



# Article Simulation and State Feedback Control of a Pressure Swing Adsorption Process to Produce Hydrogen

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Abstract: One of the separation processes used for the production and purification of hydrogen is molecular sieve adsorption using the Pressure Swing Adsorption (PSA) method. The process uses two beds containing activated carbon and a sequence of four steps (adsorption, depressurization, purge, and repressurization) for hydrogen production and purification. The initial composition is 0.11 CO, 0.61 H<sub>2</sub>, and 0.28 CH<sub>4</sub> in molar fractions. The aim of this work is to bring the purity of hydrogen to 0.99 in molar fraction and implement controllers that can maintain the desired purity even in the presence of the disturbances that occur in the PSA process. The controller design (discrete PID and state feedback control) was based on the Hammerstein–Wiener model, which had an 80% fit over the rigorous PSA model. Both controllers were validated on a virtual plant of the PSA process, showing great performance and robustness against disturbances. The results obtained show that it is possible to follow the desired trajectory and attenuate double disturbances, while managing to maintain the purity of hydrogen at a value of 0.99 in molar fraction, which meets the international standards to be used as a biofuel.

Keywords: state feedback control; pressure swing adsorption; hydrogen purification; biogas

**MSC:** 03B70

## 1. Introduction

Biogas, an alternative and less-polluting fuel, is a resource that has great potential to replace fuels that have an impact on climate change, since it is a renewable product that can be obtained from alternative processes that use Third-Generation Biomass (3G). In general, biogas production is a decentralized and low-carbon energy option that can help mitigate and reduce climate impact. This clean energy can be generated from organic waste [1–4].

This biogas (biohydrogen) can be used as a fuel, but for this, it must have a low content of nitrogen, methane, CO, CO<sub>2</sub>, and other elements. There are various processes that can purify and produce 99% biohydrogen, one of them being PSA [5–9].



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The PSA process has two stages, production and regeneration, and these stages are discretized into certain steps; these are: adsorption, depressurization, purge, equalization, and repressurization. PSA presents an oscillatory nature generated by pressure changes, giving a highly nonlinear model represented by Partial Differential Equations (PDEs) [10-14]. The PSA process is cyclical until reaching the Cyclic Steady State (CSS), delivering high hydrogen purity results from 97% to 99%. This process includes the variables of temperature, pressure, flow, speed, and concentration. There are other ways to model and optimize the PSA process, some of these techniques being through Artificial Neural Networks (ANNs). ANNs are capable of completing nonlinear actions and parallel computation, due to the different structures that can be implemented. For this case, ANNs are given the task of improving performance in the PSA process in terms of predicting the adsorption isotherms, optimizing the times of each process step, obtaining optimal values for the depression change values and temperature, and optimizing the calculation of the highest production of a certain mixture [15-19]. However, this rigorous model represented by PDEs is not suitable for the controller design due to the cyclical nature, and for this reason, it is necessary to identify a reduced model that captures the important dynamics of the PSA process and can be used to design controllers that mitigate or eliminate certain disturbances that occur in the PSA process that alter the purity of hydrogen obtained as a final product. There are very few works on the control of the PSA process oriented toward the production of hydrogen, but the control strategy has great relevance for the production of hydrogen fuel.

The work in [20] presented a model predictive controller for the PSA process. They identified a reduced model in state space, and their results showed that MPC is better than the PI controller under disturbances, with a recovery of 61% in different scenarios with unwanted inputs. Likewise, in [21], they developed a mathematical model of the PSA process applied to the production of hydrogen. They studied adsorption dynamics under disturbances and different regions of admissible operating modes. They applied an adaptive control for the PSA cyclic process. The proposed strategy is able to attenuate certain disturbances in the input such as pressure and temperature or increase the composition of the other elements. In [22], they developed a hybrid controller of a PSA process for hydrogen purification. The developed a Local Reduced-Order Model (LROM). The proposed controller system had great performance in the presence of external disturbances, which was presented in the simulation of a two-bed, six-step PSA process for H<sub>2</sub> purification.

There are other relevant works on the control of the PSA process, but they are focused on the production and purification of ethanol, methane, or oxygen, one of the works being [23], who obtained a reduced model by using identification systems. They used the Hammerstein–Winner model to design the MPC. The control used was able to attenuate disturbances and follow the desired trajectories. The result obtained was an ethanol purity of 99% wt, and this purity allows the ethanol to be used as a fuel or oxygen additive. In [24], they designed a Fault-Tolerant Control (FTC), which allows eliminating faults in the actuators or sensors of the PSA process for the production of ethanol. The achievements obtained in this work made a great contribution toward determining the FTC to be better than a fuzzy PID controller, allowing maintaining the desired purity in the face of constant and variable faults. In [25], they presented the design and optimized study of a PSA process for the recovery of CH<sub>4</sub>. The optimal control they used managed to synthesize the optimal PSA cycle and could generate a CH<sub>4</sub> recovery of up to 97.30%. Likewise, in the work of [26], they presented a control,  $H_{\infty}$ , applied to a PSA for oxygen production. The control they designed followed the trajectories of oxygen purity and rejected disturbances in the input composition. The controller design was based on a Hammerstein model that approximated the cyclic process. This controller demonstrated better robustness than PI control.

