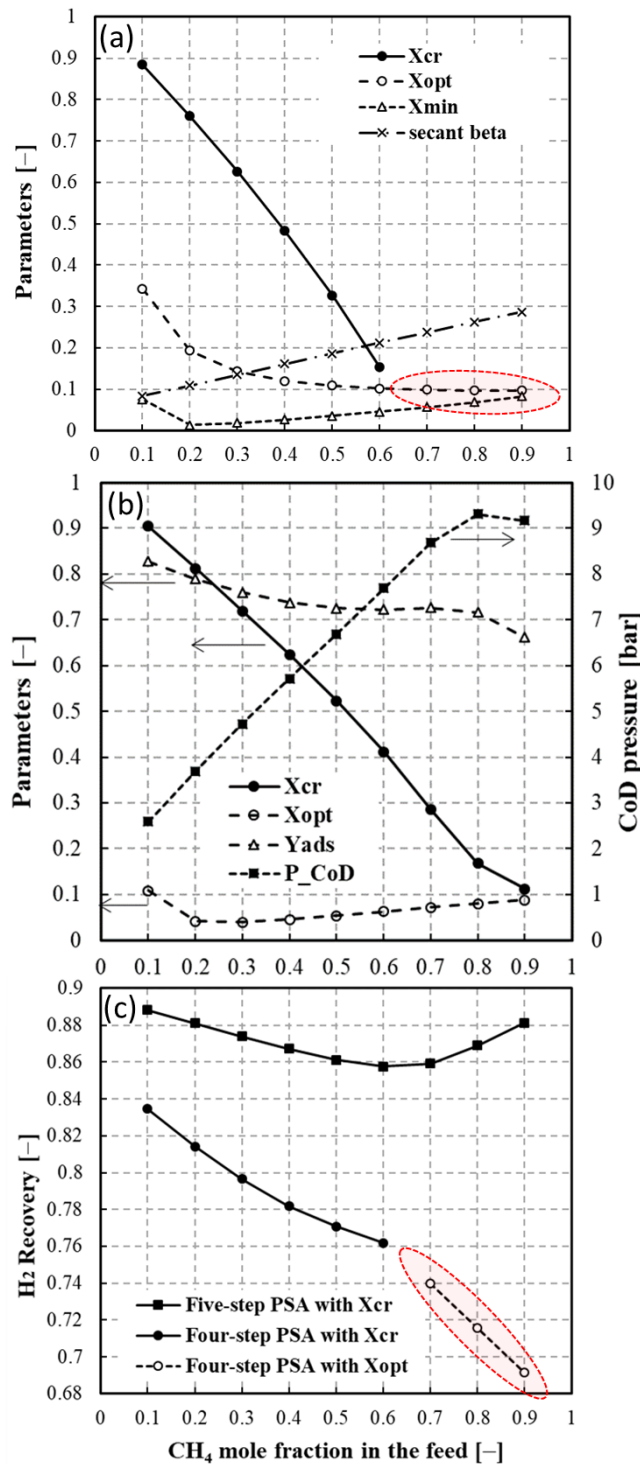


Figure 4. Critical, optimum and minimum extents of purging for (a) a four-step PSA and (b) a five-step PSA with co-current depressurisation step and (c) comparison of H₂ product recovery between the 4-step and 5-step PSAs at varying feed composition (secant beta case). P_H = 14.8 bar and P_L = 1.36 bar for all cases.

The fixed beta case in Figure 3 was computed with the slope of the single linear isotherm that is the best approximation of the non-linear CH₄ isotherm over the entire range of the PSA operating pressure. As shown in Figure 2, however, the actual methane isotherm deviates so significantly from the straight line, which indicates that the fixed beta model may not be good enough to predict the PSA performance.

To address this issue, the secant beta model was proposed. In this case, the methane isotherm was still linear but it was approximated to the secant line of the isotherm connecting zero to the equilibrium adsorption amount at the methane partial pressure in the feed. Therefore, the slope of the secant line varies with the feed composition and accordingly so does β_A .

