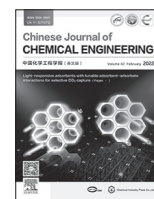




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Full Length Article

Comparative study on pressure swing adsorption system for industrial hydrogen and fuel cell hydrogen

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ARTICLE INFO

Article history:

Received 11 May 2021

Received in revised form 29 September 2021

Accepted 29 September 2021

Available online 25 October 2021

Keywords:

Pressure swing adsorption

Hydrogen

Fuel cell hydrogen

Industrial hydrogen

Numerical simulation

ABSTRACT

In order to improve the design of PSA system for fuel cell hydrogen production, a non-isothermal model of eight-bed PSA hydrogen process with five-component ($\text{H}_2/\text{N}_2/\text{CH}_4/\text{CO}/\text{CO}_2 = 74.59\%/0.01\%/4.2\%/2.5\%/18.7\%$ (vol)) four-stage pressure equalization was developed in this article. The model adopts a composite adsorption bed of activated carbon and zeolite 5A. In this article, pressure variation, temperature field and separation performance are stimulated, and also effect of providing purge (PP) differential pressure and the ratio of activated carbon to zeolite 5A on separation performance in the process of producing industrial hydrogen (CO content in hydrogen is $10 \mu\text{L}\cdot\text{L}^{-1}$) and fuel cell hydrogen (CO content is $0.2 \mu\text{L}\cdot\text{L}^{-1}$) are compared. The results show that Run 3, when the CO content in hydrogen is $10 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery is 89.8%, and the average flow rate of feed gas is $0.529 \text{ mol}\cdot\text{s}^{-1}$; When the CO content in hydrogen is $0.2 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery is 85.2%, and the average flow rate of feed gas is $0.43 \text{ mol}\cdot\text{s}^{-1}$. With the increase of PP differential pressure, hydrogen recovery first increases and then decreases, reaching the maximum when PP differential pressure is 0.263 MPa; With the decrease of the ratio of activated carbon to zeolite 5A, the hydrogen recovery increases gradually. When the CO content in hydrogen is $0.2 \mu\text{L}\cdot\text{L}^{-1}$ the hydrogen recovery increases more obviously, from 83.96% to 86.37%, until the ratio of activated carbon to zeolite 5A decreases to 1. At the end of PP step, no large amount of CO_2 in gas or solid phase enters the zeolite 5A adsorption bed, while when the CO content in hydrogen is $10 \mu\text{L}\cdot\text{L}^{-1}$, and the ratio of carbon to zeolite 5A is less than 1.4, more CO_2 will enter the zeolite 5A bed.

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1. Introduction

Pressure swing adsorption (PSA) technology is a kind of gas separation technology, which makes use of the characteristics that the adsorption capacity of adsorbent for hydrogen is weaker than that of other gases, and the adsorption capacity for gases increases with the increase of pressure and decreases with the decrease of pressure. Compared with low temperature method and membrane separation, PSA technology has the advantages of high purity of product gas, wide adaptability of feed gas, high degree of automation and flexible operation [1,2]. PSA hydrogen purification technology is widely used in the purification of various H_2 -containing gases. The purity of hydrogen can reach 99.9–99.9999 % (vol), and the trace impurities in hydrogen can meet the requirements of various chemical reactions and hydrogen for proton exchange membrane fuel cell [3]. In order to adapt to different applications and improve the efficiency and performance of the system, researchers

have carried out in-depth research on the mathematical model [4–6], process [7–9], adsorbent [10–13], gas distribution technology [14] of PSA. With the development of computer technology and the improvement of PSA mathematical model, more and more researchers use numerical simulation to study on PSA process [15–25]. Moon *et al.* [26] simulated a PSA process for hydrogen recovery from H_2 -rich gas. Firstly, he compares four- and eight-bed process, finding that eight-bed process has more pressure equalization step, and the hydrogen recovery is higher than that of four-bed process. On the basis, the separation performance of eight-bed process with three different kinds of purge gas, *i.e.* product gas, remaining gas after the first pressure equalization step, and remaining gas after the last pressure equalization step, was compared. The results show that when the hydrogen purity is greater than 99.99% (vol), the recovery is increased by 3%–6% using remaining gas after the first equalization depressurization step as purge gas than using product gas; When the hydrogen purity is 99.99% (vol), using remaining gas after the last equalization depressurization step as purge gas, the highest recovery is 89.7%. Niu *et al.* [27] simulated and studied the six-bed PSA hydrogen process with

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four-components ($H_2/CH_4/CO/CO_2 = 76\%/3.5\%/0.5\%/20\%$ (vol)) in three-stage pressure equalization twelve-step. He also compared the separation effect of rPSA and PSA. Under conditions that adsorption at 2.2 MPa, desorption at 1.0 MPa, volume ratio of activated carbon to zeolite 5A is 1:1, hydrogen purity is 99.9 % (vol), recovery of rPSA is 69.88%, dealing capacity is $0.8875 \text{ mol}\cdot\text{s}^{-1}$, and H_2 yield is $0.4713 \text{ mol}\cdot\text{s}^{-1}$; while the recovery of PSA is 83.04%, dealing capacity is $0.39 \text{ mol}\cdot\text{s}^{-1}$, and H_2 yield is $0.2472 \text{ mol}\cdot\text{s}^{-1}$.

