SIMULATION AND ANALYSIS OF A HYDROGEN GENERATION UNIT

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ABSTRACT

High Purity Grade Hydrogen is an important compound used in oil refineries due to its capacity for withdrawing sulfur impurities from fuels. This feedstock is commonly produced in Hydrogen Generation Units through a Steam Methane Reforming process. Due to the high-grade requirements in Hydrotreatment processes, a hydrogen purification step is mandatory, often conducted in batteries of adsorption columns, known as Pressure Swing Adsorption (PSA). The main objective of this paper is to present the simulation of a Hydrogen Generation Unit, focusing on the PSA process modeling approach. ASPEN PLUS[™] was utilized in conjunction with MATLAB[®] to model the whole process. The link between both simulators was established through VBA (Visual Basic for Applications) macros developed in Excel. This two-way connection did not affect the results obtained by the simulation of the whole process unit, as it was carried out successfully and was able to represent the global mass and energy balances satisfactory, obtaining hydrogen with 99.8% purity.

KEYWORDS

hydrogen purification; pressure swing adsorption (PSA); processes simulation; ASPEN PLUS[™]; MATLAB[®]

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1. INTRODUCTION

Hydrogen gas plays a fundamental role in sulfur impurities removal processes from oil derivate products, such as gasoline and diesel. Steam reforming of natural gas, or steam methane reforming (SMR), is the most common method of hydrogen production on a large scale in oil refineries (Meyers, 2003). In this process, a mixture of steam and natural gas, (or in certain situations, naphtha) substitutes methane (Fahim et al., 2010) and reacts at a high temperature in the presence of a catalyst, forming a mixture of carbon dioxide and hydrogen, according to the following equations (Fahim et al., 2010):

Before shipping the hydrogen produced by this reforming process to consumers, the hydrogen is sent to a purification unit that removes any unconverted methane and steam along with carbon monoxide and carbon dioxide (Biswas et al., 2010).

The impurities are adsorbed by charging a column (containing the adsorbent) with the gas mixture and, then, raising its inside pressure until it reaches a value that is high enough to cause the gases to be adsorbed. Hydrogen is not adsorbed as the impurities are pulled from the gas stream. When the column pressure is reduced to atmospheric pressure levels, the column is

evacuated countercurrentwise, so as to withdraw the impurities adsorbed in it. The present operation is particularly advantageous when a very high level of purified hydrogen is required (Fahim et al., 2010).

1.1 Operation of PSA column

The PSA adsorption process is based on the modulation of the internal pressure of the column vessel. The pressure swings that occur during an operation cycle determine the degree of retention of each gas by the adsorption bed inside the column, as well the level of impurities inside the column. In general, a single PSA vessel goes through five elementary steps along an operation cycle, as shown in the Fig. 1 (Fahim et al., 2010).

The regeneration step of the adsorption bed after its saturation is always essential, as the PSA cycle is a batch process. The vessels in a PSA unit operate simultaneously under different pressures and physical conditions, depending on which step each column is in the moment (Fahim et al., 2010). More information about the cycle steps is available in Fahim et al. (2010).

1.2 Mathematical modeling of a single PSA column

The dynamic behavior of PSA columns results from the interaction of mass balances, mass transfer, and adsorption equilibrium. For that reason, the mathematical modeling of such systems requires corresponding models for each of these items.

Figure 1. Elementary steps of a PSA cycle operation.

The mass balance for a one-dimensional flow in PSA systems can be written as shown in Eq. 4.

$$\frac{\partial y_i}{\partial t} + u \frac{\partial y_i}{\partial z} + \rho_p \frac{RT(1-\varepsilon)}{P} \frac{\partial \bar{q}_i}{\partial t} = 0 \quad (4)$$

The mass transfer between gas and solid phases, represented by the term $\partial \bar{q}_i / \partial t$, is described by the linear driving-force model (LDF), given by Eq. 5.

$$\frac{\partial \bar{q}_i}{\partial t} = k_i(q_i^* - q_i) \quad (5)$$

This model assumes that the rate of mass transfer is directly proportional to the mass transfer driving-force, namely, the difference between the actual concentration of adsorbed gas in the solid phase and the theoretical concentration that would exist under equilibrium conditions (Biswas et al., 2010; Huang et al., 2008).