The calculated extents of purging of the secant beta case were very similar to those of the fixed beta case as shown in Figure 4a, except for the lowest methane composition in the feed where the optimum extent of purging becomes greater than the critical extent of purging. The lowest y_f at which the reversal occurred was 0.7 in the secant beta case, while it was 0.6 in the fixed beta case. This is because the two extents of purging were affected by the slope of methane isotherm. Given that the slope of the secant line at $y_f = 0.6$ is larger than the slope of the linear fit, the secant beta case must have a larger critical extent of purging than the fixed beta case. The resulting critical extent of purging was larger than the optimum extent of purging in case of the secant beta case, while it was not in the fixed beta case (see Figures 3a and 4a). The effect of isotherm slope on X_{cr} can be explained by the equilibrium theory model. The greater slope of isotherm leads to the less amount of heavy component remaining in the column at the end of purge step, thus it takes longer to see a perfect shock wave form

during the feed step. As a result, the column has to be regenerated more rigorously to keep the heavy component from breaking through the column before the shock wave is formed.

The most striking difference between the fixed and secant beta cases is the trends of the product recovery changing with the feed composition. In case of the fixed bed case, the results in Figure 3 exhibits the effect of feed mole fraction very well, indicating that the greater heavy component fraction in the feed makes it easier to form a shock wave, requiring less critical extent of purging and resulting in the light component recovery being greater. In contrast, both feed gas composition and slope of isotherm come into play in the secant beta case. Given the trends of the decreasing H₂ recovery with increasing CH₄ fraction in Figure 4c, it appears that the effect of reducing isotherm slope that is typical of Langmuir isotherm is more dominant than the effect of feed composition.

All in all, substantial enhancement of the H₂ recovery resulting from adding the CoD step was captured very well in both fixed and secant beta methods, but a caveat is that the fixed beta method may not be relevant if the adsorption isotherm deviates greatly from linearity.

3.3. Optimising the number of Pressure Equalisation (PE) steps

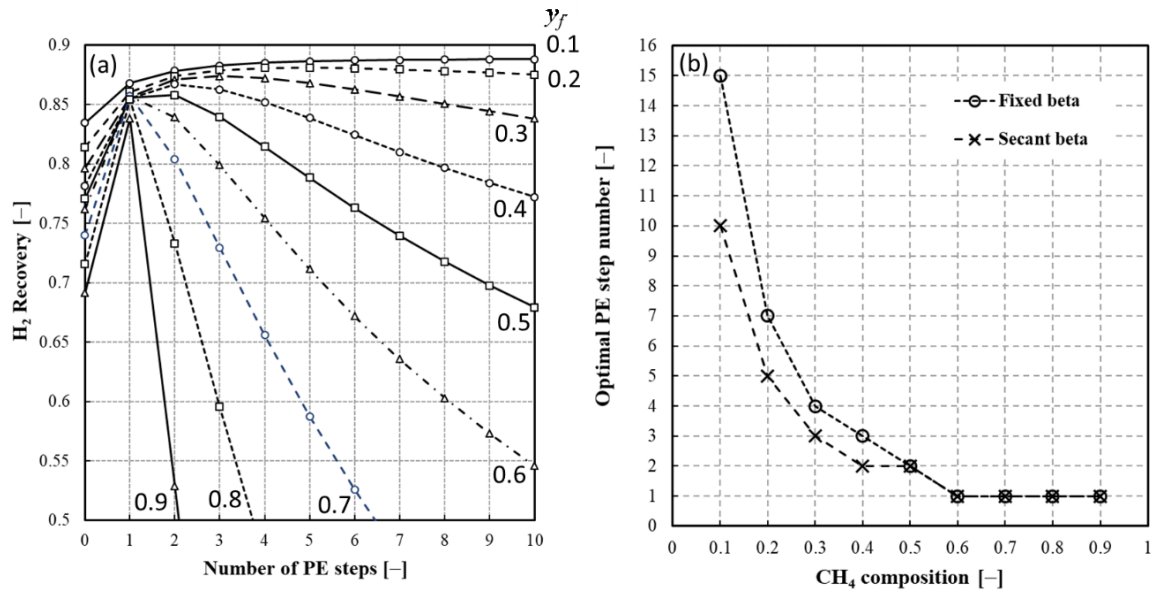


Figure 5. Optimising the number of pressure equalisation steps of a H₂ purification PSA cycle with respect to the H₂ recovery: (a) Effects of the number of pressure equalisation step on the H₂ recovery at varying feed mole fractions (secant beta case) and (b) the resulting optimal pressure equalisation step number in either fixed or secant beta case. $P_H = 14.8$ bar and $P_L = 1.36$ bar for all cases.