As a clean secondary energy, hydrogen energy has developed rapidly in recent years. The market demand for fuel cell hydrogen is increasing, so more PSA devices are needed to produce fuel cell hydrogen. The content of CO in product hydrogen is generally required to be $10 \mu\text{L}\cdot\text{L}^{-1}$ for conventional industrial hydrogen, while that for fuel cell hydrogen is $0.2 \mu\text{L}\cdot\text{L}^{-1}$. That is to say, hydrogen for fuel cell has more stringent requirements on trace impurities. Therefore, more study should be taken on the difference between fuel cell hydrogen PSA and conventional industrial hydrogen, to optimize the engineering design and unit performance. Based on this, a mathematical model of eight-bed PSA hydrogen process with 5-component ($H_2/N_2/CH_4/CO/CO_2 = 74.59\%/0.01\%/4.2\%/2.5\%/18.7\%$ (vol)), four-stage pressure equalization was established. The model adopts composite adsorption bed with activated carbon and zeolite 5A. The performance differences between industrial hydrogen (CO content $10 \mu\text{L}\cdot\text{L}^{-1}$) and fuel cell hydrogen (CO content $0.2 \mu\text{L}\cdot\text{L}^{-1}$) production under the same conditions were compared. The model also evaluates the effect of PP differential pressure and ratio of activated carbon to zeolite 5A on the separation performance.

2. Process Flow

This article evaluates a 8-1-4/P PSA hydrogen process, that is eight adsorbers, one feeding vessel, four-stage pressure equalization process. Each adsorber will successively go through the following sixteen steps: adsorption(A), equalization 1 depressurization(E1D), equalization 2 depressurization(E2D), equalization 3 depressurization(E3D), equalization 4 depressurization(E4D), providing purge 1(PP1), providing purge 2(PP2), dump (D), purge 1(P1), purge 2(P2), equalization 4 repressurization (E4R), equalization 3 repressurization(E3R), equalization 2 repressurization(E2R), equalization 1 repressurization(E1R), final repressurization(FR). Each step takes 50 s and each cycle takes 800 seconds. The process flow is shown in Fig. 1. A product gas valve, E1/E2 valve, E3/E4 valve, PP valve and inlet P valve are set at the outlet of the adsorber; and a feed gas valve, dump (D) valve, purging (P) outlet valve and final repressurization (FR) valve are set at the inlet of the adsorber. The operation process of each adsorber is shown in Fig. 2, and the sequence of the whole process is shown in Table 1.

3. Mathematical Model

To develop a mathematical simulation of PSA hydrogen process, a non-isothermal dynamic model is selected. In this study, the following assumptions were adopted: (i) the gas conforms to the ideal gas property; (ii) ignore the gradient of temperature, concentration, pressure and adsorption capacity in radial direction; (iii) the adsorption kinetic model uses the linear driving force (LDF) model; (iv) Ergun equation is used for momentum balance; (v) the adsorption equilibrium uses loading ratio correlation (LRC) equation; (vi) the heat balance between gas and solid phase is achieved quickly. The mathematical equation is as follows [28,29]:

The component mass balance:

$$\frac{\partial y_i}{\partial t} + \frac{RT}{P} \rho_s \left(\frac{1-\varepsilon}{\varepsilon} \right) \frac{\partial q_i}{\partial t} + \frac{\partial (uy_i)}{\partial z} - D_{ax} \frac{\partial^2 y_i}{\partial z^2} = 0 \quad (1)$$

The overall mass balance:

$$\frac{RT}{P} \rho_s \left(\frac{1-\varepsilon}{\varepsilon} \right) \frac{\partial q_i}{\partial t} + \frac{\partial (u)}{\partial z} = 0 \quad (2)$$

The energy balance for gas and solid phases:

$$\begin{aligned} & \varepsilon \rho_g c_{pg} + (1-\varepsilon) \rho_s c_{ps} \left(\frac{\partial T}{\partial t} + \varepsilon \rho_g c_{pg} \frac{\partial (uT)}{\partial z} \right. \\ & \left. - K_z \frac{\partial^2 T}{\partial z^2} - (1-\varepsilon) \rho_s \sum_{i=1}^n \left(\Delta H_i \frac{\partial q_i}{\partial t} \right) + \frac{2h_i}{R_i} (T - T_w) \right) = 0 \end{aligned} \quad (3)$$

The energy balance for wall:

$$\rho_w c_{pw} A_w \frac{\partial T_w}{\partial t} = 2\pi R_i h_i (T - T_w) - 2\pi R_o h_o (T_w - T_{atm}) \quad (4a)$$

$$A_w = \pi (R_o^2 - R_i^2) \quad (4b)$$

The momentum balance (Ergun eqn):

$$-\frac{dp}{dz} = \frac{1.5 \times 10^{-3} (1-\varepsilon)^2}{(2r_p \psi)^2 \varepsilon^3} \mu v + 1.75 \times 10^{-5} \rho_g \frac{(1-\varepsilon)}{2r_p \psi \varepsilon^3} |v| v \quad (5)$$

The multi-component adsorption equilibrium (loading ratio correlation (LRC) model):

$$q_i^* = \frac{(IP_{1,i} - IP_{2,i} T) IP_{3,i} \exp^{IP_{4,i}/T P_i}}{1 + \sum_{j=1}^n IP_{3,j} \exp^{IP_{4,j}/T P_j}} \quad (6)$$

Mass transfer (linear driving force (LDF) model):

$$\frac{\partial q_i}{\partial t} = k_{LDF,i} (q_i^* - q_i) \quad (7)$$

The feed gas for the simulation uses five-component ($H_2/N_2/CH_4/CO/CO_2 = 74.59\%/0.01\%/4.2\%/2.5\%/18.7\%$ (vol)) hydrogen-containing gas, under pressure of 2.6 MPa, temperature of 298.15 K, desorption pressure of 0.12 MPa. A composite adsorption bed of activated carbon (AC) and zeolite 5A (5A) was used, in which zeolite 5A was located in the upper part of the adsorption bed and activated carbon was located in the lower part. The parameters of adsorption bed are shown in Table 2. The length of adsorption bed is 2.4 m, and the diameter is 0.14 m. The adsorption isotherm parameters of components on adsorbent were obtained by adsorption isotherm data simulation. The adsorption heat value of each component gas is calculated by Clausius Clapeyron Eq. (8) [30]. The dynamic mass transfer coefficients of each component are calculated from the breakthrough curve data, and the specific values are shown in Table 3.