The relation between the amount adsorbed of a given compound and the total pressure at a fixed temperature is called an adsorption isotherm. There are many isotherm models and one of the most common is the Langmuir single-site model (Ruthven et al., 1994), which is the model chosen for this paper and presented in Eq. 6.

$$q_i^* = \frac{q_i^{sat} b_i P_i}{1 + \sum_{j=1}^n b_j P_j} \quad (6)$$

The terms q_i^{sat} and b_i , from Eq. 6, are obtained by empirical correlations represented by Eq. 7 and 8. The Langmuir parameters $a_{1,i}$, $a_{2,i}$, $b_{0,i}$ and $b_{1,i}$ are specific for each gas component and for each adsorbent bed.

$$q_i^{sat} = a_{1,i} + \frac{a_{2,i}}{T} \quad (7)$$

$$b_i = b_{0,i} e^{\left(\frac{b_{1,i}}{T}\right)} \quad (8)$$

The main objective of this paper is to present the simulation of the entire Hydrogen Generation Unit. Special focus was given to the PSA modeling approach, aiming to validate it to be used in the future to evaluate the behavior of the purification process when different adsorbent materials are used. ASPEN PLUS™ was applied in conjunction with MATLAB® to model the whole process. The former was used to simulate the Steam Methane Reforming conversion, whereas the Pressure Swing Adsorption operation calculation was executed by

a customized MATLAB® module previously developed and validated.

2. MATERIALS AND METHODS

2.1 Hydrogen Production Unit (HPU) simulation

From the investigation of the basic ASPEN™ functionalities it was possible to elaborate a process flow diagram for the simulation of the hydrogen generation unit. The simulated unit had the following unit operations (equipment): flash tank, compressor, reactors, mixers, heat exchangers, and adsorption column.

It was assumed that the Hydrogen Production Unit might use either natural gas or naphtha as feed. In this study, natural gas, with molar compositions of 80% CH₄, 10% C₂H₆ and 10% C₃H₈, was used. Since steam reforming is a catalytic process, there are several components that can poison the catalyst, such as sulfur and chloride-containing compounds. As the experiments used natural gas as the unit feed, the sulfur compounds were removed by the pre-treatment (hydrodesulfurization) stage, which occurred at a relatively low pressure (from 1 to 4 MPa) when compared to the hydrotreating conditions employed for distillates (naphtha) and at temperatures of 613 – 673 K (Brasil et al., 2014; Fahim et al., 2010).

Due to the fact that the reaction consumes hydrogen, it is usually necessary to recirculate the generated hydrogen to the reactor inlet. In this reactor, the reactions responsible for breaking the hydrocarbons of larger chains begin to occur, producing the methane that will later undergo the reform reaction.

The pretreated charge receives the addition of water vapor at a vapor/carbon ratio equal to 4, and goes to the pre-reform stage. The pre-reforming reactor operates at a lower temperature (783 K) than the reforming reactor (1093 – 1153 K). This setting allows for the conversion of hydrocarbons into methane, the production of hydrogen, and the use of more active catalysts than the ones employed during steam reform (Brasil et al., 2014; Fahim et al., 2010).

Figure 2. Information shared between ASPEN PLUS™ and MATLAB® through Excel.

In the reforming furnace, the reform reaction occurs to a greater extent. The charge is preheated in the convection section and fed to the reformer tubes where the catalyst is located. The reaction between hydrocarbons and water vapor produces hydrogen (H_2) and carbon monoxide (CO). This reaction, which is strongly endothermic, occurs in a pressure range between 2 and 2.5 MPa and at temperatures between 1093 and 1153 K (Brasil et al., 2014; Fahim et al., 2010).

The pretreated reactor, pre-reforming and steam-reforming reactor were simulated in an equilibrium reactor (Gibbs reactor), which minimizes Gibbs' free energy subject to atom balance constraints, therefore yielding the equilibrium composition. This model does not require the reaction stoichiometry as an input. However, it is necessary to provide the temperature and the pressure at which it occurs.

The effluent from the steam-reforming reactor proceeds to the shift reactor. In the first reactor, High Temperature Shift - HTS, the charge is fed to the reactor at a temperature of 613 to 673 K. In the Low Temperature Shift - LTS, the inlet temperature is 483 to 503 K (Brasil et al., 2014; Fahim et al., 2010). In the model, the fixed-conversion reactor was used to represent the shift reactor, establishing that 90% of carbon monoxide (CO) is converted to carbon dioxide (CO_2).

The effluent from the shift reactor then proceeds to the purification stage. The remaining water of the steam reform reactions is removed by condensation. The other contaminants were separated through a PSA column. The adsorption column operates at 293.15 K and 1.49 MPa for 75 seconds of adsorption.