It is of great importance to have controllers implemented in this type of process due to the number of variables that influence it and its cyclical nature, which determines the nonlinear behavior.

The main contributions of this work focus on two important points: The first is to find a simplified model from the input and output data of the PSA process that can be used for the design of controllers. The second contribution is to implement and validate the controllers of the PSA process, with the aim of rejecting external disturbances and maintaining the desired purity, complying with international standards of hydrogen purity to be used as a biofuel.

The structure of this work is as follows. In Section 2, the description and simplifications of the mathematical model represented by EDP that describes the dynamic behavior of the PSA process are presented. In Section 3, the simulations performed with the PDEs and initial and boundary conditions are given; in this section, the molar fraction and pressure profiles are described as a function of the time and length of the beds, and an analysis of the sensitivity in defining the control loops is given. In Section 4, the identification of a reduced model (Hammerstein–Wiener) capable of following the dynamics of the PSA process for hydrogen production is carried out. Section 5 describes the design of the state feedback controller, which was implemented in the identified model and validated in the rigorous model of the PSA process, and Section 6 shows the results, demonstrating the trajectory tracking and disturbance rejections.

## 2. Description of the PSA Model

A structure of 2 columns connected to 10 valves was implemented, and the process uses 4 steps (adsorption, depressurization, purge, and repressurization) (see Figure 1). It has as the feed a mixture composed of 0.11 CO, 0.61 H<sub>2</sub>, and 0.28 CH<sub>4</sub> of a molar composition and uses a pressure of 980 kPa and a temperature of 298.15 K [27–30].

In order to establish the PSA model for the production and purification of hydrogen, it was necessary to make some considerations, which are shown below:

- (1) There are no reactions between the elements of the mixture  $(H_2, CH_4, CO)$ .
- (2) The mass transfer coefficient was considered constant.
- (3) The gaseous phase is only convective, and axial dispersion was considered negligible.
- (4) The energy balance was considered to be isothermal.
- (5) The adsorption equilibrium was modeled by the Langmuir model, expressed in terms of partial-pressure-dependent parameters.

From the assumptions mentioned above, a summary of the equations used in this work to determine the cyclical behavior of the PSA process is made. The model was obtained from the works [28,29,31]:

Mass balance for gas phase:

In this Equation (1), the terms of the axial dispersion, convection, and accumulation are shown.

$$-D_L \frac{\partial^2 c_i}{\partial z^2} + \frac{\partial (U_z c_i)}{\partial z} + \frac{\partial c_i}{\partial t} + \frac{(1 - \epsilon_b)}{\epsilon_b} J = 0$$
(1)

The flow over the solid surface is given as:

$$J = -\rho \frac{\partial W_i}{\partial t} \tag{2}$$

• Gas phase energy balance:

In this Equation (3), the solid phase enthalpy accumulation, solid thermal conduction, heat of adsorption, heat of the adsorbed phase, and gas–solid heat transfer are included in the solid phase energy balance.

$$K_L \frac{\partial^2 T}{\partial z^2} + \left[\epsilon_b c C_{pg} + (1 - \epsilon_b) \rho_p C_{ps}\right] \frac{\partial T}{\partial t} - \epsilon_b c C_{pg} U_z \frac{\partial T}{\partial z} = (1 - \epsilon_b) \rho_p \sum_{j=1}^N \Delta H_j \frac{\partial n_j}{\partial t} + \frac{2h_{in}}{R_{in}} (T_w - T)$$
(3)

## • Momentum balance:

The momentum balance is a combination of the Karman–Kozeny and Burke–Plummer equations. It is shown in Equation (4).

$$-\frac{\partial P}{\partial z} = \left(\frac{150\mu V_z (1-\epsilon_b)^2}{4R_p^2 \epsilon_b^3} + \frac{1.75\rho_g (1-\epsilon_b)}{(2R_p)\epsilon_b^3} v_g^2\right)$$
(4)

• Langmuir equation:

This isotherm is a function of partial pressure. This equation is shown in (5).

$$W_i^* = \frac{(IP_1 - IP_2T_s)IP_3e^{ip_4/T_s}P_i}{1 + \sum_i (IP_3e^{ip_4/T_s}P_i)}$$
(5)

Kinetic model:

The mass transfer driving force is shown in Equation (6)

$$\frac{\partial W_i}{\partial t} = MTC_{si}(W_i^* - W_i) \tag{6}$$

where:

$$MTC_{si} = \frac{\Omega D_{ei}}{r_n^2} \tag{7}$$

 $\Omega$  is the parameter in the Glueckauf expression. The initial and boundary conditions are shown in Table 1.

Table 1. Boundary and initial conditions.

