



# Optimization of Hydrogen Purification Using a Temperature Swing Adsorption System (TSA) Method in the Natural Gas Processing

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**Abstract.** Hydrogen is an example of renewable and environmentally friendly future energy. Hydrogen as a fuel for emission-free vehicles has attracted much attention recently. The level of hydrogen purity greatly determines its use and economic selling price. Increasing the hydrogen purity can be done in two ways: TSA (Temperature Swing Adsorption) and PSA/VSA (Pressure/Vacuum Swing Adsorption). This study systematically uses the TSA method to optimize the hydrogen purification process. This study aims to obtain a TSA system design based on adsorbents' ability to absorb impurities in hydrogen purification. The designed TSA system also observes the energy-saving operating conditions to produce high-purity hydrogen products. The TSA process consists of six stages, and stages are feed pressurization (FP), adsorption (AD), depressurizing pressure equalization (DPE), depressurization (DP), purge (P), and pressurizing pressure equalization (PPE). The principle of TSA is to use temperature to separate hydrogen from impurities and improve hydrogen purity. All the processes simulated in Aspen Adsorption v11. TSA is carried out by varying temperatures at 278.15 K, 298.15 K, 303.15 K, and 318.15 K. The results of system separation optimization ( $H_2/CO_2/CH_4/CO/N_2 = 0.564/0.031/0.266/0.084/0.055$ ) using the TSA method obtained high purity at a temperature of 298.15 K so that it achieved hydrogen purity for the single-bed column of 99.97% and the two-bed column of 99.99%.

**Keywords:** Aspen Adsorption, Adsorption, Hydrogen, Purification, Temperature Swing Adsorption

## 1 Introduction

Hydrogen is a form of energy carrier that can be generated from both fossil fuels and renewable energy sources. It can be utilized for fuel cell-based applications free of pollution emissions, like hydrogen fuel cell vehicles (HFCV). To maintain high-purity hydrogen and sustain the fuel cell applications' good performance, an efficient hydrogen purification process is crucial to the hydrogen economy [1]. The two main techniques for adsorption operations for hydrogen purification are TSA (Temperature Swing Adsorption) and PSA/VSA (Pressure/Vacuum Swing Adsorption) [2].

Adsorption is the phenomenon whereby gas molecules attach themselves to a solid surface and form bonds with the molecules on the surface that are attracted to each other in different amounts because their forces are not balanced. Adsorbents are specific solids that have varying affinities for different gases to adsorb [3]. The adsorbent is packed into multiple columns during the adsorption process. The packed columns are also called absorbers. In comparison to an empty column, the pressure drop in adsorption processes is increased when packed beds are used [4].

Adsorption-based gas separation processes employ preferential adsorption of component gases on solid adsorbent materials, and this phenomenon is controlled. This phenomenon is controlled by adjusting the temperature (TSA) and pressure (PSA) of the gases being separated. Bed-based PSA systems are well established and a vast body of literature describing the flow and adsorption kinetics in the adsorbent bed is available. Various adsorbents including zeolite 5A [5], LTA-zeolites [6], silica molecular sieve [7], and activated carbon [8] have been studied to determine the  $N_2$ ,  $CH_4$ , and  $CO_2$  adsorption capacities, and their applications have been documented for use in PSA-based gas separation systems.

Adsorption systems can be less expensive in some aspects compared to absorption systems. The operating cost of amine absorption systems is reduced because solid adsorbents rarely need to be replaced, in contrast to liquid solvents. On the other hand, complicated valve operation and high compressor operation costs are associated with PSA-based processes. In this regard, TSA-based procedures are preferred since they don't require significant pressure swings to function, which lowers the complexity of the system, and the power needed for the compressor [9]. Despite these benefits, PSA-based systems are still thought to be more energy-efficient than TSA-based systems, primarily because it is more difficult to heat adsorbent that is packed in a bed form [10, 11].