2.2 Mathematical modeling of a single PSA column

The proposed model is a system of partial differential equations (PDE). The numerical technique employed in solving the model was finite differences. From algebraic approximations of the derivatives in a rectangular grid, a linear system is obtained, from which the PDE can be solved.

The mass balance for the one-dimensional flow of a PSA system can be written as presented by Eq. 4. The mass transfer between gas and solid is described by the linear driving-force model (LDF), given by Eq. 5, and the adsorption equilibrium is described by the Langmuir single-site model, as presented by Eq. 6.

The initial values and boundary conditions used in the modeling for the simulation of the hydrogen adsorption column are represented by Eq. 9, 10, 11, and 12. Hydrogen is considered to be inert with respect to the adsorbent bed and, therefore, its composition is given by the total molar fraction constraint in the system, given by Eq. 13.

$$y_i = y_{i_{alim}}, \text{ for } \forall t \text{ and } z = 0 \quad (9)$$

$$y_{H_2} = 1, \text{ for } t = 0 \text{ and } z \neq 0 \quad (10)$$

$$q_i = 0, \text{ for } t = 0 \text{ e } \forall z \quad (11)$$

$$\frac{\partial y_i}{\partial t} = 0, \text{ for } \forall t \text{ and } z = 0 \quad (12)$$

$$y_{H_2} = 1 - y_{CH_4} - y_{CO} - y_{CO_2} \quad (13)$$

2.3 Interface between ASPEN PLUS™ and MATLAB®

A MATLAB® model can be embedded in ASPEN PLUS™ by means of an Excel-VBA interface, as is described in Fig. 2. The software Excel was used so

Table 1. Adsorption column data.

Parameter	Value	Unit
L	4.80	m
T	313.15	K
P	1.49	MPa
CA/Z5A	5/3	Vol/Vol

Table 2. User arrays input for a single bed PSA unit.

Parameter		Character
Int.	Real	
100	75	Number of divisions of column length / absorption time (s)
100	4.8	Time unit number/Column length (m)
	0.4	Active Carbon Porosity
	0.4	Zeolite Z5A Porosity
	1.5	Interstitial velocity (m/s)

as to establish a connection between ASPEN PLUS™ and MATLAB®, allowing data to be both read and written by both applications.

The User 2 block from ASPEN PLUS™ supplies Excel with the inlet stream properties and the real and integer parameters of the model. This block allows the user to execute equations or models that are in Excel worksheets as part of ASPEN™ simulation. There is a ready-to-use Excel template file containing the inlet and outlet stream structure used by ASPEN PLUS™. The Excel file template contains special functions, called Aspenhooks, required to send and receive information to and from ASPEN PLUS™. For more information about these functions refer to **Aspen Plus™ (2000; 2008)**.

The Excel template file organizes this information and sends it to the MATLAB® routine. The interface between Excel and MATLAB® is established through an Excel add-in, the

Spreadsheet Link, which connects it to the MATLAB® workspace, allowing access to the MATLAB® environment from an Excel worksheet through their functions. This information exchange is done through a macro implemented in VBA, responsible for sending it to the user model in MATLAB®.

The MATLAB® routine calculates the properties of the output streams and, if required, additional parameters. This information returns to Excel and the template file sends it back to ASPEN PLUS™.

The Spreadsheet Link add-in includes some functions to the Excel file that allows sending and receiving information to and from the MATLAB® model. More information about these functions can be found in Excel Link available in **Mathworks™ (2017)**.

A user unit built by embedding a MATLAB® user model in ASPEN PLUS™ was used to simulate the single bed adsorption. The simulated adsorption column is composed of two layers of adsorbents: activated carbon (AC) and zeolite 5A (Z5A), as shown in Fig. 3. Langmuir parameters, mass transfer coefficients, and adsorbent bed characteristics were obtained from the work of **Rahimpour et al. (2013)**. The adjusted gas velocity was $u = 1.5 \text{ m/s}$ and the parameters are shown in Table 1.

The connection of the model implemented in MATLAB® to ASPEN PLUS™ was carried out using the ASPEN™ interface, the Excel file template and the spreadsheet link add-in. The parameters of the user block are presented in the Table 2.

Figure 3. Detail of the adsorbent layers on a PSA column. Measurements are given in meters.

Table 3. Composition (mol/mol) of the currents and operational condition of the steam reforming reactors.