I. Adsorption	
t = 0	$c_{H_2}(z,0) = c_0, c_{CO}(z,0) = 0, c_{CH_4}(z,0) = 0, c_{CO_2}(z,0) = 0$ $T(z,0) = T_0, T_w(z,0) = T_0, p(z,0) = p_0, \eta_i(z,0) = \eta_i^*$
z = 0	$ \begin{array}{l} -D_L \frac{\partial c_i}{\partial z} = u[c_i(0^-,t) - c_i(0^+,t)], \ p = p_0, \ U_z = U_{zo} \\ -K_L \frac{\partial I}{\partial t} = \varepsilon_b c C_{pg} U_z[T(0^-,t) - T(0^+,t)] \end{array} $
z = L	$-D_L rac{\partial c_i}{\partial z} = 0, \; rac{\partial U_z}{\partial z} = 0, \; -K_L rac{\partial T}{\partial t} = 0, \; rac{\partial p}{\partial z} = 0$
II. Depressurization	
t = 0	$c_{H_2}(z,0) = c_0^{(I)}, \ c_{CO}(z,0) = c_{CO}^{(I)}, \ c_{CH_4}(z,0) = c_{CH_4}^{(I)}, \ c_{CO_2}(z,0) = c_{CO_2}^{(I)}$
1 – 0	$T(z,0) = T^{(I)}, T_w(z,0) = T^{(I)}, p(z,0) = p^{(I)}, \eta_i(z,0) = \eta_i^{*(I)}$
z = 0	$rac{\partial_{c_i}}{\partial z}=0, \ rac{\partial p}{\partial z}=0, \ rac{\partial U_z}{\partial z}=0, \ rac{\partial T}{\partial t}=0$
z = L	$\frac{\partial_{c_i}}{\partial z} = 0, \ \frac{\partial U_z}{\partial z} = 0, \ \frac{\partial T}{\partial t} = 0, \ \frac{\partial p}{\partial z} = 0, \ F = F_{valve}$
III. Purge	
	$c_{H_2}(z,0) = c_0^{(II)}, c_{CO}(z,0) = c_{CO}^{(II)}, c_{CH_2}(z,0) = c_{CH_2}^{(II)}, c_{CO_2}(z,0) = c_{CO_2}^{(II)}$
t = 0	$T(z,0) = T^{(II)}, \ T_m(z,0) = T^{(II)}, \ p(z,0) = p^{(II)}, \ \eta_i(z,0) = \eta_i^{*(II)}$
z = 0	$\frac{\partial c_i}{\partial z} = 0, \ \frac{\partial p}{\partial z} = 0, \ \frac{\partial U_z}{\partial z} = 0, \ \frac{\partial T}{\partial t} = 0, \ F = F_{valve}$
z = L	$rac{\partial c_i}{\partial z}=0, \ rac{\partial U_z}{\partial z}=0, \ rac{\partial T}{\partial t}=0, \ rac{\partial p}{\partial z}=0$
IV. Repressurization	
	$c_{H_2}(z,0) = c_0^{(III)}, c_{CO}(z,0) = c_{CO}^{(III)}, c_{CH_4}(z,0) = c_{CH_4}^{(III)}, c_{CO_2}(z,0) = c_{CO_2}^{(III)}$
t = 0	$T(z,0) = T^{(III)}, T_w(z,0) = T^{(III)}, p(z,0) = p^{(III)}, \eta_i(z,0) = \eta_i^{*(III)}$
z = 0	$\frac{\partial_{c_i}}{\partial z} = 0, \ \frac{\partial p}{\partial z} = 0, \ \frac{\partial U_z}{\partial z} = 0, \ \frac{\partial T}{\partial t} = 0, \ F = F_{valve}$
z = L	$rac{\partial \widetilde{c_i}}{\partial z}=0, \ rac{\partial \widetilde{U_z}}{\partial z}=0, \ rac{\partial \widetilde{T}}{\partial t}=0, \ rac{\partial \widetilde{p}}{\partial z}=0$

The operating conditions were defined considering as a reference the processes reported by [28,31]. To purify and produce hydrogen, the use of activated carbon was considered. The schematic diagram of the PSA process is shown in Figure 1. This schematic presents the execution times of each step and includes 10 valves to control the flow in both directions through the beds.

The PSA process uses pressure, temperature, flow, and composition as the inputs (see Figure 1). For this reason, each of the expressed equations represents a solution to each variable. For the material balance, the purity results of each element can be obtained, either in concentration or molar fraction; in the moment balance, the pressure profiles that exist within the beds can be observed, determining if there are pressure drops, which can occur when changing from one step to another. For the material balance, the temperature increases within the bed can be known. It is important to observe this variable since the temperature must be kept stable so as not to affect the adsorption of the molecules or atoms. The kinetic and thermodynamic model determines the number of molecules that have been adsorbed and how quickly they reach their saturation points.



Figure 1. Scheme of the PSA process for the purification and production of hydrogen.

#### 3. Simulation of the PSA Process

The simulations presented in this work were based on the start-up of the process and the CSS. Figure 2 shows the purity profiles of CO, methane, and hydrogen in molar fractions. It was observed that the CO and methane decreased, resulting in pure hydrogen. The hydrogen purity levels were 97% and were reached in a time of 4380 s, which is equivalent to 30 cycles. After this time, the CSS was reached, and the pressure, temperature, and composition profiles showed an oscillatory repetitive dynamics.