Since heat is required for the regenerating gas, TSA uses a lot of energy. By absorbing them on the packed bed's surface, pollutants are extracted from the air stream. During the feed period, the cycle time is prolonged, and the heat pulse is permitted to leave the adsorbent bed. Regeneration uses heat to desorb contaminants. Heat pulse is produced and travels counter-currently through the bed in the direction of feed. More than one unwanted gas is removed. To regenerate the gas, a high temperature is needed [12].

TSA is superior to PSA in that it can separate contaminants that can form a strong bond with the adsorbent through a process known as chemisorption (adsorption is the chemical reaction between the adsorbent and surface area). The loading amount that

results in effective separation can also vary with a few degrees of temperature change [13].

Bulk separation is rarely used in favor of the TSA technique during the purification process. The process of adsorption functions in a cycle. The regeneration of the adsorbent bed through temperature increases is the foundation of the TSA method. Regenerating the adsorbent bed through a reduction in overall pressure is the foundation of the PSA method. While the TSA method works well for adsorbents with moderate volatility, PSA is primarily preferred for adsorbents with high volatility [12].

Numerous researchers have concentrated on the process design from the perspectives of numerical simulation and experimental study in order to achieve an applicable and efficient design of the TSA cycle. For example, a thorough modeling study utilizing five distinct materials [14] and considering the thermodynamic properties of the adsorbed phase [15] to evaluate a four-step TSA cycle of the fixed bed. Parametric analysis has also been used to examine the effects of various step types on process performance [16] the optimization of a six-step TSA cycle [17], and a two-step indirect TSA cycle [2].

Tong et al. enhanced the adsorption bed's structure and employed a dual activated carbon bed to replicate the breakthrough curves of the four-component mixture  $H_2/CO/CO_2/CH_4$  [1]. They discovered that the dual bed could effectively lessen the impact of the thermal effect in addition to reducing spatial occupancy and enhancing hydrogen purity. Jee et al. used activated carbon and zeolite as the layer of adsorbent and studied the influence of adsorption pressure, carbon ratio on break through curves and bed temperatures [18].

The objective of this work is to create a model for the breakthrough and TSA process in the Carbon-Zeolite bed for hydrogen purification using Aspen Adsorption. This work not only evaluated the performance but also conducted a parametric study on the performance of hydrogen purification. The influence of adsorption pressure and temperature in the breakthrough process as well as adsorption pressure, temperature, feed time and feed rate in TSA cycles on the hydrogen purification performance are studied.

## 2 Materials and Methods

The research methodology is a decision-making process to obtain an optimization design of the TSA hydrogen purification and recovery system. The aim of this study is to obtain a TSA system design based on the ability of adsorbents to absorb impurities in the hydrogen purification process. The design of the TSA system also considers the aspect of operating conditions in energy saving to produce high-purity hydrogen products.

**Table 1.** Data for Simulation

Components	Compositions
$H_2$	0.564
$CO_2$	0.031
$CH_4$	0.266

	CO		0.084
	N <sub>2</sub>		0.055
	Total		1
<b>Specifications</b>		<b>Value</b>	<b>Unit</b>
	Internal bed radius ( $R_{in}$ )	0.021	M
	Bed length (L)	1.2	M
	Bed porosity ( $\epsilon_b$ )	0.433	m <sup>3</sup> (void)/m <sup>3</sup> (bed)
<b>Specifications</b>		<b>Activated Carbon</b>	<b>Zeolite</b>
	Particle Radius ( $R_p$ )	0.00115	0.00157
	Intra-particle voidage ( $\epsilon_p$ )	0.433	0.357
	Adsorbent density ( $\rho_s$ )	850	1160
			kg/m <sup>3</sup>
<b>Components</b>		<b>Value (1/s)</b>	
	H <sub>2</sub>		0.7
	CO <sub>2</sub>		0.0355
	CH <sub>4</sub>		0.195
	CO		0.15
	N <sub>2</sub>		0.261

