

## Control for Hydrogen Recovery in Pressuring Swing Adsorption System Modeling

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**Abstract:** Pressure swing adsorption (PSA) process can be used to purify and isolate various gases from complex mixtures. Creating ultra-pure (99.999+ %) hydrogen (H<sub>2</sub>) is its primary use in industries. This is accomplished by applying various pressures on adsorbents with varying loading capacities. The PSA system designed in this work, was more of an experimental basis, despite the huge development in real-world uses of this technology. Because of high adsorption, the flow rate in a realistic model of a PSA cycle varies. However, due to adiabatic temperature conditions, the heat impact must also be considered. As part of this study, a PSA cycle for H<sub>2</sub> purification was modelled and simulated. With this goal in mind, a two-bed, six-cycle system was considered, with activated carbon (AC) and zeolite layers in each bed. CO<sub>2</sub>, H<sub>2</sub>, CH<sub>4</sub>, CO, and N<sub>2</sub> were all present in the feed stream, as is commonplace when purifying H<sub>2</sub>. The PSA process simulation indicated that the concentration of H<sub>2</sub> went from 75.7% (vol%) in the feed to 99.996% (vol%) in the final product including CO<sub>2</sub>, H<sub>2</sub>, CH<sub>4</sub>, CO, N<sub>2</sub>.

**Keywords:** Hydrogen Production; Pressure Swing Adsorption; Zeolite; Skarstrom Cycle; Activated Carbon; Gas Purification; Blown down Bed; Modeling and Simulation.

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### 1. Introduction

Hydrogen is an essential compound in many chemical process industries, such as ammonium production, petroleum processing, metallurgical processing, electronics, town gas, and aerospace. It is produced normally by removing or recovering other components from gaseous mixtures generated in various chemical processes. Several separation and purification methods could be employed for H<sub>2</sub> recovery from the gas mixtures, such as extraction, distillation and adsorption techniques [5]. While distillation may seem like the easier option, rising energy prices have forced scientists to focus on adsorption. Because of their useful properties for separating gas mixtures, adsorption methods, particularly pressure swing adsorption (PSA), are widely employed. In gas separation, adsorption is defined as the adhesion or retention of selective components of a gas mixture stream coming into contact with the surface of a specific solid adsorbent as a result of the force of the field at the surface.

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PSA is an adsorption process in which bulk separation of the gas mixture is achieved by repetitive adsorption at high pressure and desorption at low pressure [6]. PSA like any other adsorption process, requires an adsorbent that preferentially adsorbs one or more components/species of a gas mixture via two main steps: an adsorption step in which the species to be adsorbed are selected from the feed, and a desorption step in which the species have been removed from the adsorbent and are ready to be used again. PSA is a separation process widely used for gas separation. Its major application is in the separation of H<sub>2</sub>, and O<sub>2</sub> from air, normal and iso-paraffins, and various drying operations [3]. Hence, this research aims to develop an H<sub>2</sub> separation and purification unit for industrial applications. It would be achieved by developing models for the PSA system, simulating the process models, and designing a control strategy for the PSA unit for efficient purification and separation.

## 2. Previous Work

Modelling of PSA involves the development of mathematical models that describe the adsorption and desorption processes occurring in the adsorbent bed. Several models have been proposed in the literature, ranging from simple equilibrium models to more complex dynamic models. For example, Yang et al. [11] developed a dynamic model of PSA for H<sub>2</sub> recovery from natural gas, which accounted for the mass transfer and heat transfer processes occurring in the adsorbent bed. The model was validated using experimental data and was found to accurately predict the performance of the PSA system. Control of PSA involves the development of control strategies that optimize the performance of the PSA system. Several control strategies have been proposed in the literature, including model-based control, fuzzy logic control, and artificial neural network control. For example, Zhang et al. [12] proposed a model-based predictive control strategy for H<sub>2</sub> recovery, which uses a dynamic model of the PSA system to predict the behaviour of the system and optimize the operating conditions. The control strategy was shown to improve the H<sub>2</sub> recovery efficiency and reduce the energy consumption of the PSA system.