The pressure profiles as a function of time are shown in Figures 3 and 4. The pressure profile presented an oscillatory dynamics due to the pressure changes that exist within the packed columns, and these oscillations ranged from 9.8 bar to 1 bar since, by raising the adsorption pressure, a greater number of CO and methane molecules are adsorbed. For the

regeneration of the bed, it is necessary to depressurize and purge, and for this reason, it is necessary to lower the pressure to 1 bar.



**Figure 2.** Purity of the CO, methane, and hydrogen obtained from the start-up of the PSA process until reaching the CSS.



**Figure 3.** Pressure profiles as a function of time obtained from the start-up of the PSA process until reaching the CSS.

When the CSS is reached after 30 cycles, the profiles tend to be cyclic, but stable. Figure 4 shows the 3D pressure profiles as a function of the length (nodes) of the bed with one completed cycle (146 s). These profiles show how each step of the PSA process is contemplated, resulting in a pressure swing of 9.8 bar until a purge that is generated for 3 s to reach 1 bar, then it is pressurized until 9.8 bar, which is needed again to purify and produce hydrogen.

At the end of the simulation of the PSA process from the start-up to reaching the CSS, which is achieved after 30 cycles (4380 s), it is necessary to perform a sensitivity analysis to define the primary control loop, and this must be done in the CSS, with changes in the possible input variables (manipulated variables) that can affect the purity of the hydrogen (controlled variables); these variables are: pressure, temperature, composition, and feed flow.



Figure 4. Pressure profiles in the CSS.

## Sensitivity Analysis of the PSA Process

After reaching the CSS, a sensitivity analysis was established with the possible input variables that can manipulate the purity of the hydrogen (controlled variables). The input variables are pressure, temperature, flow, and feed composition. The changes made to these variables were  $\pm 0.5$ , resulting in certain effects on the output variable (hydrogen purity). The results can be seen in Figure 5 and Table 2.

In Figure 5, the effects of the possible input variables can be observed. In the case of temperature, we can see the behavior of the hydrogen purity obtained from changes in temperature in the feed, and these changes that occur have a great effect compared to the other input variables.

The other variable that can generate a change is the feed composition (see Figure 5g,h), which could be considered for a secondary control loop.

In the case of pressure and feed flow, it can be seen that both have little effect on the purity of the hydrogen, but they can be considered as possible unwanted inputs (disturbances) that can affect the PSA process.

From the results shown in Table 2, it can be determined how the primary control loop is at the feed temperature and the purity of hydrogen, which is obtained from the two beds.

After having performed the sensitivity analysis, a Pseudo Random Binary Sequence (PRBS) was constructed with the  $\pm 5\%$  feed temperature values. This input signal was introduced to the rigorous PSA model to obtain an output (purity of hydrogen). With the input and output data obtained, a reduced model was identified, which should be able to capture the important dynamics of the PSA process for hydrogen production.

Run	Temperature (K)	Flow kmol $h^{-1}$	Pressure kPa	Composition Hydrogen-Methane-CO	Purity % Molar Fraction of Hydrogen	Number of Cycles
1	298.15	0.162	980	0.61-0.28-0.11	94.85	30
2	283.24	0.162	980	0.61-0.28-0.11	98.01	2
3	313.05	0.162	980	0.61-0.28-0.11	90.02	2
4	298.15	0.153	980	0.61-0.28-0.11	94.95	1
5	298.15	0.170	980	0.61-0.28-0.11	94.75	1
6	298.15	0.162	931	0.61-0.28-0.11	95.00	1
7	298.15	0.162	1029	0.61-0.28-0.11	94.65	1
8	298.15	0.162	980	0.6405-0.2495-0.11	94.00	2
9	298.15	0.162	980	0.5795-0.3105-0.11	95.50	2

Table 2. Sensitivity analysis to define the control loops.



**Figure 5.** Results of the sensitivity analysis to define the possible input variables that can control the purity of hydrogen. (a) Purity obtained using 283 K. (b) Purity obtained using 313 K. (c) Purity obtained using 0.153 kg h<sup>-1</sup>. (d) Purity obtained using 0.170 kg h<sup>-1</sup>. (e) Purity obtained using 931 kPa. (f) Purity obtained using 1029 kPa. (g) Purity obtained using 0.6405 hydrogen, 0.2495 methane, and 0.11 CO. (h) Purity obtained using 0.5795 hydrogen, 0.0.3105 methane, and 0.11 CO.

## 4. System Identification

The identified model that was obtained with the input and output data of the PSA process was a Hammerstein–Wiener model, and this model was obtained with the MatLab toolbox. The input and output data used are shown in Figure 6a,b.

The PRBS signal was built with the following points:

- At least 1 step must take into account the time for the hydrogen purity to converge to a new CSS.
- A percentage that generates effects on the output variable must be established.
  - A number of bits must be proposed, in such a way that they do not excite the PSA process too much, since this can generate a bad identification. However, this cannot be an input with a minimum value of bits either.