The light product recovery can also be improved by adding pressure equalisation steps, instead of co-current depressurisation step, to the step sequence of a PSA cycle. Since the extent of purging affects the hydrogen recovery greatly, it has to be optimised for maximising the product recovery. The strategy for finding the optimum number of PE steps is to check the effect of the number of PE steps on the maximum product recovery of the PSA system..

Addition of PE steps requires Y_{ads} to decrease, resulting in X_{cr} being increased accordingly. In this case, even one DPE step gives rise to Y_{ads} decreasing so significantly that it turns out that X_{cr} is always larger than X_{opt} for every PSA containing PE case. Therefore X_{cr} was chosen to estimate the H₂ recovery for each PSA containing PE step(s) in this study.

As shown in Figure 5, the optimised number of PE steps was affected greatly by the feed composition. The lower heavy component fraction the feed contains, the more PE steps are required to maximise the light product recovery. For example, the optimised number of PE steps was estimated to be 10 at $y_f = 0.1$ while it was 1 if y_f is equal to or greater than 0.6. This is because the DPE step would not be able to increase the heavy component mole fraction much if y_f was already high, and as a result the PE could not compensate the adverse effect resulting from the shorter feed step that addition of PE step(s) induced.

It is not uncommon to see researchers claim that increasing the PE steps would always be beneficial for improving the product recovery and nevertheless they decide not to increase the number of PE stages indefinitely simply because it would require too many columns to accommodate such a high number of PE steps in the process of PSA cycle scheduling. The results of Figure 5 implies that adding the PE steps does not always guarantee increasing the hydrogen product recovery against the stereotype conceived. In case of a hydrogen purification PSA working for a reforming-based hydrogen production process^{3,22}, the hydrogen mole fraction in the feed lies in the range of 0.7 – 0.9. Under the conditions of the light component fraction being so high, at least three PE steps would be able to enhance the hydrogen product recovery as shown in Figure 5b. In the stark contrast, only 1 PE step would be advisable for improving the H₂ recovery in case of the hydrogen fraction in the feed being so low. Having more than one PE steps would deteriorate the product recovery.

It should be noted that the effect of adding PE steps is basically the same as the effect of adding a CoD step. Accordingly the final column pressure at the last PE step must be very close to the final column pressure at the CoD step when they are compared under the optimised operating conditions. But the two final column pressures cannot be the same as each other because the column pressure can be adjusted minutely during the CoD step while it cannot but change stepwisely given the number of PE steps. In this respect, the H₂ recovery achievable by the optimised CoD pressure must be always larger than the product recovery

that a PSA can attain through optimising the number of PE steps, even though the difference is negligible.