PSA involves adsorbent materials such as zeolite and Activated Carbon (AC), most commonly used to selectively adsorb H<sub>2</sub> from a gas mixture. Zeolite is a porous material with a high surface area and a regular network of channels and cavities, which makes it highly selective for certain gas molecules, including H<sub>2</sub>. They are also chemically stable and easily regenerated, making them ideal for repeated adsorption and desorption cycles. Using zeolites in PSA systems allows for efficient and cost-effective separation of gas mixtures, making them valuable tools in various industrial applications. Conversely, AC is a highly porous material with a high adsorption capacity for a wide range of gas molecules, including H<sub>2</sub>. In a PSA cycle that utilizes AC, a gas mixture is fed into a PSA vessel containing a bed of AC. The gas mixture is then pressurized, allowing the more strongly adsorbed gas components to be selectively adsorbed onto the AC. In contrast, the less strongly adsorbing gas components pass through the bed and are collected as the product gas. The efficiency of the PSA system using AC depends on several factors, including the characteristics of the AC, the operating conditions, and the nature of the gas mixture. Some of the effects of AC in PSA include selectivity, capacity, regeneration and temperature. Generally, using AC in PSA systems provides an efficient and cost-effective gas purification and separation solution. It is widely used in food and beverage, pharmaceuticals, and chemical processing industries.

### 2.1. Principles of PSA Cycle

The PSA or Pressure Swing Adsorption cycle is a process used in gas separation based on the adsorption principle. It involves the selective adsorption of certain gases from a mixture of gases under high pressure and then releasing them under low pressure. It is widely used in petroleum refining, natural gas processing, and air separation industries. This article will discuss the PSA cycle in detail, including its applications, principles, and advantages. The PSA cycle operates based on the principle of adsorption. Adsorption is a process in which a gas or liquid is attracted to the surface of a solid material and adheres to it. In the case of the PSA cycle, the solid material is a porous adsorbent, such as AC or zeolites. When a mixture of gases is passed through the adsorbent bed under high pressure, certain gases are selectively adsorbed onto the surface of the adsorbent. In contrast, others pass through the bed and are collected as the product stream. Once the adsorbent bed is saturated with the selectively adsorbed gas, the pressure is reduced, and the adsorbed gas is released from the adsorbent and collected as the waste stream. This process is repeated in cycles to produce a continuous supply of the desired gas.

The PSA cycle involves two main steps: the adsorption step and the desorption step. The gas mixture is passed through the adsorbent bed under high pressure during adsorption. The selective adsorption of certain gases occurs on the adsorbent's surface. The adsorption is highly dependent on the properties of the adsorbent, such as its pore size, surface area, and affinity for certain gases. The adsorption step typically lasts for a few minutes to several hours, depending on the size of the adsorbent bed and the properties of the gas mixture. During the desorption step, the pressure is reduced, causing the adsorbed gas to be released from the adsorbent and collected as the waste stream. The desorption step is typically shorter than the adsorption step and lasts a few seconds to a few minutes. The pressure is then increased again, and the adsorption step is repeated to produce a continuous supply of the desired gas.

## 2.2. Applications and challenges of the PSA Cycle

The PSA cycle is widely used in petroleum refining, natural gas processing, and air separation industries. Some of the specific applications of the PSA cycle include [9]:

- Hydrogen purification: H<sub>2</sub> is widely used in petrochemicals, electronics, and metallurgy industries. The PSA cycle is used to purify H<sub>2</sub> from a mixture of gases, such as methane, ammonia, and carbon monoxide.
- Nitrogen generation: Nitrogen is used in various industries, including food processing, electronics, and pharmaceuticals. The PSA cycle is used to separate N<sub>2</sub> from the air.
- Oxygen generation: Oxygen is used in various industries, including medical, chemical, and metallurgical. The PSA cycle may be used to separate oxygen from air.
- Carbon dioxide removal: Carbon dioxide is a greenhouse gas responsible for global warming. The PSA cycle removes CO<sub>2</sub> from industrial processes like power plants and cement factories.