The variables considered when optimizing hydrogen purification using the TSA method are pressure, temperature, type of adsorbent, and stage cycle. The temperature in the adsorption process can affect the adsorption capacity and reaction speed of the adsorption material, so that in addition to the pressure variations used, which are 0.1 and 0.01 bar, a temperature variation is also required in the adsorption process. The adsorption process was also tried by varying the temperature of 298.15 and 303.15 K to find out whether the temperature also affects the adsorption process. Selecting the correct type of adsorbent can affect the adsorption capacity and selectivity of certain compounds, as well as the long-term stability of the adsorbent material. The type of adsorbent commonly used in the adsorption process is activated carbon. The stage cycle used in the simulation may affect the simulation results and the optimization of the hydrogen purification process, so it needs to be varied after several cycles that tried and there are 1, 5, 7, 9, 10, 11, 12, 15, 20, and 25.

Some of the assumptions used in simulation are the thermodynamic properties of the gas are calculated using the Peng-Robinson state equation, the equilibrium isotherm for each adsorbent layer is described using the Langmuir model which is affected by partial pressure, the kinetic rate model on a solid film is explained by the linear driving force (LDF) equation with a constant mass transfer coefficient, the pressure in the column is described using the Ergun equation, and the equilibrium of the material is described as convection with constant diffusion.