Steam	FEED-MIX	REF1	REF-GAS
T (K)	772.95	1123.15	1123.15
P (MPa)	29	27	25
y CH₄ (%)	20.69192	24.95481	5.550656
y C₂H₆ (%)	2.396716	0.000878	0.000296
y C₃H₈ (%)	2.284316	1.7E-07	5.54E-08
y CO (%)	0	0.178176	10.64461
y CO₂ (%)	0	4.304397	5.621848
y H₂ (%)	0.000178	11.41227	49.72233
y H₂O (%)	74.62687	59.14949	28.46025
F (kmol/h)	4.02	4.41589	5.95846

3. RESULTS AND DISCUSSION

After performing the activities previously presented, the results obtained through the simulations were treated and analyzed. The inferences and interpretations that stemmed from the analysis of these data are presented and discussed next.

3.1 Hydrogen Production Unit (HPU) simulation

The unit was simulated assuming a molar flow rate of 1 kmol/h of natural gas, with the following composition: 80% (mol/mol) H₄, 10% C₂H₆ and 10% C₃H₈ with the objective of generating 3.6 kmol/h of purified product, H₂, with 99.8% purity.

Some contaminants that may be present in lesser proportions in the gas were suppressed from the feed. However, a pretreatment reactor, where a hydrodesulfurization (HDS) reaction would occur was included in the simulation because it already begins the hydrocarbon breaks reactions for methane formation, which will subsequently undergo the steam reforming reaction. For the pre-reform reaction, the modular unit representing a Gibbs equilibrium reactor available in ASPEN PLUS™ was used. In this reactor, about 99.99% of all ethane and propane were converted to methane.

After being heated, the output stream from the pre-reforming reactor was sent to the reforming reactor, an ASPEN™ Gibbs reactor, where

methane reacted with water vapor to form hydrogen. In this reactor, 70% of the methane was converted by the reforming reaction to carbon monoxide and hydrogen. The composition of the currents and operational condition of the pre-reform and steam reforming reactors are shown in Table 3.

The output current from the reforming reactor went to the CO conversion stage, to produce more hydrogen through the displacement reaction. This step was performed in two stages, HTS and LTS. For both reactors, the ASPEN conversion reactor was used and the CO conversion was set at 90%. The composition of the currents and operational condition of the conversion reactors are shown in Table 4.

The output current of the converter contained the excess water vapor used in the process and the other contaminants. It was thus necessary for it to undergo purification steps. The water was removed by cooling and condensation. For the removal of the other contaminants, one adsorption column was used. As almost all ethane and propane were converted to methane and the water was removed by cooling, the components in the feed column were: CH₄, CO, CO₂, and H₂. The adsorption time considered was 75s. With this last stage of purification, the hydrogen leaves with a purity of 99.8%. Due to the fact that the ethane, propane, and water components were not incorporated into the adsorption process model (the same amount that enters the column, exits), the composition of

Table 4. The composition (mol/mol) of the currents and operational condition of the conversion reactors.

Steam	FEED-HTS	HTS-OUT	LTS-OUT
T (K)	623.15	623.15	483.15
P (MPa)	24	23	21
y CH₄ (%)	5.550656	5.550656	5.550656
y C₂H₆ (%)	0.000296	0.000296	0.000296
y C₃H₈ (%)	5.54E-08	5.54E-08	5.54E-08
y CO (%)	10.64461	1.064461	0.106446
y CO₂ (%)	5.621848	15.202	16.16002
y H₂ (%)	49.72233	59.30248	60.2605
y H₂O (%)	28.46025	18.88011	17.92209
F (kmol/h)	5.95846	5.95846	5.95846

Table 5. Composition (mol/mol) of the currents and operational condition of the water remover and the PSA unit.

Steam	LTS-OUT	PSA-IN	H2-PURE
T (K)	483.15	293.15	293.15
P (MPa)	21	15	15
y CH₄ (%)	5.550656	6.753829	2.09E-17
y C₂H₆ (%)	0.000296	0.00036	0.000491
y C₃H₈ (%)	5.54E-08	6.74E-08	9.17E-08
y CO (%)	0.106446	0.12952	1.08E-16
y CO₂ (%)	16.16002	19.6629	1.58E-68
y H₂ (%)	60.2605	73.32271	99.8216
y H₂O (%)	17.92209	0.130686	0.177917
F (kmol/h)	5.95846	4.89698	3.59702

the hydrogen in the column output stream was slightly below the values obtained in real processes. The composition of the currents and operational condition of the water removal and the PSA unit are show in Table 5.

By analyzing the results of the simulation of the UGH, one can conclude that it satisfactorily models the process of hydrogen production and purification. In the simulation, it was possible to observe all stages of the process as well as the unit operations involved. It was also possible to evaluate the compositions of the intermediate streams as well as the compositions of the final product, providing a broad view of the evaluated unit.