3.4. Comparison with numerical simulation results

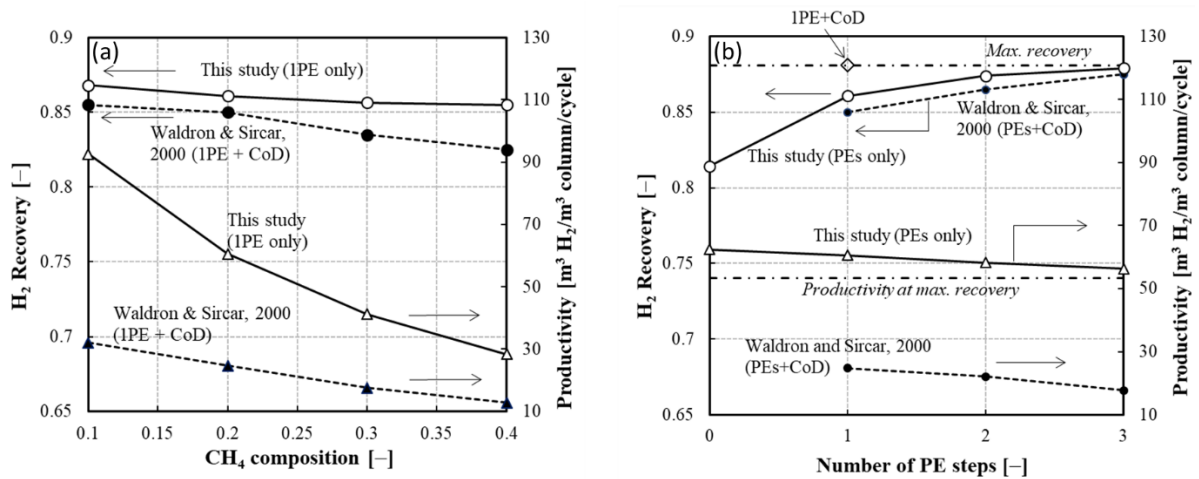


Figure 6. Comparison of the equilibrium theory results of this study with the corresponding numerical simulation results in the reference¹³ with respect to H₂ recovery and bed productivity: (a) effect of the feed composition and (b) effect of the number of PE steps with the CH₄ mole fraction in the feed fixed at 0.2. P_H = 14.8 bar and P_L = 1.36 bar for all cases.

In Figure 6, the equilibrium theory analysis results are compared with the numerical simulation results presented in the reference¹³. We must take heed not to compare the two results directly, because the reference work considered step sequences where both PE and CoD steps exist, and it is also not clear that the PSA operating conditions of the reference study were optimised for maximum H₂ recovery. That said, it is worth comparing the trends of the two results when either the feed composition or the number of PE steps are varied. In Figure 6a, the PSA with one PE step cases are compared with the CH₄ composition in the feed varying from 0.1 to 0.4. The trends of H₂ recovery decreasing rather than increasing with increasing CH₄ composition in the feed were consistent with the H₂ recovery that the equilibrium theory model using secant

beta estimated. This comparison corroborates that secant beta model is more accurate than the fixed beta model.

In Figure 6b, the theoretical and numerical results are compared with each other at the PSA where the numbers of PE steps are 1, 2 or 3 with the CH₄ mole fraction in the feed fixed at 0.2. The trends of the H₂ recovery increasing with the number of PE steps were observed in both theoretical and numerical results. It is shown that the theoretical H₂ recovery converges to its maximum value that would be reached by the five stage PSA with its CoD pressure optimised. It is notable that the bed productivity is reduced so significantly by addition of PE steps. This is because the feed step had to be finished early considering the distance of the shock wave travelling during the DPE steps, which means that less amount of fresh feed would be processed in each cycle.

3.5. Effect of adsorption pressure

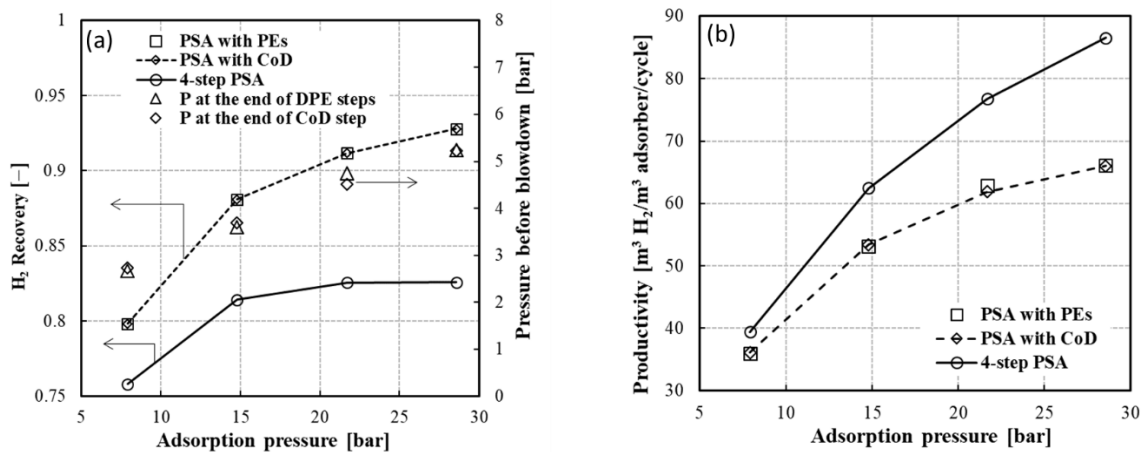


Figure 7. Effect of adsorption pressure on the two H₂/CH₄ separation PSAs with or without CoD (or PEs) with respect to (a) H₂ recovery and (b) bed productivity. P_L = 1.36 bar and y_f = 0.2.

