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Utilization of dual - PSA technology for natural gas upgrading and integrated CO_2 capture

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Abstract

In this study, we have considered Pressure Swing Adsorption (PSA) technology natural gas purification and integrated CO_2 capture. We present the utilization of Dual-PSA technology for obtaining pipeline-quality methane and integrated CO_2 capture. In the initial simulation example, a binary stream of 10% CO_2 – 90% CH_4 could be separated into two streams where the CH_4-rich stream has a purity of 98% and the CO_2-rich stream has a purity of 91%. The overall recovery of methane is higher than 99% and unit productivity of the Dual-PSA is 3.3 mol CH_4 /(kg_{ads}.h).

Keywords: natural gas; CO2 capture; kinetic adsorbents; Pressure Swing Adsorption; modeling

1. Introduction

The utilization of fossil fuels is expected to remain important in the production of power and fuels in the next decades [1]. Natural gas is a fossil fuel with an increased demand in the last years and with proved reserves exceeding 187 trillion cubic meters [2]. The natural gas (NG) value chain can be divided in: production (extraction), conditioning (removal of contaminants), transportation (pipeline or liquefied natural gas, LNG) and consumption. Carbon dioxide emissions are associated to this entire value chain [3]. Particularly, when the natural gas is contaminated with significant amounts of CO_2, the overall emissions are quite high. If the CO_2 is separated and re-injected to the NG field, it will boost the production of oil by Enhanced Oil Recovery. Current technology for CO_2 removal from natural gas is amine scrubbing [4]. Other alternative to remove CO_2 is by cryogenic methods [5] and by membranes [6].

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The aim of this work is to design and evaluate an adsorption-based process to make this separation as an alternative technology.

Adsorption processes for CH_4-CO_2 separation are commercially used in the upgrading of biogas [7] to reduce the content of CO_2 to pipeline-quality methane or fuel-grade methane. The content of CO_2 varies from some per cent to almost 50% in landfill gas, while the feed pressure ranges from 2-12 bars. Plants up to 10000 Nm^3/h are available but common flowrates are 100 - 1000 Nm^3/h [7]. The only application of PSA for CO_2 removal from natural gas is carried out by Xebec Inc. and was designed to treat around 6000 Nm^3/h at a feed pressure of 20 bar with a total content of CO_2 of 13%. The unit is actually treating 3500 Nm^3/h with a methane recovery of around 62% with 1.42% of CO_2. The content of methane in the exhaust gas of the PSA is 40% and this stream is used to produce energy [8].

Our aim is to design a PSA unit to remove CO_2 from natural gas streams. However, the target of our design is quite different from existing PSA technology. The feed stream specifications are: 250000 Sm^3/h with an inlet pressure of 70 bar and with a CO_2 content of 10%. The target is to produce pipeline-quality methane ($CO_2 < 2.5$%), but also to produce a high-purity stream to be re-injected to reduce environmental impact of exploration. As can be seen, the flowrate to be treated is two orders of magnitude higher than existing technology and also the feed pressure is more than 3 times higher. Furthermore, there is an imposition of CO_2 purity. All these factors together indicate that the adsorbent material that should perform the separation should be very selective to CO_2.

Moreover, the PSA configuration and regeneration steps comprising the cycle should be specifically designed. The aim of this study is to suggest a PSA design to be employed for natural gas sweetening. For this reason, we have assumed some properties of the adsorbent material and develop a sequential analysis of the PSA variables that should be optimized to obtain two streams with high purity. Finally, the design and results obtained with a dual-PSA configuration are presented and described in detail.

Nomenclature

C concentration: of component i (C_i) and total (C_T)

q_i^* adsorption equilibrium of component i

$K_{LDF,i}$ Linear Driving Force constant for diffusion in the micropores for component i

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2. Simulation of PSA process

The most important requirement to simulate an adsorption process is the adsorption equilibrium isotherms. Since our purpose is to design an adsorption process and evaluate its potential, we have started by selecting some specific adsorption properties. The chosen isotherms of the material at 313 K are shown in Figure 1. They obey the Langmuir isotherm and the parameters are reported in Table 1. We have also assumed that when the binary gas mixture is fed to the PSA unit, the binary adsorption equilibrium can be described by the extension of the Langmuir model for multicomponent mixtures, given by:

$$q_i^* = \frac{q_{m,i}K_{eq,i}P_i}{1+\sum_{i=1}^{N}K_{eq,i}P_i} \tag{1}$$

$$K_{eq,i} = K_{i,\infty}\,exp\left(\frac{-\Delta H_i}{RT}\right) \tag{2}$$

Table 1. Adsorption equilibrium parameters (Langmuir model) and diffusion time constants of methane and carbon dioxide.