The PSA cycle has several advantages over other gas separation techniques. Some of the advantages of the PSA cycle includes: high selectivity, in which the PSA cycle can selectively adsorb certain gases from a mixture of gases and makes it a highly efficient and cost-effective gas separation technique; low energy consumption, in which the PSA cycle requires less energy than other gas separation techniques, such as cryogenic distillation and makes it a more environmentally friendly and cost-effective option; continuous operation, where the PSA cycle can operate continuously, providing a constant supply of the desired gas and makes it a highly efficient gas separation technique for industrial applications; compact size, where which the PSA cycle can be designed to be compact and portable, making it ideal for use in remote locations or for mobile applications; and versatility, in which the PSA cycle can be used to separate a wide range of gases, including H<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub> and CO<sub>2</sub>, making it a versatile gas separation technique for a variety of industries.

Although the PSA cycle has several advantages over other gas separation techniques, it also has some challenges. Some of the challenges of the PSA cycle include [1]:

- Adsorbent regeneration: The adsorbent used in the PSA cycle needs to be regenerated periodically. The regeneration process involves purging the adsorbent with a gas to remove the adsorbed gas. This process can be time-consuming and energy-intensive, reducing the overall efficiency of the PSA cycle.
- Adsorbent fouling: Adsorbent used in the PSA cycle can become fouled over time, reducing effectiveness. This can be caused by contamination, high humidity, or exposure to corrosive gases.
- High capital cost: The capital cost of a PSA system can be high, making it less feasible for small-scale applications.
- Dependence on adsorbent properties: The effectiveness of the PSA cycle is highly dependent on the properties of the adsorbent used. Selection of the right adsorbent can be a complex process, and the performance of the PSA cycle can vary depending on the adsorbent used.
- Despite these challenges, the PSA cycle remains a popular and effective gas separation technique for various industrial applications.

## 3. Methodology

### 3.1. Materials

An X64-based Windows 10 Pro Personal Computer type (HP 15 Notebook), manufactured by Hewlett-Packard with Intel® Celeron® CPU N2840 @ 2.16GHz, 2159Mhz, @ core(s) processor and an Aspen Adsorption® Simulation software Version 8.8 were used.

### 3.2. Modeling Procedure

It involves the following stepwise procedures: the opening of Aspen adsorption software, defining components and property models, configuring CSS\_Info and adding cyclor organizer, building flowsheet, specifying feed stream (Table 1), specifying valve schedule (Table 1), specifying absorbent properties (Table 2), specifying energy and heat transfer values (Table 3), specifying equilibrium parameters (Table 4), performing gCSS initialization and lastly, configure and generate cycle organizer and task respectively.

**Table 1:** Feed Composition and Valve Coefficient ( $C_V$ ) [10]

Component	Composition
H <sub>2</sub>	0.757

N <sub>2</sub>	0.024
CO	0.007
CO <sub>2</sub>	0.18
CH <sub>4</sub>	0.032
<b>Valve</b>	<b>C<sub>v</sub> (kmol/(s bar<sup>-1</sup>))</b>
VF	$2 \times 10^{-5}$
VP	$5.6 \times 10^{-9}$
VW	$1 \times 10^{-5}$
V-Purge	$1.8 \times 10^{-7}$

As shown in Table 2, adsorbents with smaller particle radii are preferred in PSA systems because they provide higher surface area and better mass transfer characteristics, resulting in higher adsorption capacity and faster adsorption kinetics.