One of the most conventional strategies we can take to improve the product recovery of a PSA is to elevate the adsorption pressure. The effects of adsorption pressure on the PSA performance were estimated very well by solving the equilibrium theory model as shown in Figure 7a. Substantial improvement of H₂ recovery resulting from either CoD or PE was shown clearly over the wide range of adsorption pressures investigated. Accordingly, the addition of either CoD or PE makes the bed productivity worse due to the feed step terminating earlier.

It should be noted that the two column pressures at the end of the CoD and last PE steps are very close to each other, as the corresponding PSA cases are both optimised for maximum H₂ recovery. As a result, the H₂ recovery and bed productivity values estimated by the two PSA cases are almost identical. Therefore it would not matter which method between PE and CoD was taken to enhance the product recovery if there was no restriction on the number of PE steps.

3.6. Light Product Pressurisation (LPP) and Feed Pressurisation (FP)

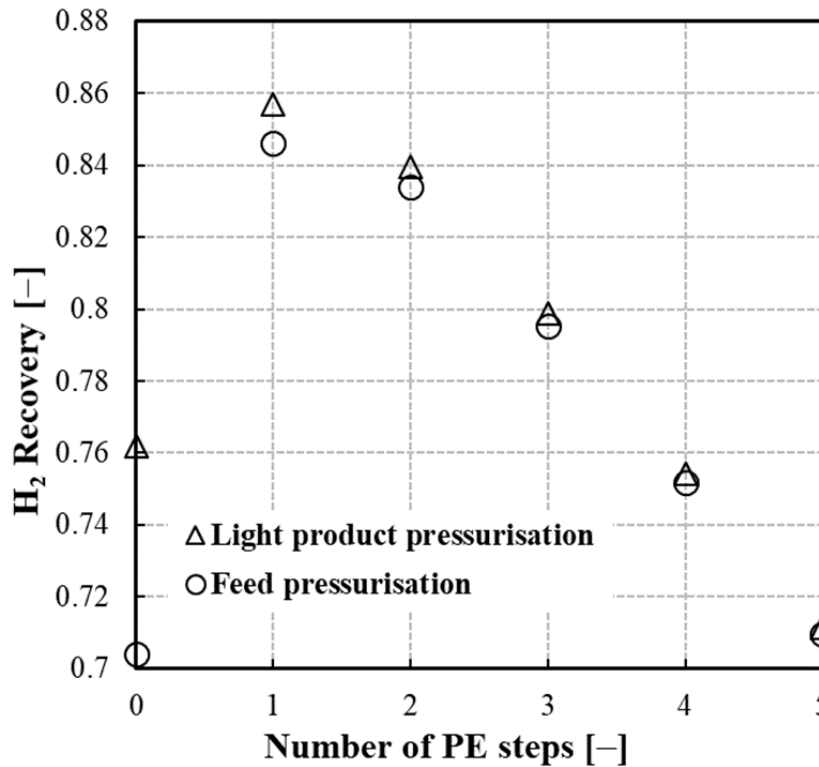


Figure 8. Comparison of H₂ recovery between light product pressurisation (LPP) and feed pressurisation (FP) of a H₂ purification PSA at varying number of PE steps. $P_H = 14.8$ bar, $P_L = 1.36$ bar and $y_f = 0.6$.

The equilibrium theory model used in this study is not only applicable to light product pressurisation but also feed pressurisation. The two different pressurisation methods were compared by solving the equilibrium theory model for the 4-step PSA under the same operation conditions and feed composition. It turned out that the LPP case would be able to give greater H₂ recovery than the FP case as shown in Figure 8. The similar results were reported in the past research²³ where they compared the LPP and FP cases by running a numerical simulator combined with an optimising algorithm. Under the conditions of the H₂ purity close to 100%, it was reported that the LPP case would provide greater H₂ recovery than the FP case. It is interesting that this theoretical research led us to the same conclusion as what the highly sophisticated optimisation work resulted in.

While the difference of H₂ recovery between the LPP and FP cases is vast in case of the 4-step PSA, it get diminished notably with pressure equalisation stages added. In case of a PSA with five pressure equalisation stages, the two H₂ recovery values estimated by the LPP and FP cases are almost identical to each other. This is because, in case of FP case, incorporating more PE steps into the step sequence incurs the extent of backfill, Z, to get increased, in other words, the effect of feed pressurisation gets smaller.