Clausius Clapeyron equation:

$$-\Delta H_i = RT \left(\frac{\partial \ln P_i}{\partial T} \right) \quad (8)$$

The calculation length is divided into 100 nodes along the axial direction of the adsorption bed. The partial differential equations are discretized by the upwind first-order difference method (uds1) with a time step of 1 s. In the simulation calculation, the object quality of product are two kinds, namely industrial hydrogen with CO content of $10 \mu\text{L}\cdot\text{L}^{-1}$ and fuel cell hydrogen with CO content $0.2 \mu\text{L}\cdot\text{L}^{-1}$. Under the condition of fixed adsorption bed parameters and process parameters, different quality of hydrogen can be produced by changing the flowrate of feed gas. In order to fully compare the difference of PSA performance when producing two kinds of hydrogen, evaluation were made on PP different pressure and ratio of activated carbon to zeolite 5A. The calculation conditions are shown in Table 4.

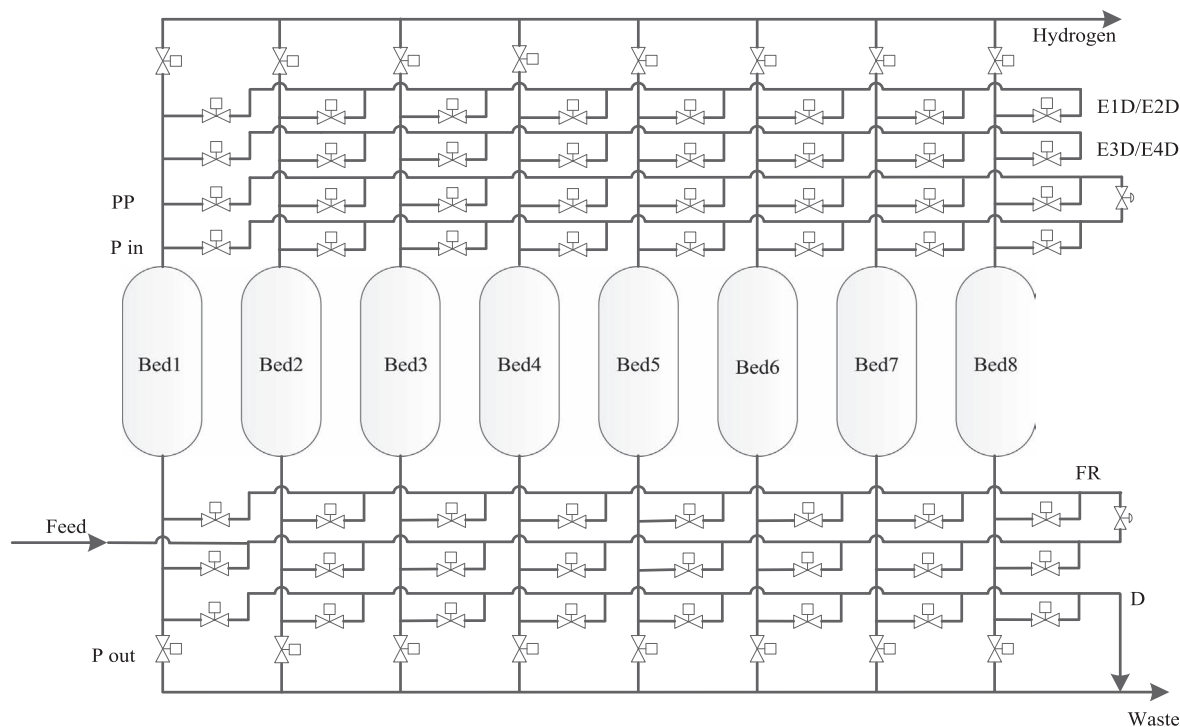


Fig. 1. Schematic diagram of the eight-bed PSA process.

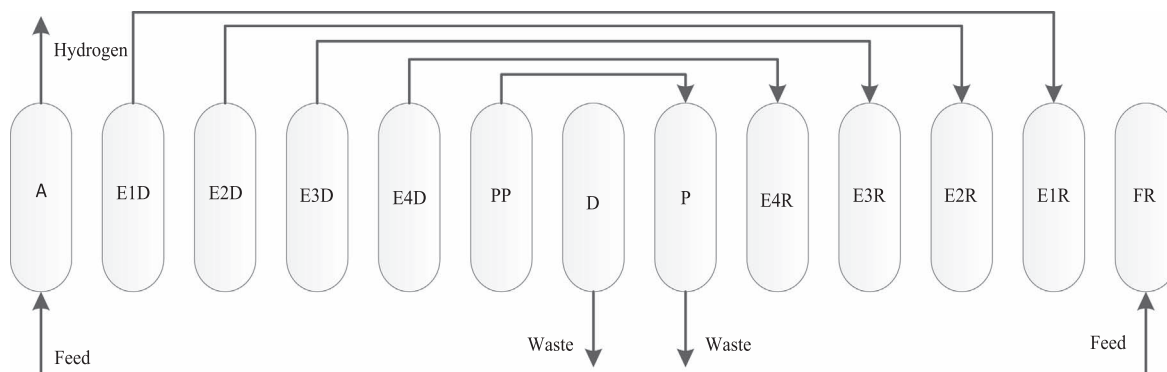


Fig. 2. Elementary steps of the PSA cycle.