**Table 2:** Adsorbent Property Specifications [2]

Property	Units	Activated Carbon	Zeolite
Particle radius	M	0.00115	0.00157
Void fraction	m <sup>3</sup> void /m <sup>3</sup> bed	0.35	0.35
Particle porosity	m <sup>3</sup> void /m <sup>3</sup> bed	0.61	0.65
Particle density	Kg/m <sup>3</sup>	850	1160
Bed density	Kg/m <sup>3</sup>	482	746
Particle external area to surface volume	m <sup>-1</sup>	2608.7	1910.83

However, using an adsorbent with a larger particle radius in a PSA system will cause a reduced surface area, mass transfer, pressure drop, and mechanical stability. The heat of adsorption of various components in a gas mixture can significantly affect the performance of a PSA system. The heat of adsorption refers to the heat released or absorbed when a gas molecule is adsorbed onto an adsorbent surface. In a PSA system, gas mixtures are separated based on the differential adsorption of the components onto the adsorbent surface. Components (Table 3) with higher heat of adsorption are typically more strongly adsorbed onto the adsorbent and require higher desorption temperatures or longer desorption times to be released.

**Table 3:** Energy Balance Properties [2]

<b>Heat of Adsorption</b>			
Component	Units	Activated Carbon	Zeolite
CH <sub>4</sub>	MJ/Kmol	-22.175	-22.175
CO	MJ/Kmol	-20.92	-20.92
CO <sub>2</sub>	MJ/Kmol	-20.0	-20.0
H <sub>2</sub>	MJ/Kmol	-11.715	-11.715
N <sub>2</sub>	MJ/Kmol	-20.0	-20.0
<b>Heat Transfer Coefficient</b>			
	Units	Value	
Activated Carbon	Mw/Wm <sup>2</sup> k	$3.851 \times 10^5$	
Zeolite	Mw/Wm <sup>2</sup> k	$3.851 \times 10^5$	
<b>Heat Capacity</b>			
Activated Carbon	MJ/Kg/K	0.0010465	
Zeolite	MJ/Kg/K	$9.209 \times 10^4$	

Equilibrium properties of an adsorbent, such as the adsorption isotherm and selectivity, can significantly affect the performance of a PSA system. Adsorption isotherm is a relationship between the amount of gas adsorbed onto the adsorbent and the gas pressure. At the same time, selectivity refers to the ability of an adsorbent to preferentially adsorb one component over another. Effects of equilibrium properties of the adsorbents (Table 4) are adsorption capacity, selectivity of adsorbents, cycle time, and energy consumption.

**Table 4:** Equilibrium Property Specification [2]

Equilibrium Property	Activated Carbon	Zeolite
IP1,CH <sub>4</sub>	0.023860	0.005833
IP2, CH <sub>4</sub>	$-5.62 \times 10^{-5}$	$-1.19 \times 10^{-5}$
IP3, CH <sub>4</sub>	0.003478	$6.51 \times 10^{-4}$
IP4, CH <sub>4</sub>	1159.000	1731
IP5,CH <sub>4</sub>	1.618.000	0.82
IP6, CH <sub>4</sub>	-248.900	53.15
IP1,CO	0.03385	0.011845
IP2, CO	$-9.07 \times 10^{-5}$	$-3.13 \times 10^{-5}$
IP3, CO	$2.31 \times 10^{-4}$	0.0202
IP4, CO	1751.00	763
IP5, CO	3.05300	3.823
IP6, CO	-654.400	-931.3
IP1,CO <sub>2</sub>	0.028797	0.01003
IP2, CO <sub>2</sub>	-7.00E-05	-1.86E-05
IP3, CO <sub>2</sub>	0.01000	1.58E-04
IP4, CO <sub>2</sub>	1030.00	207
IP5, CO <sub>2</sub>	0.99900	-5.648
IP6, CO <sub>2</sub>	-37.400	2098
IP1,H <sub>2</sub>	0.016943	0.004314
IP2, H <sub>2</sub>	$-2.1 \times 10^{-5}$	$-1.06 \times 10^{-5}$
IP3, H <sub>2</sub>	$6.25 \times 10^{-5}$	0.002515
IP4, H <sub>2</sub>	1229.00	458
IP5, H <sub>2</sub>	0.9800	0.986
IP6, H <sub>2</sub>	43.03	43.03
IP1,N <sub>2</sub>	0.001644	0.004813
IP2, N <sub>2</sub>	$-7.3 \times 10^{-7}$	-6.68E-06
IP3, N <sub>2</sub>	0.0545	$5.7 \times 10^{-4}$
IP4, N <sub>2</sub>	326.00	1531
IP5, N <sub>2</sub>	0.908	0.842
IP6, N <sub>2</sub>	0.991	-7.467