4. Conclusions

This study was initiated to answer the question: is there an optimal number of the pressure equalisation steps for maximising the PSA product recovery and if an optimised number of PE steps exists, how can we find it? In this study, an equilibrium theory model developed by Chiang¹⁶ was taken to estimate the product recovery and the bed productivity of the step sequences concerned. The results of this study elucidated that there exists an optimal number of pressure equalisation steps for maximising the product recovery and it turned out that the optimal number of pressure equalisation steps can be very different depending on the feed composition. Co-current depressurisation can also be taken instead of pressure equalisation with a view to enhancing the product recovery. The column pressure at the end of co-current depressurisation step has to be optimised for the maximum product recovery in a fashion similar to searching for optimal number of pressure equalisation steps.

If the isotherm is highly non-linear, it would be recommended using the secant of isotherm, rather than linear fit of the isotherm, in solving the equilibrium theory model that was developed for an adsorption system with linear adsorption isotherms. The Equilibrium theory model of a PSA system turned out to be an excellent tool to understand the effects of adsorption pressure and column pressurisation methods (light product pressurisation and feed pressurisation). In the end, the equilibrium theory analysis was able to produce the PSA performances that was

qualitatively consistent with the results obtained by numerical simulation combined with an optimiser.

Notations

A_{cs} = cross-sectional area of adsorption column, m²

b_i = Langmuir isotherm parameter of component i, 1/bar

k_i = Henry constant, -

L = length of adsorption column, m

N_{BF} = total number of moles of light component consumed for pressurising the column in a cycle, mole

N_{CoD} = total number of moles of light component product obtained during the CoD step, mole

N_{DPE} = total number of moles of light component product obtained during all DPE steps, mole

N_H = total number of moles of feed gas entering the PSA system in a cycle, mole

N_{HP} = total number of moles of light component product obtained during the feed step, mole

N_{pg} = total number of moles of light component consumed as a purge gas during the purge step, mole

P = pressure, bar

P_{CoD} = pressure at the end of cocurrent depressurisation step, bar

P_{E_i} = pressure at the end of i th DPE step, bar

P_H = pressure of feed step, bar

P_L = pressure of purge step, bar

q_0 = saturated adsorption capacity of Langmuir isotherm, mol/kg

R = light component product recovery, -

T = throughput of a cycle, total number of moles of the net light component product in one cycle, mole

u = interstitial gas velocity, m/s

u_s = velocity of shock wave, m/s

y = gas mole fraction, -

y_b = heavy component mole fraction at the end of blowdown step, -

y_{E_i} = heavy component mole fraction behind the shock wave at the end of i th DPE step, -

y_f = heavy component mole fraction in the feed, -

X = extent of purging, -

X_{cr} = critical extent of purging, -

X_{min} = minimum extent of purging, -

X_{opt} = optimum extent of purging, -

Y_s = dimensionless position of shock wave the moment that a shock wave develops into a perfect shock wave during the feed step, -

Y_{ads} = dimensionless position of shock wave at the end of adsorption step, -

Z = extent of backfill, -

z = axial position of adsorption column with $z = 0$ at the feed end, m

z_f = axial position of the heavy component concentration front at $y = 0$ at the beginning of feed step, m

Greek symbols

ε = column void fraction. -

ρ = adsorbent particle density, kg/m³

$$\phi = \frac{\varepsilon A_{cs} L P_L}{\beta_A R T}$$

$$\psi_H = P_H / P_L$$

$$\psi_{Ei} = P_{Ei} / P_L$$

$$\psi_{CoD} = P_{CoD} / P_L$$

Abbreviations

CoD: co-current depressurisation

DPE: depressurising pressure equalisation

PE: pressure equalisation

PPE: pressurising pressure equalisation

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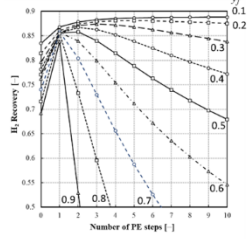
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Generic PSA cycle: Feed \rightarrow DPE(s) \rightarrow CoD \rightarrow BD \rightarrow Purge \rightarrow PPE(s) \rightarrow Pressurisation

Optimising a H₂ PSA for maximising the light product recovery by adjusting:

Number of pressure equalisation steps



Co-current depressurisation pressure