4. Results and Discussion

4.1. Simulation results

In order to compare the PSA performance when producing different hydrogen products, researchers control the PP differential pressure through adjusting the valve flow in PP step, and controls the CO content in the product hydrogen through adjusting the feed flow in adsorption step.

The simulation results show that, in Run3, when CO content in product hydrogen is $10 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen purity is above 99.99%

(vol), recovery is 89.8%, the average feed flow is $0.529 \text{ mol}\cdot\text{s}^{-1}$, and treatment capacity per unit adsorbent is $1.433 \text{ kmol}\cdot\text{m}^{-3}$. The simulation results are consistent with the operation data from industrial units. When CO content in product hydrogen is $0.2 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen purity is above 99.999% (vol), recovery is 85.2%, the average feed flow is $0.43 \text{ mol}\cdot\text{s}^{-1}$, and treatment capacity per unit adsorbent is $1.165 \text{ kmol}\cdot\text{m}^{-3}$. That is, when the CO content decreases from $10 \mu\text{L}\cdot\text{L}^{-1}$ to $0.2 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery decreases by 4.6% and the average feed flow decreases by 18.7%. After circulation stabilized, the pressure curve in the adsorber is shown in Fig. 3. It can be seen from Fig. 3 that the when CO content

Table 1
Sequence of the 16-step PSA Cycle

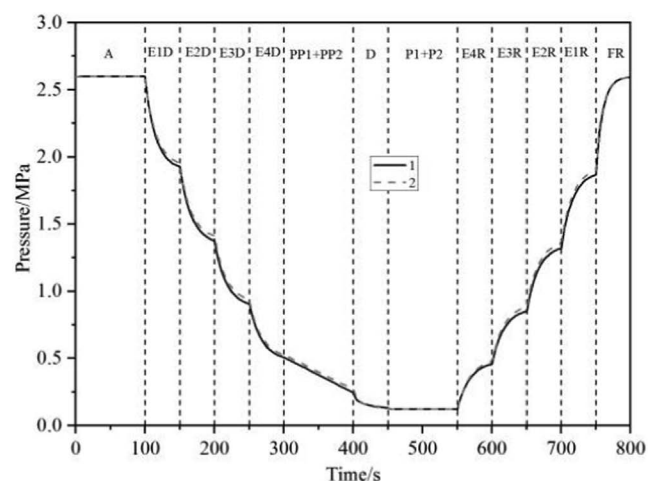
	Step															
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
Time/s	50	50	50	50	50	50	50	50	50	50	50	50	50	50	50	50
Bed	A	A	E1D	E2D	E3D	E4D	PP1	PP2	D	P1	P2	E4R	E3R	E2R	E1R	FR

Table 2
Characteristics of adsorbents and adsorption bed

Properties	Adsorbents	
	Activated Carbon	Zeolite 5A
r_p/m	0.0012	0.001
ψ	0.83	1
$\rho_B/kg \cdot m^{-3}$	630	750
ε	0.35	0.35
$c_{ps}/kJ \cdot kg^{-1} \cdot K^{-1}$	0.89	0.921
Adsorption bed		
L/m	2.4	
R_i/m	0.07	
R_o/m	0.073	
$\rho_w/kg \cdot m^{-3}$	7800	
$c_{pw}/kJ \cdot kg^{-1} \cdot K^{-1}$	0.5	
$h_i/kW \cdot m^{-2} \cdot K^{-1}$	0.03851	
$h_o/kW \cdot m^{-2} \cdot K^{-1}$	0.01423	

in product hydrogen is $0.2 \mu L \cdot L^{-1}$ pressure at the end of each pressure equalization is higher than that when CO content is $10 \mu L \cdot L^{-1}$. This is mainly due to the desorption and secondary adsorption of gas components in the adsorption bed during equalization depressurization step. Therefore, the differential pressure of equalization is not ideal, but it decreases with the increase of equalization. That is $\Delta P_{E4D} < \Delta P_{E3D} < \Delta P_{E2D} < \Delta P_{E1D}$. Also with the increase of impurities adsorbed in the adsorption bed, the differential pressure increases. As a result, when CO content in the product gas is low, the feeding capacity of the adsorption bed, the adsorbed impurities, and the differential pressure is reduced, while the pressure at the end of each equalization is higher.

In Run3, the temperature field in the adsorption bed and the temperature change curves at different positions are shown in Figs. 4 and 5. It can be seen from Figs. 4 and 5 that the temperature in the adsorption bed changes periodically with the PSA process,

**Fig. 3.** Pressure history during one cycle at Run3(1-CO:10 $\mu L \cdot L^{-1}$, 2-CO:0.2 $\mu L \cdot L^{-1}$).

and the temperature change in the lower part is much greater than that in the upper part of the adsorption bed. In the process of equalization depressurization and providing purge, the temperature in the lower part of the adsorption bed is decreasing, while that in the middle and lower part is increasing. In the process of dump and purge, the temperature below the middle part of adsorption bed decreases rapidly, which indicates that there are many gas desorption and secondary adsorption during equalization depressurization and providing purge. Besides, the endothermic capacity of desorption in the lower part of the adsorption bed is greater than the exothermic capacity of secondary adsorption, while the exothermic capacity of adsorption in the middle and lower part of the adsorption bed is greater than the endothermic