IP: Isotherm Parameter

Afterwards, the dynamic simulation was run, and the plot of production composition was generated.

### 3.3. PSA Cycle

In this piece, the concept of two beds (based on Skarstrom cycle) will serve as the foundation [7]. There are six fundamental steps in PSA, and they include pressurisation, adsorption, blowdown, purge, and pressure equilibration [8]. Step 1 involves increasing the operating pressure of the feed from the feed end through Bed 1 (by opening valve VF1) to enrich the selectively adsorbed species in the gas phase. Bed 2 is blown down to the atmospheric pressure (by opening valve VW2) in the opposite direction to Bed 1 to prevent the contamination of the product end (H<sub>2</sub>) with more of the strongly adsorbed species (CO<sub>2</sub>, CO, CH<sub>4</sub> and N<sub>2</sub>). This is schematically represented in Figure 1a.

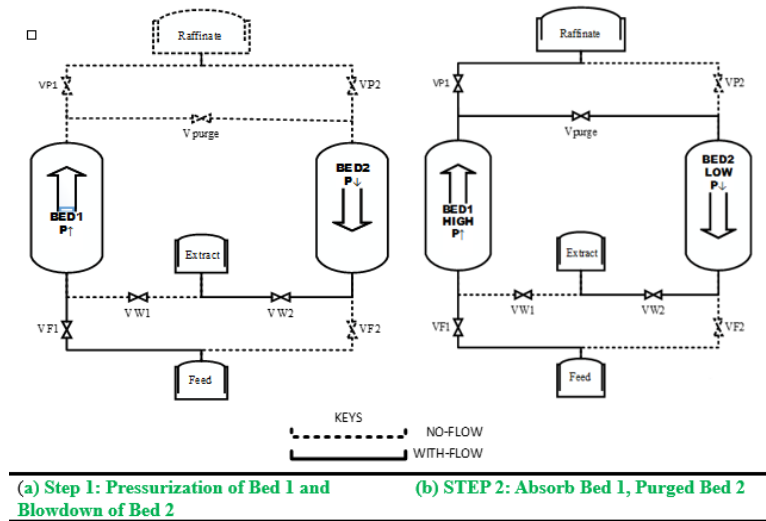


Figure 1: Step 1 and Step 2

To retain the most strongly adsorbed species ( $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_4$ , and  $\text{N}_2$ ), the high-pressure feed is sent through Bed 1 (by opening valve VF 1) in Step 2. The product-enriched gas stream ( $\text{H}_2$ ) is discharged at a somewhat lower pressure than the feeds. By opening valve VP1, a portion of the effluent is diverted as a raffinate product, while the remaining volume is used to purge Bed 2 at a low pressure (by opening valve V-purge). The direction of the purge bed is also opposite to the feed flow, as shown in Figure 1b. Step 3 is the pressure equalization step, after Bed 2 has been purged to blow down the raffinate product and Bed 1 has completed the high-pressure adsorption step and becomes saturated with the strongly adsorbed components of the gas mixture. Instead of blowing down Bed 1 directly, the two beds are connected through their product end (by opening valve V-purge) to equalize their pressure, as shown in Figure 2a.