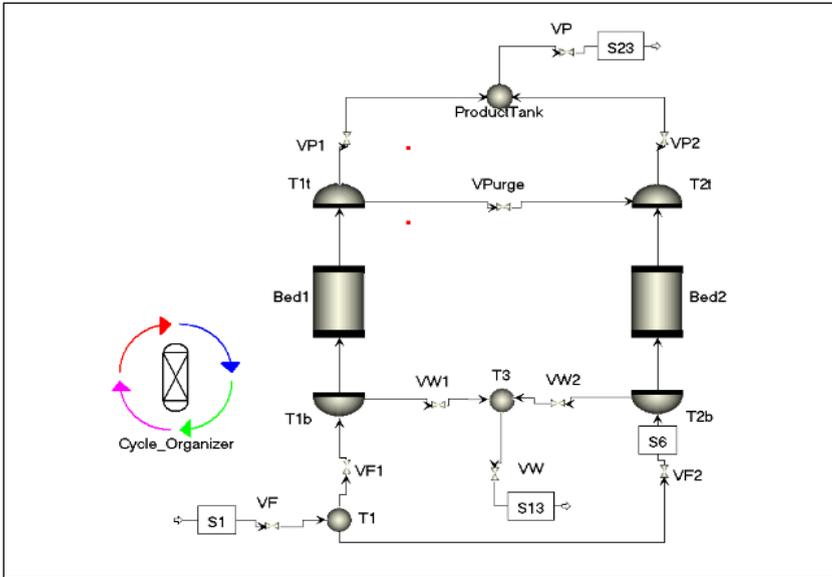


Fig. 1. Simulation for hydrogen purification

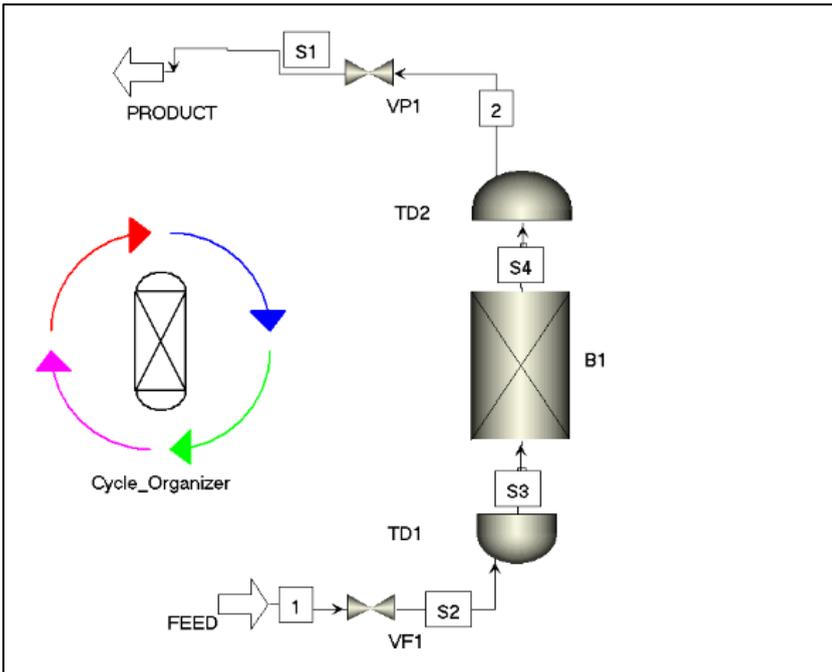
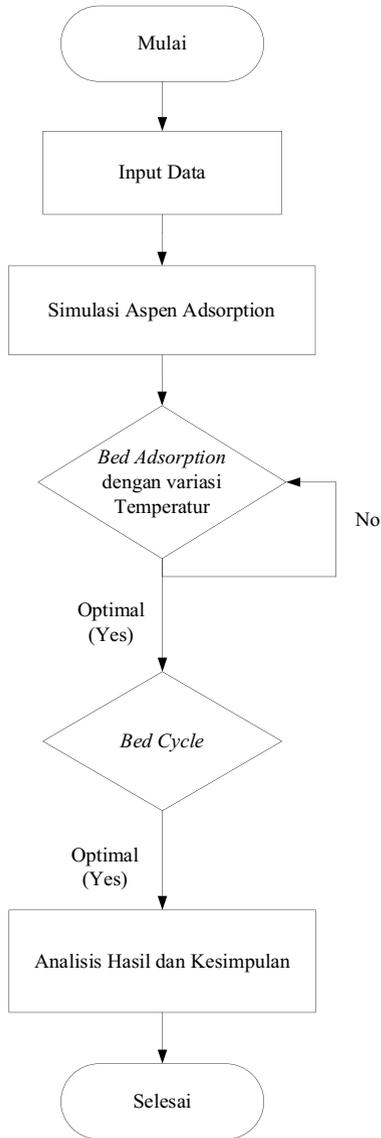