Table 3
LRC parameters and LDF coefficients for activated carbon and zeolite 5A

	H ₂	N ₂	CH ₄	CO	CO ₂
Activated Carbon					
$IP_1/kmol \cdot kg^{-1}$	-0.04327	0.014104	0.01759	0.016872	0.043169
$IP_2/kmol \cdot kg^{-1} \cdot K^{-1}$	-1.20×10^{-4}	3.61×10^{-5}	4.22×10^{-5}	4.39×10^{-5}	1.18×10^{-4}
IP_3/MPa^{-1}	-0.00334	0.12658	0.9344	0.65161	0.4706
IP_4/K	0.94667	517.173	157.22	81.1195	473.567
$\Delta H_i/kJ \cdot mol^{-1}$	7.85	15.71	18.73	16.66	25.54
K_{LDF}/s^{-1}	0.9	0.261	0.195	0.15	0.036
Zeolite 5A					
$IP_1/kmol \cdot kg^{-1}$	0.023095	0.010397	0.018391	0.008285	0.024742
$IP_2/kmol \cdot kg^{-1} \cdot K^{-1}$	6.32×10^{-5}	2.39×10^{-5}	4.87×10^{-5}	1.92×10^{-5}	6.62×10^{-5}
IP_3/MPa^{-1}	0.042	0.982	1.042	2.486	5.997
IP_4/K	4.74	66.881	145.587	271.919	501.839
$-\Delta H_i/kJ \cdot mol^{-1}$	8	18.5	19	29	50.69
K_{LDF}/s^{-1}	0.9	0.147	0.099	0.063	0.014

Table 4
Calculation conditions

Run no.	Adsorption pressure/MPa	Numbers of pressure equalization	$\Delta P^{(D)}/MPa$	AC to 5A ratio
Run1	2.6	4	0.203	1.4/1
Run2	2.6	4	0.233	1.4/1
Run3	2.6	4	0.263	1.4/1
Run4	2.6	4	0.308	1.4/1
Run5	2.6	4	0.263	1.6/0.8
Run6	2.6	4	0.263	1.5/0.9
Run7	2.6	4	0.263	1.4/1.0
Run8	2.6	4	0.263	1.3/1.1
Run9	2.6	4	0.263	1.2/1.2

① Differential pressure before and after PP step.

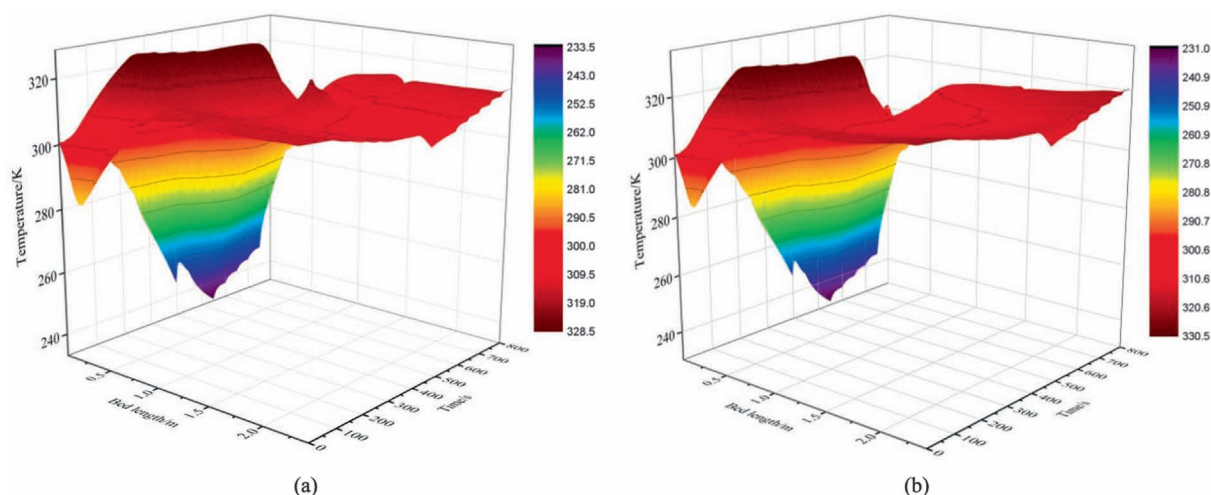


Fig. 4. Bed temperature field during one cycle at Run3 ((a):CO:10 $\mu\text{L}\cdot\text{L}^{-1}$, (b):CO:0.2 $\mu\text{L}\cdot\text{L}^{-1}$).

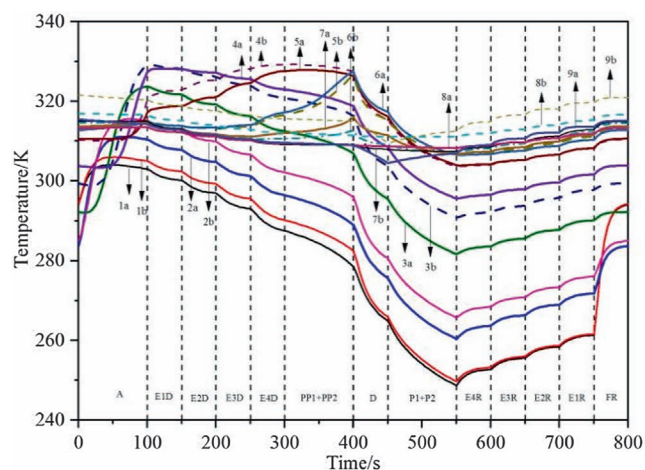


Fig. 5. Bed temperature curve at different positions during one cycle at Run3 (1–0.1 m, 2–0.2 m, 3–0.4 m, 4–0.6 m, 5–0.8 m, 6–1.0 m, 7–1.2 m, 8–1.8 m, 9–2.4 m, a–CO:10 $\mu\text{L}\cdot\text{L}^{-1}$, b–CO:0.2 $\mu\text{L}\cdot\text{L}^{-1}$).

mic capacity of desorption. In addition, it can be seen from Fig. 5 that when CO content of product hydrogen is low, the position where the temperature in the adsorption bed changes from decreasing to increasing is closer to the lower part of the adsorption bed.