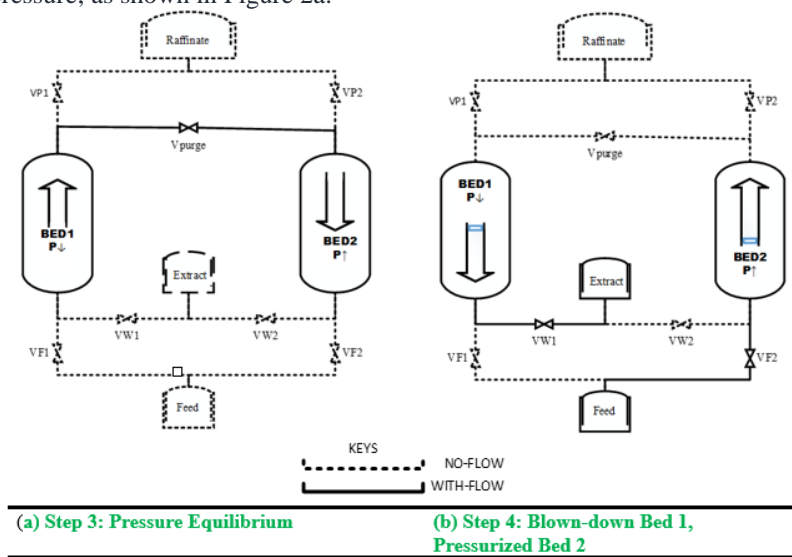
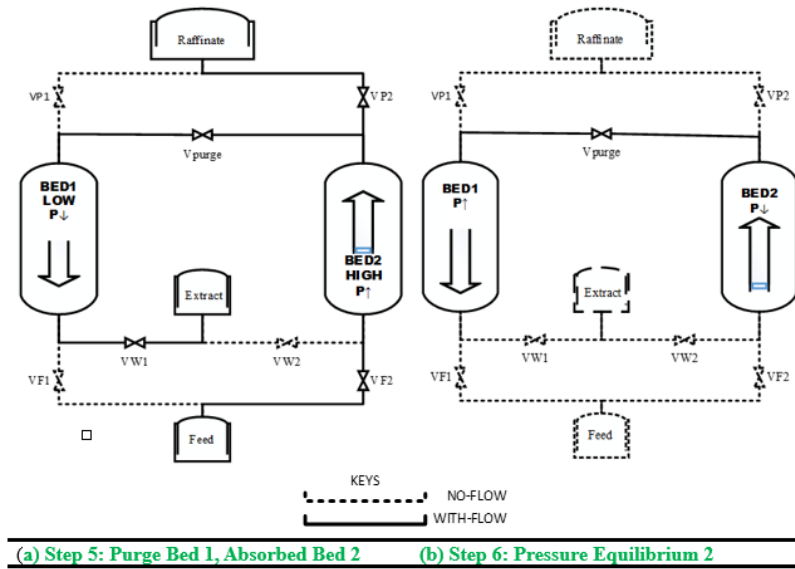


Figure 2: Steps 3 and Step 4

In Stage 4, following the initial pressure equilibration step, the two beds' order of operations is reversed. In order to enrich the less selectively adsorbed species in the gas phase, we now pressurise Bed 2 (by opening valve VF2) using the feed from the feed end. To avoid Bed 2's more strongly adsorbed species contaminating the product end, Bed 1 is blown down to atmospheric pressure (by the opening of valve VW1) in the reverse direction. This is depicted in a simplified form in Figure 2b. In Step 5, the high-pressure feed flows through Bed 2 (through the opened VF 2), where the more strongly adsorbed component remains in the bed and the gas stream enriched with the less strongly adsorbed component exits as an effluent component at a pressure slightly below that of the feed. By releasing pressure on Bed 1 through valve VP2, some of the effluent can be collected as a raffinate product, while the rest is used to flush the bed (by opening valve V-purge). Similar to the feed flow, the purge bed flows counter clockwise in Figure 3a.