For this case, a 4 bit PRBS with a convergence time of 300 s and a variation of  $\pm 0.5\%$  was generated. Then, 300 s were chosen from the estimated time in which it reached the new CSS, and these data were obtained from the sensitivity analysis, which is shown in Figure 5a,b.



**Figure 6.** Output data obtained from the 4 bit PRBS input with a sampling time of 1 s. (**a**) Input signal generated from a 4 bit PRBS with a sampling time of 1 s. (**b**) Output signals (hydrogen purity).

Figure 7 shows how the identified model follows the dynamics of the PSA process, resulting in a fit of 80%. Since the dynamics of the PSA process is highly nonlinear with an oscillatory cyclic nature, being able to have a model above 90% is very complicated, and it becomes more complicated when one wants to obtain a linear model in the state space or transfer function; the results with these representations were below 40% in the fit. For this reason, a nonlinear model (Hammerstein–Wiener) was used, which has a better structure, which allows better approximations of the oscillatory behavior of the PSA process.



Figure 7. Comparison between the identified model and the rigorous model of the PSA process.

## Structure of the Hammerstein–Wiener Model

The Hammerstein–Wiener model is used to estimate static nonlinearities in a complex process. It is made up of three blocks: the blocks at the ends are nonlinear inputs and outputs, and these are defined as piecewise linear; the central block is a linear block, and it is the dynamic part of the PSA process.

Figure 8 shows the structured scheme of the Hammerstein–Wiener model.



Figure 8. Block diagram of the Hammerstein–Wiener model.

A detailed description of the characteristics and variables of the Hammerstein–Wiener model can be found in the work of [23].

However, to design controllers using the Hammerstein–Wiener model, new blocks representing the inverse functions of the input and output blocks were obtained (see Figure 9).



Figure 9. Block diagram of the Hammerstein–Wiener model with inverse functions.

The inverse functions of the static nonlinear input and output blocks that were obtained are shown in the following Equations (8) and (9).

$$u_{t} = \begin{cases} 3.5155w_{t} + 305.2763 & 49.993987424472000 < w_{t} \le 50.844481371683270 \\ 2.6620w_{t} + 306.6717 & 50.844481371683270 < w_{t} \le 51.967677299012884 \\ 2.3525w_{t} + 307.7555 & 51.967677299012884 < w_{t} \le 53.238642233028040 \\ 2.3110w_{t} + 308.9661 & 53.238642233028040 < w_{t} \le 54.532443188664950 \\ 2.3525w_{t} + 309.1693 & 54.532443188664950 < w_{t} \le 55.724147176214060 \\ 3.0994w_{t} + 310.2660 & 55.724147176214060 < w_{t} \le 56.688821214343890 \end{cases}$$
(8)

	$0.00012y_t - 11.55$	$0.965337951237799 < y_t \le 0.975337951237799$	
× )	$160.551y_t - 182.085$	$0.975337951237799 < y_t \le 0.991127869506668$	(0)
$x_t - y$	$-539.8071y_t + 512.0845$	$0.881994750982603 < y_t \le 0.965337951237799$	(9)
	$-302.4829y_t + 302.7657$	$0.766910025770996 < y_t \le 0.881994750982603$	

In the case of the linear block model, the transfer function was converted to state space representation, since the controller was designed to work in  $\dot{x} = Ax + Bu$ , y = cx. The model was as follows (Equation (10)):

	2.930	-1.446	0.504	0.085	-0.334	0.337	-0.124	0.007	0.039	-0.113	
A =	2	0	0	0	0	0	0	0	0	0	
	0	1	0	0	0	0	0	0	0	0	
	0	0	1	0	0	0	0	0	0	0	
	0	0	0	1	0	0	0	0	0	0	
	0	0	0	0	1	0	0	0	0	0	'
	0	0	0	0	0	1	0	0	0	0	(10)
	0	0	0	0	0	0	1	0	0	0	
	0	0	0	0	0	0	0	0.500	0	0	
	0	0	0	0	0	0	0	0	0.250	0	
C = [	-0.340	0.500	-0.490	0.160	0 0	0 0	0 0],				
B = [	1 0	0 0 0	0 0	0 0	$0 ]^{ op}$						

where  $x \in \mathbb{R}^n$ ,  $u \in \mathbb{R}^m$ ,  $y \in \mathbb{R}^p$ , *k* represent the states, the inputs of control, the system outputs, and the particular sampling instant (1 s).

After having obtained the reduced model, we continued to design the controller on the Hammerstein–Wiener model using the inverse functions of the input and output blocks and the linear block.

## 5. Discrete State Feedback Controller Design

In this section, the controller design for the PSA process is explained. The scheme of the controller system is illustrated in Figure 10. Note that the unmeasured states were estimated using a discrete Luenberger observer. A discrete state variable feedback controller was designed such that the molar fraction of hydrogen followed the desired reference.



Figure 10. Discrete state variable feedback controller scheme applied to the PSA system.