4.2. The influence of PP differential pressure

After the equalization depressurization step, hydrogen with high purity still exists in the top part of the adsorption bed. Part of this hydrogen is used as purging gas for other adsorption beds in PP step, and other part is used as purging gas for the lower position of adsorption bed in D step. The gas flow in PP step depends on the differential pressure. The larger the PP differential pressure is, the larger the purging capacity is, and the more thorough the regeneration is. However, the larger the PP differential pressure, the farther the adsorption front moves to the outlet, and more purging gas will be needed for regeneration. Therefore, there is an optimal value of the PP differential pressure between. Fig. 6 shows the variation of hydrogen recovery and feed gas average flow with PP differential pressure. The recovery of hydrogen and the feed gas average flow rate increase first and then decrease with

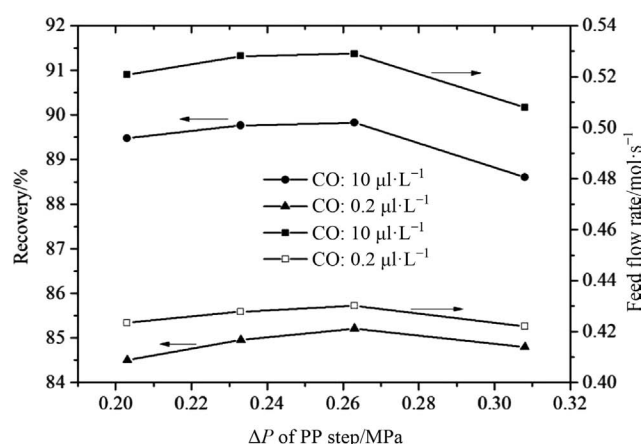


Fig. 6. Performance at different ΔP of PP step.

the increase of the PP differential pressure, and reach the maximum value when the PP differential pressure is 0.263 MPa.

In many researches on PSA process, the purge-to-feed ratio(P/F), i.e. the ratio of hydrogen volume in purging gas to that in feed gas, is regarded as an important parameter to evaluate the process. The providing purge capacity is also purging capacity. The PP differential pressure will directly affect the P/F. Fig. 7 shows the change of P/F with PP differential pressure. It can be seen from Fig. 7 that the P/F increases with the increase of the PP differential pressure. When the PP differential pressure is 0.263 MPa and CO in product gas is 10 $\mu\text{L}\cdot\text{L}^{-1}$ the corresponding optimal P/F is 9.12% and when CO is 0.2 $\mu\text{L}\cdot\text{L}^{-1}$ optimal P/F is 11.22% respectively. That is to say, the lower the CO content in the product, the greater the P/F is required.

4.3. Influence of AC-to-5A ratio

Different adsorbents have different adsorption capability for different impurities. The hydrogen purification process generally adopts composite adsorption bed, in which the heavy component is adsorbed at the lower part, and the light component at the upper part. According to the impurities composition, the appropriate proportion of adsorbent is selected to achieve the best separation effect.

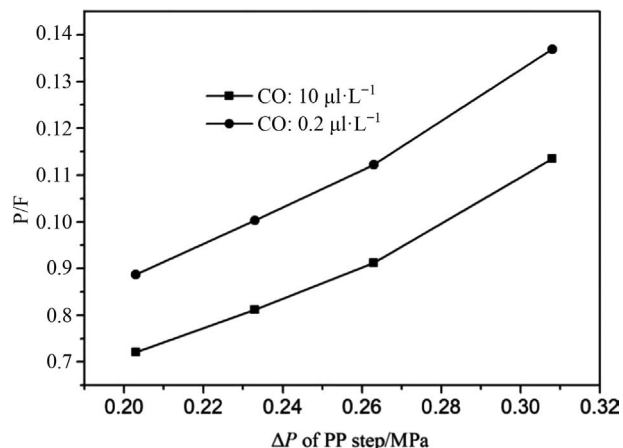
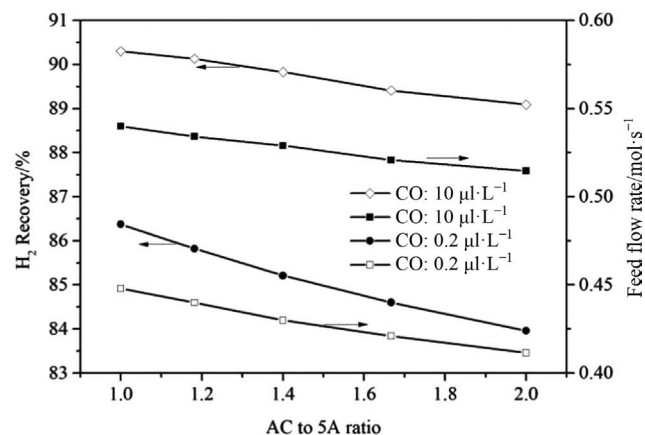
Fig. 7. P/F at different ΔP of PP step.

Fig. 8. Performance at different AC to 5A ratio.