**Figure 3:** Step 5 and Step 6

Step 6: After Bed 1 has been purged to blowdown the raffinate product and Bed 2 has completed the high-pressure adsorption step and becomes saturated with the strongly adsorbed components of the gas mixture, the second pressure equilibration step follows to conserve energy and separate work. Instead of blowing down Bed 2 directly, the two beds are connected through their product end (by opening valve V-purge) to equalize their pressure, as shown in Figure 3b. To accomplish this kind of switching, when to open and close the valves needs to be carefully scheduled. Since the schedule of the steps above and the valve layout described in the Figures are carefully examined. The valve opening and closing schedule in Table 5 can be easily created.

**Table 5:** Valve Schedule

Steps	Function	VF1	VF2	VP1	VP2	VW1	VW2	VPURGE
Step 1	Pressurizing Bed 1 and blowing down Bed 2	ON	OFF	OFF	OFF	OFF	ON	OFF
Step 2	Adsorb Bed 1 and purge Bed 2	ON	OFF	ON	OFF	OFF	ON	ON
Step 3	Pressure Equilibration 1	OFF	OFF	OFF	OFF	OFF	OFF	ON
Step 4	Pressurizing Bed 2 and blow down Bed 1	OFF	ON	OFF	OFF	ON	OFF	OFF
Step 5	Adsorb Bed 2 and purge Bed 1	OFF	ON	OFF	ON	ON	OFF	ON
Step 6	Pressure Equilibration 2	OFF	OFF	OFF	OFF	OFF	OFF	ON

To set up valve opening and closing schedules in Aspen Plus, first, define the valve, then define the operating conditions, define the schedule and simulate and optimize the process to ensure the valve operates as expected.

## 4. Results and Discussion

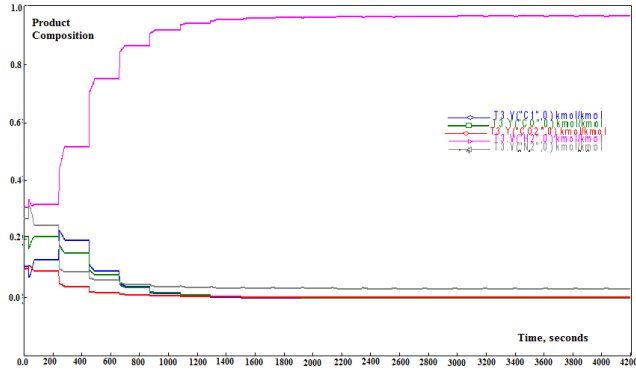
### 4.1. Simulation Result

The model of PSA for H<sub>2</sub> recovery from synthesis gas was simulated for ten cycles to attain a steady state, which was attained after 4200 seconds. A typical cycle of a normal PSA is 10 minutes [4]. The variation of concentration of gas components (H<sub>2</sub>, N<sub>2</sub>, CO, CH<sub>4</sub>, and CO<sub>2</sub>) after the feed step along the bed in the steady state are shown in Table 6 [13].

**Table 6:** Comparison Between Feed Composition and Final Composition

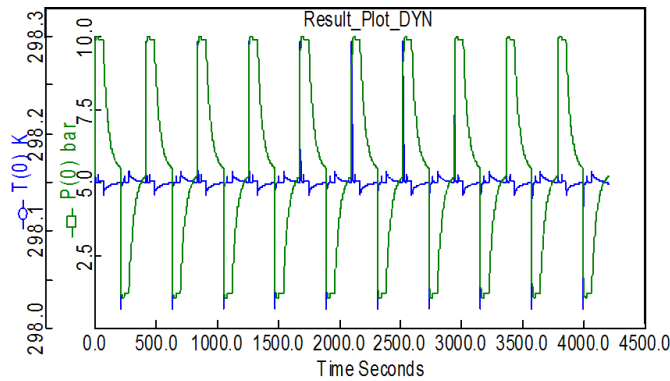
Component	Initial Feed Composition	Final Composition
H <sub>2</sub>	0.757	0.999983
N <sub>2</sub>	0.024	0.007018
CO	0.007	8.414373 × 10 <sup>-5</sup>
CO <sub>2</sub>	0.18	0.009261
CH <sub>4</sub>	0.032	6.168925 × 10 <sup>-6</sup>