The continuous system (Equation (10)) was converted into a discrete dynamics by using the Euler method. Then, the following system was obtained:

$$\begin{aligned} x(k+1) &= A_d x(k) + B_d u(k), \\ y(k) &= C_d x(k), \end{aligned} (11)$$

where  $A_d$ ,  $B_d$ , and  $C_d$  are matrices with proper dimensions. An integrator comparator was used in order to reduce the steady-state error by defining the error dynamics  $e(k) = r_f(k) - C_d x(k)$  as follows:

$$z(k+1) = z(k) + T_s(e(k)) = z(k) + T_s(r_f(k) - C_d x(k)),$$
(12)

where  $T_s$  denotes the sample time and z(k) is an auxiliary variable for integral control design. Then, System (11) and (12) is rewritten in an augmented system form  $\bar{x}(k) = [x(k), z(k)]^{\top}$  as follows:

$$\bar{x}(k+1) = \bar{A}_c \bar{x}(k) + \bar{B}_c u(k) + \bar{R}_c r_f(k) 
\bar{y}(k) = \bar{C}_c \bar{x}(k),$$
(13)

with

$$\bar{A}_{c} = \begin{bmatrix} A_{d} & 0 \\ -C_{d}T_{s} & 1 \end{bmatrix}, \bar{B}_{c} = \begin{bmatrix} B_{d} \\ 0 \end{bmatrix},$$
$$\bar{C}_{c} = \begin{bmatrix} C_{d} & 0 \end{bmatrix}, \bar{R}_{c} = \begin{bmatrix} 0 \\ T_{s} \end{bmatrix}.$$

If the pair  $(\bar{A}_c, \bar{B}_c)$  in (13) is controllable, then the control law u(k) is calculated as:

$$u(k) = -\bar{K}_c \bar{x}(k) = -\begin{bmatrix} K_1 & K_2 \end{bmatrix} \begin{bmatrix} x(k) \\ z(k) \end{bmatrix},$$
(14)

The controller gains matrices are  $K_1$  and  $K_2$ . The optimal controller gains can be calculated by substituting the control input (14) into the linear system (13), resulting in the following closed-loop system:

$$\bar{x}(k+1) = (\bar{A}_c + \bar{B}_c \bar{K}_c) \bar{x}(k).$$
 (15)

The global asymptotic convergence of the controller is guaranteed if matrices  $X = X^{\top} > 0$  and *W* exist, such that

$$\begin{bmatrix} X & \bar{A}_c X + \bar{B}_c W \\ (\bar{A}_c X + \bar{B}_c W)^\top & X \end{bmatrix} > 0.$$
(16)

The controller gain matrix is obtained by  $\bar{K}_c = WX^{-1}$ , and the proof can be consulted in [32]. Usually, the linear matrix inequality (16) produce slows dynamics. This problem was solved using disk-stability with the eigenvalues of  $(\bar{A}_c - \bar{B}_c \bar{K}_c)$  located inside the unit circle, as defined in [32,33]. The area associated with the decay rate consisted of a circle centered at the origin, with its radius being the parameter, which was defined by the desired decay rate.

Then, in order to avoid slow dynamics, (16) was replaced with (17), by considering that the close-loop error system (12) is asymptotically stable if there exists a positive scalar r, a symmetric matrix  $X = X^{\top} > 0$ , and a matrix W, such that

$$\begin{bmatrix} -rX & \bar{A}_c X + \bar{B}_c W\\ (\bar{A}_c X + \bar{B}_c W)^\top & -rX \end{bmatrix} < 0.$$
(17)

#### 6. Results

As a result, two scenarios related to the implementation of the controllers on the Hammerstein–Wiener model are presented in order to tune the parameters and two scenarios for the validation of the controllers applied to the PSA process.

In the first scenario, the discrete PID control scheme is presented (see Figure 11), applied to the Hammerstein–Wiener model. The PID controller gains were tuned using the Ziegler–Nichols method.



Figure 11. Discrete PID control implementation on the Hammerstein–Wiener model.

In the second scenario, the state feedback control scheme is presented (see Figure 12), applied to the Hammerstein–Wiener model. The controller gain, from (17), was solved using the Yalmip Toolbox [34], obtaining the following results:

$$K_c = [-5.52, 14.66, -35.86, 53.97, -55.11, 39.01, -18.87, 5.16, -2.20, 0.64, -0.01].$$
(18)



Figure 12. Implementation of state feedback control on the Hammerstein–Wiener model.

In these two scenarios, four simulations were carried out to tune and observe the effort of the controllers when making reference changes, achieving the control of the purity of hydrogen obtained as the final product.

In Figure 13, a trajectory is established with three changes, the changes in purities being from 0.95 to 0.6, 0.6 to 0.75, and 0.75 to 0.95. This was done to show that both controllers were following the desired trajectory. However, there were some control signals that generated peaks that altered the desired purity. In the case of the discrete PID controller, it can be seen that it had a better follow-up in the face of these three changes. On the other hand, state feedback control followed the path, but generated control signals with small increments at the end of each trajectory change.