The writer calculates the separation performance under different ratios of activated carbon to zeolite 5A. Fig. 8 shows the changes of hydrogen recovery and feed gas flow when the ratio of activated carbon to zeolite 5A changes from 1:1 to 2:1. When CO content in product gas is $10 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery increases from 89.09% to 90.3%, and the feed gas flow rate increases from $0.5146 \text{ mol}\cdot\text{s}^{-1}$ to $0.54 \text{ mol}\cdot\text{s}^{-1}$. When CO content in the product gas is $0.2 \mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery increases from 83.96% to 86.37%, and the feed gas flow increases from

$0.4115 \text{ mol}\cdot\text{s}^{-1}$ to $0.448 \text{ mol}\cdot\text{s}^{-1}$. That is to say, when CO content in the product gas is $0.2 \mu\text{L}\cdot\text{L}^{-1}$, increasing the proportion of zeolite 5A has a greater impact on the hydrogen recovery and the feed gas capacity, and reducing CO content in the product requires a larger proportion of zeolite 5A.

Fig. 9 shows the concentration distribution of CO and CO_2 in the gas phase, and the adsorption capacity distribution of CO and CO_2 in the solid phase at the end of the PP step. Fig. 9 shows that no matter the concentration front of CO and CO_2 in the gas phase or

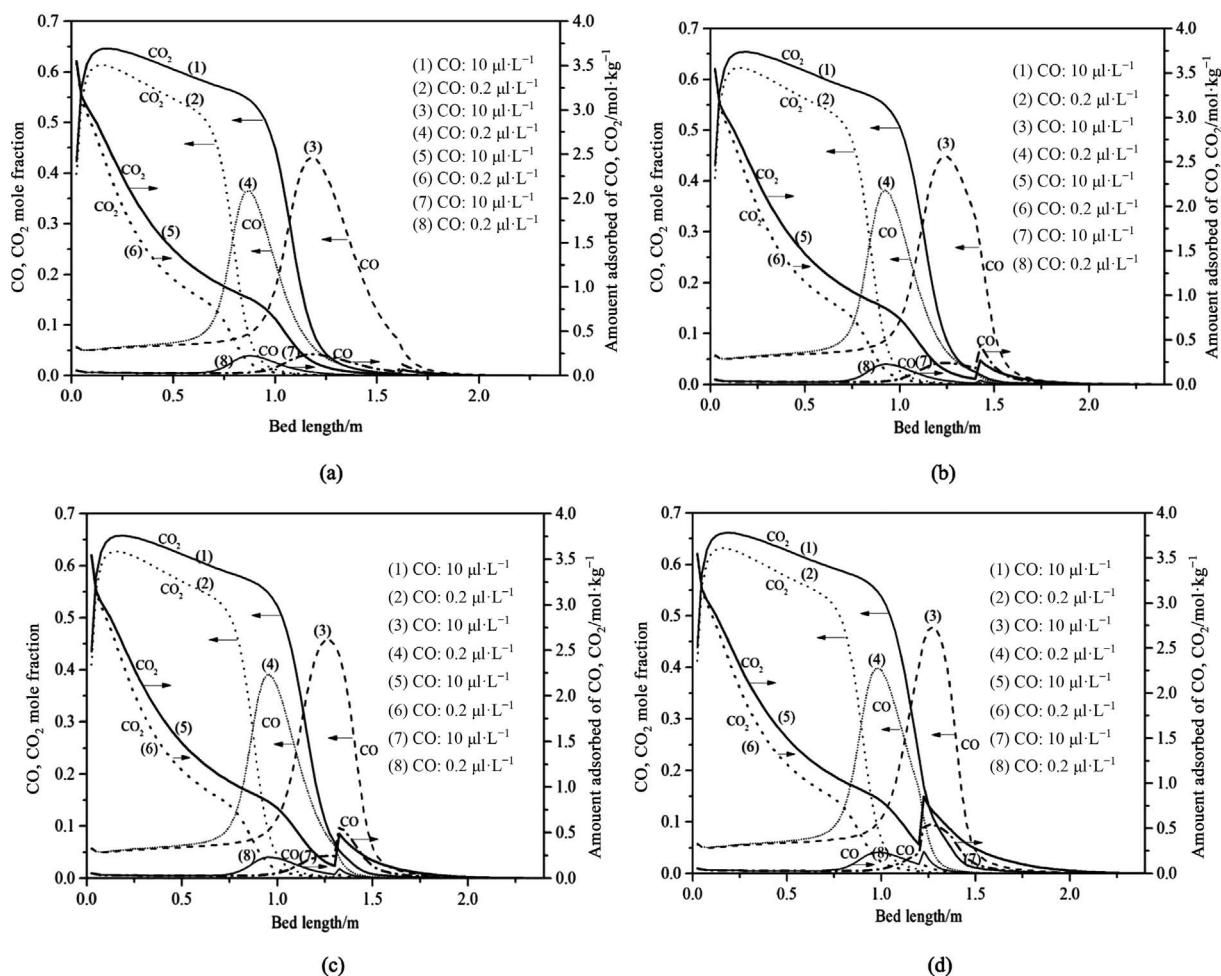


Fig. 9. Solid and gas-phase concentration profiles with the AC-to-5A ratio at the end of PP step((a):1.6/0.8,(b):1.4/1,(c):1.3/1.1,(d):1.2/1.2).

the adsorption front of CO and CO₂ in the solid phase, when the CO content in product is high, it is forward (at the outlet of the adsorption bed), and when the CO content is low, it is backward. That is, the lower the CO content in the product, the longer the mass transfer zone of impurities in adsorption bed. When the ratio of activated carbon to zeolite 5A is 2:1, the concentration front of CO in the gas phase after PP step does not enter the zeolite 5A bed, and the CO₂ in the solid phase is mainly distributed in the activated carbon layer. When CO content in product is 10 $\mu\text{L}\cdot\text{L}^{-1}$, and the ratio of activated carbon to zeolite 5A reaches 1.4, a small amount of CO₂ in gas and solid phase enters the zeolite 5A layer. With the increase of ratio of zeolite 5A, the amount of CO₂ entering the zeolite 5A bed continues to increase. When CO content in product is 0.2 $\mu\text{L}\cdot\text{L}^{-1}$, and the ratio of activated carbon to zeolite 5A reaches 1:1, only a small amount of CO₂ in gas and solid phase enters the zeolite 5A layer. Therefore, when producing hydrogen with CO content of 10 $\mu\text{L}\cdot\text{L}^{-1}$, the optimal ratio of activated carbon to zeolite 5A is 1.4, while when producing hydrogen with CO content of 0.2 $\mu\text{L}\cdot\text{L}^{-1}$, the ratio of activated carbon to zeolite 5A can reach 1:1.