	CH_4	CO_2
Maximum loading, $q_{m,i}$ [mol/kg]	3.0	5.0
Infinite adsorption constant, $K_{i,\infty}$ [1/bar]	2×10^{-5}	1×10^{-5}
Heat of adsorption, $(-\Delta H_i)$ [J/mol]	15000	25000
Adsorption time constant, $K_{LDF,i}$ [1/s]	$\sim 10^{-4}$	3×10^{-2}

In Table 1 we are also indicating a time constant for diffusion. We have assumed that the particle is bidisperse and that diffusion can be described by a bi-LDF (linear driving force) model and in this case diffusion is controlled in the micropores of the adsorbent. With these time constants, methane diffusion in the micropores of the adsorbent is extremely slow (kinetic adsorbent). Such effect is observed in some materials like titanosilicates, alumino-phosphates and carbon molecular sieves.
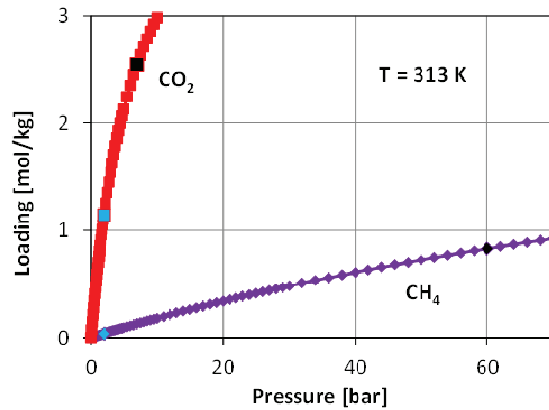

Figure 1. Adsorption isotherms of CO_2 and CH_4 at 313 K used to design a PSA process for separation of the binary mixture 10% CO_2 – 90% CH_4.

The conservation equations of the PSA model were derived using the following assumptions [9]:
a. Ideal gas behaviour inside the column,
b. No mass, heat or velocity variations in the radial direction,
c. Axial dispersed plug flow,
d. External mass and heat transfer resistances expressed with the film model,
e. Bidisperse adsorbent particle: macropore and micropore mass transfer resistances expressed with the Linear Driving Force (LDF) model,
f. No temperature gradients inside each particle,
g. Inside the column, porosity is considered constant.

The mass balance for each component in the gas phase is given by:

$$\varepsilon_c \frac{\partial C_i}{\partial t} = \varepsilon_c \frac{\partial}{\partial z}\left(D_{ax} C_T \frac{\partial y_i}{\partial t}\right) - \frac{\partial\left(u_o C_i\right)}{\partial z} - \left(1 - \varepsilon_c\right)\frac{a' k_{fi}}{Bi_i / 5 + 1}\left(C_i - \overline{c_i}\right)$$ (3)

where C_i is the gas-phase concentration, D_{ax} is the axial dispersion coefficient, u_o is the superficial velocity, ε_c is the column porosity, y_i is the molar fraction, k_{fi} is the film mass transfer resistance, Bi_i is the Biot number and $\overline{c_i}$ is the averaged concentration in the macropores, all valid for component i, while C_T is the total gas concentration and a' is the pellet specific area.

We have assumed a LDF model for the mass transfer rate from the gas phase to the macropores. The mass balances in the particles is given by:

$$\varepsilon_p \frac{\partial \overline{c_i}}{\partial t} + \rho_p \frac{\partial \langle \overline{q_i} \rangle}{\partial t} = \varepsilon_p \frac{15 D_{p,i}}{R_p^{\,2}} \frac{1}{1 + 5 / Bi_i}\left(C_i - \overline{c_i}\right)$$ (4)

where $D_{p,i}$ is the pore diffusivity, R_p is the extrudate radius, ρ_p is the particle density, ε_p is the particle porosity and $\langle q_i \rangle$ is the extrudate averaged adsorbed phase concentration.

The LDF equation for the crystals averaged over the entire extrudates is expressed by:

$$\frac{\partial \langle \overline{q_i} \rangle}{\partial t} = K_{LDF,i}\left(q_i - \langle \overline{q_i} \rangle\right)$$ (5)

where q_i is the adsorbed phase concentration in the equilibrium state.

In the momentum balance we have considered that the pressure drop and velocity change are related through the Ergun equation, defined by:

$$-\frac{\partial P}{\partial z} = \frac{150\,\mu\left(1 - \varepsilon\right)^2}{\varepsilon_c^{\,3} d_p^{\,2}} u_0 + \frac{1.75\left(1 - \varepsilon\right)\rho_g}{\varepsilon_c^{\,3} d_p}\left|u_0\right|u_0$$ (5)

where P is the total gas pressure, μ is the gas viscosity, ρ_g is the gas density and d_p is the particle diameter.