In Figure 14, the simulations are given to verify that the hydrogen purity can also be increased, contrary to the simulations shown in Figure 13. In these simulations, it can be shown that both controllers worked correctly and followed the desired trajectory until obtaining a higher purity. For this, the controller signals by the state feedback were better than the PID control signals, since they followed the path with a lower percentage of error compared to the PID control. It was possible to achieve a purity from 0.949 to 0.96 and from 0.96 to 0.949.

For the tests of the four trajectory changes (see Figure 15), the results showed that both controllers had good tracking of the desired purity; however, in the case of control by the state feedback, it continued to have problems in the changes of purity from 0.97 to 0.6 and 0.725 to 0.97, and it can be seen how there were increases that generated an error; consequently, increases and decreases in hydrogen purity were generated.



**Figure 13.** Control signals to reach different purity values and verify the performance of the controllers. (a) Control signals generated before the block of inverse input functions. (b) Control signals generated using the input inverse functions' block. (c) Hydrogen purity obtained from 3 trajectory changes.



**Figure 14.** Control signals to reach different purity values and verify the performance of the controllers. (**a**) Control signals generated before the block of inverse input functions. (**b**) Control signals generated using the input inverse functions block. (**c**) Hydrogen purity obtained from 2 trajectory changes.

(a)



(b)

**Figure 15.** Control signals to reach different purity values and verify the performance of the controller. (a) Control signals generated before the block of inverse input functions. (b) Control signals generated using the input inverse functions block. (c) Hydrogen purity obtained from 4 trajectory changes.

For the following two scenarios, the two control schemes were established to validate the results using the virtual plant of the PSA process, which is represented by the PDEs. The schemes are shown in Figures 16 and 17. Both controllers worked in discrete time, since the PSA process established a sampling time of 1 s to be able to obtain results in each step (adsorption, depressurization, purge, and depressurization) of the PSA process.



Figure 16. Discrete PID control implementation for the PSA process.



Figure 17. Validation of state feedback control in the PSA process.

For the validation of the controllers applied to the PSA process, four simulations are presented that showed the trajectory changes and the presence of disturbances. These tests are shown in Figures 18–21.



**Figure 18.** Validation of the controllers from a change of trajectory. (**a**) Purity obtained with trajectory tracking from the control signals generated by the discrete PID. (**b**) Purity obtained with trajectory tracking from the control signals generated by state feedback. (**c**) Discrete PID and status feedback control signals.

For the first case, a reference change from 0.95 to 0.98 was observed. Both controllers generated the necessary control signal so that the hydrogen purity rose and converged to 0.98. However, it can be seen that the state feedback control had a faster response in 900 s; on the other hand, the PID control had a delay of 1500 s, and the effects can be shown in the hydrogen purity values. It should be noted that most hydrogen purity results will have an oscillatory response due to the nature of the PSA process.

Figure 19 shows the hydrogen purity profiles and control signals from the implementation of a perturbation. These tests were performed to observe the robustness of the controllers against unwanted inputs. The disturbance appeared at 15 s, and it was generated from the changes that the feed composition presented; for this case, it was modified from 0.11 CO, 0.61 H<sub>2</sub>, and 0.28 CH<sub>4</sub> to 0.11 CO, 0.488 H<sub>2</sub>, and 0.402 CH<sub>4</sub>, resulting in a -20% disturbance applied in the feed composition.



**Figure 19.** Validation of the controllers from a change of trajectory and the presence of a disturbance. (a) Purity obtained with trajectory tracking and attenuation of a disturbance from the control signals generated by the discrete PID. (b) Purity obtained with trajectory tracking and attenuation of a disturbance from the control signals generated by state feedback. (c) Discrete PID and status feedback control signals.

It can be seen how both controllers managed to attenuate the disturbance and made it converge again to the desired value. However, the discrete PID controller presented a higher control signal and had a purity drop of 0.88 in mole fraction. On the other hand, the state feedback control presented a better performance on the disturbance since it only decayed up to 0.91. The control signal it presented had small temperature values compared to the discrete PID signals.

In Figure 20, a case with two applied disturbances is presented in order to see the performance and robustness of the controllers. The perturbations were feed composition changes of -20% (0.11 CO, 0.61 H<sub>2</sub>, and 0.28 CH<sub>4</sub> to 0.11 CO, 0.488 H<sub>2</sub>, and 0.402 CH<sub>4</sub>) and production pressure changes of 20% (980 kPa to 1176 kPa).



**Figure 20.** Validation of the controllers from a change of trajectory and the presence of two disturbances. (a) Purity obtained with trajectory tracking and attenuation of two disturbances from the control signals generated by the discrete PID. (b) Purity obtained with trajectory tracking and attenuation of two disturbances from the control signals generated by state feedback. (c) Discrete PID and status feedback control signals.

It can be seen that both controllers attenuated the disturbances presented and managed to follow the desired trajectory, fulfilling the purpose of each controller. However, the discrete PID control presented very high control signals compared to the state feedback control, and purity values below 0.9 in the molar fraction were observed. On the other hand, the state feedback control had a control action with better performance and robustness since it did not allow the purity to decrease from the value of 0.91 in molar fraction, and this applied to both disturbances presented (see Figure 20).