A high amount of H<sub>2</sub> was recovered from the synthesis gas after ten cycles at 4200 seconds. Other gaseous components like CO, N<sub>2</sub>, CH<sub>4</sub> and CO<sub>2</sub> appear negligible in the final output composition. Figure 4 shows the plot of the product composition with time. It illustrates the dynamics of H<sub>2</sub> composition until it attains a steady state. From the simulation result in Figure 4, it was observed that 99.99+ of H<sub>2</sub> was recovered after modelling the PSA by specifying the model parameters, adsorbents properties, and energy and heat coefficients.



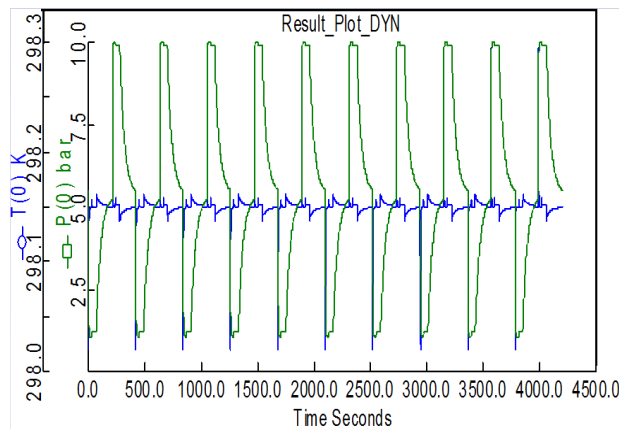
**Figure 4:** Product Composition with Time

Figure 5 shows the dynamic results of pressure and temperature in Bed 1, where the curve illustrates how pressure drops and rises in the bed with time to accomplish the required step times to complete the cycles.



**Figure 5:** Pressure and Temperature Dynamics in Bed 1

Figure 5 shows how pressure swings with time aid the cycle stages to separate H<sub>2</sub> within the bed by adsorbing H<sub>2</sub> at high pressure and desorbing the impurities at low pressures. Meanwhile, Figure 6 shows the dynamics of pressures and temperature in Bed 2.



**Figure 6:** Pressure and Temperature Dynamics in Bed 2



In Figure 6, the graph shows pressure initiates at a lower rate, which signifies the initial blowdown of Bed 2 in the first stage and subsequent pressurization when Bed 1 is saturated.

## 5. Conclusion

The development of a model for the removal of contaminants from H<sub>2</sub> using a layered activated carbon/zeolite PSA method was investigated. The feed stream mixture included CO<sub>2</sub>, CH<sub>4</sub>, CO, and N<sub>2</sub>. The Skarstrom model of PSA for H<sub>2</sub> recovery was developed by obtaining the required parameters describing the unit models. The models were simulated using Aspen Adsorption software to observe the response, where 99.9998% of H<sub>2</sub> was recovered from the result obtained. At the end of each cycle, the number of gas components adsorbed was measured and compared. Since zeolite is so pricey, cramming an entire column with it would be impractical. A column with stacked layers of packing seems like a feasible solution. Activated carbon removes N<sub>2</sub> and CO from the air and is a good candidate for the initial packing. The remaining CO<sub>2</sub> and CH<sub>4</sub> can be absorbed by zeolite packing. Water vapour in the stream can be eliminated with a first layer of alumina or silica. Water vapour has been determined to threaten AC and zeolite packing because it is not easily desorbed. In order to achieve the required level of H<sub>2</sub> purity, future research in this area should focus on cost, that will effectively improve the thickness of each layer. Other packings can be compared in the same way, too.

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**Ethics and Consent Statement:** This work is a draft from the corresponding author's B.Eng. Degree Project. Supervisors and examiners mentioned in the acknowledgement approve the work before awarding a Bachelor's Degree to the candidate. Authors of the work unanimously consent to make this publication available to all interested people for reading, teaching and learning.

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