5. Conclusions

In this paper, a non-isothermal model of eight-bed PSA hydrogen system four-stage pressure equalization was developed. The model uses composite adsorption bed of activated carbon and zeolite 5A, and hydrogen mixture with five-component (H₂/N₂/CH₄/CO/CO₂ = 74.59%/0.01%/4.2%/2.5%/18.7% (vol)) as raw material, and two kinds of product hydrogen with CO content 10 $\mu\text{L}\cdot\text{L}^{-1}$ and 0.2 $\mu\text{L}\cdot\text{L}^{-1}$ as target product. From the simulation, the pressure curve and temperature field for producing two kinds of hydrogen, and temperature change in different adsorption bed were compared. Moreover, the difference of separation performance when producing two kinds of hydrogen, and the influence of PP differential pressure and the ratio of activated carbon to zeolite 5A were compared. Under the conditions that the adsorption pressure is 2.6 MPa, desorption pressure is 0.12 MPa, PP differential pressure is 0.263 MPa and the ratio of activated carbon to zeolite 5A is 1.4:1, when the content of CO is 10 $\mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery is 89.8%, and the average flow rate of feed gas is 0.529 $\text{mol}\cdot\text{s}^{-1}$; when the content of CO is 0.2 $\mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery is 85.2%, and the average flow rate of feed gas is 0.43 $\text{mol}\cdot\text{s}^{-1}$. When the PP differential pressure increases from 0.233 MPa to 0.3 MPa, the recovery and feed flow rate first increase and then decrease, and reach the maximum when the PP differential pressure is 0.263 MPa. When CO content in hydrogen is 10 $\mu\text{L}\cdot\text{L}^{-1}$, the corresponding P/F is 9.12%, while when CO content in hydrogen is 0.2 $\mu\text{L}\cdot\text{L}^{-1}$, the corresponding P/F is 11.22%. When the ratio of activated carbon to zeolite 5A decreases from 2:1 to 1:1, the hydrogen recovery increases gradually. When CO in product gas is 0.2 $\mu\text{L}\cdot\text{L}^{-1}$, the hydrogen recovery increases more, from 83.96% to 86.37%. When the ratio of activated carbon to zeolite 5A reaches 1:1, a large amount of CO₂ in gas and solid phase does not enter the zeolite 5A layer. However, when CO in product gas is 10 $\mu\text{L}\cdot\text{L}^{-1}$, and the ratio of activated carbon to zeolite 5A is less than 1.4:1, more CO₂ in gas and solid phase will enter the zeolite 5A layer. This article can be guidance of PSA design for fuel cell hydrogen production.

Nomenclature

A_w	cross-sectional area of column wall, m^2
c_{pg}	heat capacity of gas phase, $\text{kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$
c_{ps}	heat capacity of particle, $\text{kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$
c_{pw}	heat capacity of column wall, $\text{kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$
D_{ax}	mass axial dispersion coefficient, $\text{m}^2\cdot\text{s}^{-1}$
h_i	heat transfer coefficient of inner wall, $\text{kW}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$
h_o	heat transfer coefficient of outer wall, $\text{kW}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$

$-\Delta H_{\text{ads},i}$	heat of adsorption of i component, $\text{kJ}\cdot\text{mol}^{-1}$
IP_{1i}	LRC coefficient, $\text{kmol}\cdot\text{kg}^{-1}$
IP_{2i}	LRC coefficient, $\text{kmol}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$
IP_{3i}	LRC coefficient, MPa^{-1}
IP_{4i}	LRC coefficient, K
$K_{\text{LDF},i}$	LDF coefficient, s^{-1}
K_z	effective axial thermal conductivity, $\text{kW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$
L	bed length, m
M	molecular weight
P	pressure, MPa
q_i	saturation amount adsorbed of i th component, $\text{kmol}\cdot\text{kg}^{-1}$
q_i^*	equilibrium amount adsorbed of i th component, $\text{kmol}\cdot\text{kg}^{-1}$
R	gas constant, $\text{kJ}\cdot\text{kmol}^{-1}\cdot\text{K}^{-1}$
R_i	inner radius of column, m
R_o	outer radius of column, m
r_p	particles radius, m
T	gas-phase temperature, K
T_{amb}	ambient temperature, K
T_w	wall temperature, K
t	time, s
u	interstitial velocity, $\text{m}\cdot\text{s}^{-1}$
v	superficial velocity, $\text{m}\cdot\text{s}^{-1}$
y_i	mol fraction of i th component in the gas phase,
Z	distance along the length of the column, m
ε	bed void fraction
μ	dynamic gas viscosity, $\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$
ρ_B	Bulk solid density of adsorbent, $\text{kg}\cdot\text{m}^{-3}$
ρ_g	gas density, $\text{kg}\cdot\text{m}^{-3}$
ρ_s	particles density, $\text{kg}\cdot\text{m}^{-3}$
ρ_w	density of wall, $\text{kg}\cdot\text{m}^{-3}$
Ψ	shape factor

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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