In the last test, two disturbances similar to those shown in Figure 20 and two reference changes were performed, taking it from 0.94 to 0.98 and from 0.98 to 0.99 in molar fraction. It can be seen in Figure 21 that both controllers performed adequately since they again achieved reducing the disturbance and followed the desired purity. It is important to mention that the purity achieved (0.99 in molar fraction) at the end of the process, complied with international standards to be used as a biofuel (biohydrogen).

The control signals generated by the discrete PID were high-temperature values, but the response signal was very soft at the moment of achieving the purity of 0.99 in mole fraction. On the other hand, the control signals by state feedback continued to obtain better performance and robustness in the face of disturbances and changes in trajectory. However, in the last trajectory change, an abrupt control signal can be observed, resulting in a less smooth hydrogen purity response when obtaining the value of 0.99 in molar fraction.



**Figure 21.** Validation of the controllers from two changes of trajectory and the presence of two disturbances. (**a**) Purity obtained with two trajectory tracking and attenuation of two disturbances from the control signals generated by the discrete PID. (**b**) Purity obtained with two trajectory tracking and attenuation of two disturbances from the control signals generated by state feedback. (**c**) Discrete PID and status feedback control signals.

#### 7. Conclusions

In this work, a PSA process for the production of hydrogen was presented with the objective of purifying and separating the elements and obtaining a biohydrogen that meets international standards to be used as a biofuel. The importance of this work focused on the implementation and validation of controllers applied to the PSA process in order to control the purity of hydrogen and mitigate the disturbances that may occur at the input of the process. In the Results Section, the different case studies that were performed to validate the robustness and performance of the controllers (discrete PID and state feedback control) were shown. It can be concluded that both controllers followed the desired purity and managed to attenuate up to two existing disturbances in the process input. However, in the case of the discrete PID, it can be seen that it generated higher control signals compared to the state feedback control, but the effects on hydrogen purity tended to have smooth profiles at the time the changes were generated of the trajectories or in the attenuation of the disturbances. On the other hand, the state feedback control presented control signals with small values compared to the discrete PID, but the effects on the hydrogen purity tended to have overshoots before the changes in trajectories and in the presence of the disturbances. As a final conclusion, it is possible to point out that the state feedback control had better performance and robustness since the purity values reached in the presence of disturbances were of a lower value (0.91 in molar fraction) compared to the purity decreases that they generated with the use of the discrete PID (0.88 in molar fraction). Both controllers managed to obtain a hydrogen purity of 0.99 in molar fraction, which is a purity that meets international standards and can mitigate disturbances in that purity value.

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## Nomenclature

$a_p$	Specific particle surface ( $m^2 m^{-3}$ )
<i>c</i> <sub>i</sub>	Concentration (kmol $m^{-3}$ )
$C_{ps}$	Heat capacity of adsorbent (MJ kmol <sup><math>-1</math></sup> K <sup><math>-1</math></sup> )
$C_{pq}$	Heat capacity of gas phase (MJ kmol <sup><math>-1</math></sup> K <sup><math>-1</math></sup> )
$C_{pai}$	Specific heat capacity of the adsorbed phase (MJ kg <sup><math>-1</math></sup> K <sup><math>-1</math></sup> )
$d_b$	Bed diameter (m)
$d_p$	Particle diameter (m)
$D_L$	Axial dispersion (m <sup>2</sup> s <sup><math>-1</math></sup> )
$H_s$	Heat transfer coefficient (Js <sup><math>-1</math></sup> m <sup><math>-2</math></sup> K <sup><math>-1</math></sup> )
$\Delta H_s$	Heat adsorption gradient (J mol $^{-1}$ )
$IP_{1i}, IP_{2i}, IP_{3i}, IP_{4i}$	Isothermal parameters of component i
Ķ	Langmuir isotherm equilibrium constant ( $Pa^{-1}$ )
M	Molar weight (kg mol <sup>-1</sup> )
$MTC_s$	Mass transfer coefficient solid (s $^{-1}$ )
Р	Pressure (Pa)
Q	Isosteric heat of adsorption (J mol $^{-1}$ )
R	Universal gas constant (J mol $^{-1}$ K $^{-1}$ )
t	Time (s)
$T_g$	Gas temperature (K)
$T_s$	Solid temperature (K)
Т	Temperature (K)
$v_g$	Surface gas velocity (m s <sup><math>-1</math></sup> )
W <sub>i</sub>	Adsorbed amount of the component i (kmol $kg^{-1}$ )
$W_i^*$	Adsorbed equilibrium amount of component i (kmol $kg^{-1}$ )
$y_i$	Molar fraction of the gas component i
Z	Coordinate of axial distance (m)
Greek symbols	
$\epsilon_b$	Bed porosity
$\epsilon_p$	Particle porosity
μ	Dynamic viscosity (m s $^{-1}$ )
$ ho_b$	Bed density (kg m $^{-3}$ )
$ ho_p$	Particle density (kg m $^{-3}$ )

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