Fig. 2. Simulation for hydrogen purification in one bed



**Fig. 3.** Process flow diagram for hydrogen purification simulation

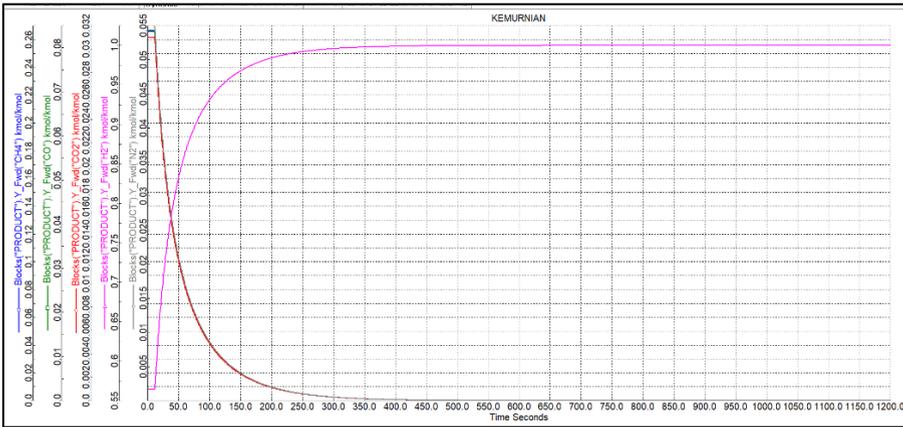
### 3 Results and Discussions

#### 3.1 Effect of Temperature on Hydrogen Purity

**Table 2.** Temperature variation on hydrogen purity with activated carbon adsorbent

Temperature (K)	Pressure (Bar)	Composition				
		CO	CO <sub>2</sub>	H <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub>
318.15	0,1	$5.71464 \times 10^{-4}$	$4.80421 \times 10^{-4}$	0.998859	$-8.286 \times 10^{-5}$	$1.72066 \times 10^{-4}$
308.15		$4.29229 \times 10^{-4}$	$4.21124 \times 10^{-4}$	0.99906	$-8.445 \times 10^{-4}$	$1.98419 \times 10^{-4}$
298.15		$4.29229 \times 10^{-4}$	$4.291124 \times 10^{-4}$	0.999431	$-8.446 \times 10^{-5}$	$1.98419 \times 10^{-4}$
278.15		$1.562271 \times 10^{-4}$	$4.70816 \times 10^{-4}$	0.999211	$-8.264 \times 10^{-5}$	$2.43899 \times 10^{-4}$

Table 2 states that the lower the temperature, the higher the purity of hydrogen. The highest hydrogen purity was obtained at a temperature of 298.15 K, which was 0.999431. To test adsorption at high temperatures, a temperature simulation was used with the purity of hydrogen obtained down, which was 0.998859; 0.99906; 0.999431; and 0.999211. The low pressure allows the adsorbent to be more detectable in absorbing unwanted molecules, thereby increasing the purity of the hydrogen produced.



**Fig. 4.** Result for hydrogen purification simulation

Fig. 3 shows the purity of hydrogen by varying the temperature at 318.15 K, 308.15 K, 298.15 K, and 278.15 K using activated carbon adsorbents. The conclusion of the simulation results shows that at a temperature of 298.15 K using an activated carbon adsorbent produces higher hydrogen purity than at temperatures of 318.15 K, 308.15 K, and 278.15 K. Research by El-Shafie states that lower temperatures provide higher hydrogen content. This is related to the increased efficiency of gas adsorption, as gases tend to be easier to adsorb at low temperatures [19].

**Table 3.** Hydrogen Purity by Bed Number

Temperature (K)	% Hydrogen Purity	
	Activated Carbon	
	1 Bed	2 Bed
298,15	99,97%	99,99%
318,15	97,89%	99,88%

### 3.2 Effect of Cycle on Hydrogen Purity

**Table 4.** Step cycle to hydrogen purity with activated carbon adsorbent

	Step					
	I	II	III	IV	V	VI
<b>Bed 1</b>	FP	AD	DPE	DP	PG	PPE
<b>Bed 2</b>	DP	PG	PPE	FP	AD	DPE
<b>Time (s)</b>	30	40	140	30	40	140

According to the research of Yang et al., the step cycles in this research consists of Feed Pressurization (FP), Adsorption (AD), Depressurizing Pressure Equalization (DPE), Depressurization (DP), Purge (P), and Pressurizing Pressure Equalization (PPE) [20]. The results of the hydrogen purity experiment reached 99.9431% with a batch system with 6 cycles using activated carbon adsorbent. It was concluded that the purity of H<sub>2</sub> was highest with 6 step cycles.

## 4 Conclusion

Hydrogen purification by the Temperature Swing Adsorption (TSA) method based on simulations using adsorption aspen indicated that activated carbon has a greater adsorption ability when purifying hydrogen from multi-component gases (H<sub>2</sub>/CO<sub>2</sub>/CH<sub>4</sub>/CO/N<sub>2</sub> = 0.564/0.031/0.266/0.084/0.055). The purity of hydrogen obtained by 1 bed activated carbon adsorbent is 99.97% and 2 bed is 99.997%. The optimization results for hydrogen purification with a single-column TSA process indicated the operating conditions at a pressure of 0.1 bar at a temperature of 298.15 K showed the highest hydrogen purity result. This is related to the increased efficiency of gas adsorption, as gases tend to be easier to adsorb at low temperatures. Activated carbon adsorbents reach optimum conditions with high hydrogen purity during the 6-step cycle.

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