The conditions used, the results achieved, and

the purity of the final product obtained in the simulation are in agreement with the values reported in the literature (Brasil et al., 2014). The complete process, PDF, of the simulated process is presented Fig. 4.

3.2 Validation of the single bed adsorption column model

To validate the mathematical model of an adsorption column, a scenario corresponding to an experiment by Park et al. (1998) was simulated, and the results obtained were compared to those reported by the authors. The conditions of the simulation were the same reported by Park et al. (1998), and the adjustment of the mathematical model to the experimental data was done by adjusting the interstitial velocity to $u = 0.45$ m/s.

Figure 4. PDF of the Hydrogen Production Unit.

Figure 5. PDF Comparison between experimental rupture curves and numerical simulation.

Figure 5 shows the comparison graphs of the experimental results obtained by [Park et al. \(1998\)](#) and the numerical model response of the adsorption column.

Comparing the graphs, one can see that there is little discrepancy between the experimental results and those obtained from the simulation, which indicates that the model adequately represented the experimental data. Greater deviations were observed for carbon monoxide and methane. In the case of carbon monoxide, the simulation curve was displaced in relation to the experimental data, but the behavior of both is similar. This displacement

between the curves is due to the adjustment of the numerical model through the interstitial velocity, which directly influences the front of the breakthrough curve. Regarding methane, a deviation almost constant with respect to the peaks was observed, however, the behavior of the curves was similar.

3.3 Connection between ASPEN PLUS™ and MATLAB®

The communication between ASPEN PLUS™ and MATLAB® was successful. To verify the efficiency of the connection and to analyze the

Figure 6. Comparison between the results of the MATLAB[®] model and the ASPEN[™] modular unit.

results generated in the simulations, the results obtained from the model developed in ASPEN PLUS[™] were compared to the results of the MATLAB[®] model, as can be seen in Fig. 6.

Figure 6 shows that the connection between the two software applications did not influence the results. It is important to highlight that the adsorption model implemented in MATLAB[®] was validated against the comparison of the simulated data with the experimental data presented by **Park et al. (1998)**. Therefore, the modular unit inserted in ASPEN PLUS[™], faithfully reproduced the results presented previously.

The model developed in MATLAB[®] provides the dynamic behavior of the adsorption process in a column, which presents the compositions of the components as a function of the operating time and the compositions along the length of the column for a given adsorption time. In ASPEN PLUS[™], this model was used to establish a static simulation, which returns the composition at the output of the column. For this, it is necessary to provide certain operating parameters, such as adsorption time, column length, bed porosity, and interstitial velocity. The connection between the ASPEN PLUS[™] process simulator and MATLAB[®] was successfully performed. Thus, the adsorption model developed in MATLAB[®] was implemented in the ASPEN[™] environment, since the simulator

does not have the unitary adsorption operation in its library of modular units.

4. CONCLUSION

From the analysis of the results obtained, one can infer that the main objective of this work was attained. A mathematical model capable of generating data describing the dynamic behavior of an adsorption column for hydrogen purification was built. Despite the simplifying assumptions made in the model, its representation was not impaired, since the data generated in the simulation displayed a good correlation with experimental results reported in the literature.

The connection between ASPEN PLUS[™] and MATLAB[®] via Excel was successful. This connection was able to perform the exchange of data between the two computational programs, sometimes sending data from ASPEN[™] to MATLAB[®], sometimes from MATLAB[®] to ASPEN[™]. The results obtained also reinforce the versatility of EXCEL[™] when using the features of the VBA programming language.

Finally, it is worth emphasizing the importance of performing the simulation of a hydrogen generation plant. This work is a step towards the

development of simulations, aiming at Operator Training (OTS). The results from a simulation, when well adjusted, provide the operator and/or user with a better understanding and follow-up to the process, and the knowledge for working with their variables in a much more conscious way, being able to evaluate their effects and impacts before actual implementation.

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SYMBOLS

$a_{n,i}$	Langmuir parameter for adsorbate i
$b_i, b_{n,i}$	Langmuir parameter for adsorbate i
ε	Adsorption column bed porosity
ρ_p	Specific mass of the adsorbent particle
P	Operating pressure of the system
q_i	Concentration of element i in the solid phase
q_i^*	Concentration of equilibrium of the adsorbate i in the solid phase
q_i^{sat}	Concentration of saturation of adsorbate i in the solid phase
R	Universal ideal gas constant
T	Operating temperature of the system
t	System operating time
u	Interstitial speed
y_i	Molar fraction of element i in the gas phase
z	Axial position on the adsorption column

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