The energy balance in the gas phase is:

$$\frac{\partial}{\partial z}\left(\lambda \frac{\partial T_g}{\partial z}\right) - u_0 C_{g,T} C_p \frac{\partial T_g}{\partial z} + \varepsilon_c R_g T_g \frac{\partial C_{g,T}}{\partial t} -$$

$$- \left(1 - \varepsilon_c\right)a_p h_f\left(T_g - T_p\right) - \frac{4 h_w}{d_{wi}}\left(T_g - T_w\right) - \varepsilon_c C_{g,T} C_v \frac{\partial T_g}{\partial t} = 0$$ (6)

with T_g, T_p and T_w respectively as the gas, particle and wall temperatures; C_v and C_p as the gas molar specific heat at constant volume and pressure respectively; R_g as the ideal gas constant; d_{wi} as the wall internal diameter, λ as the heat axial dispersion coefficient. The film heat transfer coefficient between the

gas phase and the particle is represented by h_f, while the film heat transfer coefficient between the gas phase and the wall is represented by h_w.

As it is assumed that there are no temperature gradients inside a particle, the solid phase energy balance for the column is given by:

$$\left(1-\varepsilon_c\right)\left[\rho_p \sum_{i=1}^{n} \overline{q_i}\, C_{v,ads,i} + \rho_p\, \hat{C}_{p,s}\right]\frac{\partial T_p}{\partial t} =$$

$$= \left(1-\varepsilon_c\right)\varepsilon_p\, R_g\, T_p\, \frac{\partial C_T}{\partial t} + \left(1-\varepsilon_c\right)\rho_p \sum_{i=1}^{n}\left(-\Delta H\right)_i \frac{\partial \overline{q_i}}{\partial t} + \left(1-\varepsilon_c\right)a_p\, h_f\left(T_g - T_p\right)$$

(7)

where ρ_b is the bulk density of the bed, $\hat{C}_{p,s}$ is the solid specific heat per unit sorbent mass and $\left(-\Delta H\right)_i$ is the isosteric heat of adsorption of component i.

Finally, for the energy balance of the column wall energy exchange with the gas phase inside the column and with the external environment is considered:

$$\rho_w\, \hat{C}_{p,w}\, \frac{\partial T_w}{\partial t} = \alpha_w\, h_w\left(T_g - T_w\right) - \alpha_{w\ell}\, U\left(T_w - T_\infty\right)$$

(8)

where T_∞ is the external temperature, ρw is the wall density, $\hat{C}_{p,w}$ is the wall specific heat per mass unit, U is the overall heat transfer coefficient and αw and $\alpha_{w\ell}$ are defined by:

$$\alpha_w = \frac{d_{wi}}{e\left(d_{wi} + e\right)} \quad ; \quad \alpha_{w\ell} = \frac{1}{\left(d_{wi} + e\right)\ln\left(\dfrac{d_{wi} + e}{d_{wi}}\right)}$$

(9)

where e is the wall thickness.

The main difference between this model and previous ones is related to the relationship used to describe gas behavior. In this case, the pressure of the system is quite high and ideal gas law cannot be employed. We have employed the Benedict-Webb-Rubin equation together with the mixing rules used to describe multicomponent gas behavior [10]. The BWR is given by:

$$P = R_g T C_T + \left(B_o R_g T - A_o - \frac{C_o}{T^2}\right)C_T^2 + \left(b R_g T - \alpha\right)C_T^3 + a\alpha C_T^6\left(1 + \gamma C_T^2\right)e^{-\gamma C_T^2}$$

(10)

The values of purity and recovery of methane and carbon dioxide are function of time and the values reported in this work were obtained only after achieving the cyclic steady state (CSS) of the process.

The mathematical model of the PSA unit was solved in gPROMS (PSE Enterprise, UK). The discretization method for the spatial domain was orthogonal collocation with finite elements method (OCFEM) with 70 intervals.

3. PSA cycle considerations

Adsorption processes operating in unsteady state and multiple column arrays are used to separate a continuous feed stream: while one column is processing feed, the others are being regenerated.

In this process, the feed pressure is 70 bars and the partial pressure of CO_2 is 7 bars. In order to desorb some CO_2 at high purity, the blowdown pressure should be smaller than 7 bar which means that P_{high}/P_{low} > 10. Since the methane molar fraction is 0.9 and we are interested in obtaining high purity CO_2, a very large portion of methane should be evacuated from the column before the blowdown step.

An estimative of the product composition that can be obtained in the blowdown step can be obtained by analyzing the number of moles existing inside the column at the end of the feed step. We have started assuming that the column will be at a constant temperature and that the desorption pressure is 2 bar. Furthermore, we have assumed that 86% of the overall CO_2 bed capacity of the adsorbent is used while the rest of the capacity is not employed due to mass transfer resistance [13]. The amount of CO_2 and CH_4 adsorbed (calculated with eq. 1) and in the gas phase inside the column are shown in Figure 2a. Since the amount of methane adsorbed in the feed step is considerable, it will result in an increased recycle of methane and also will increase the thermal variations in the column due to gas adsorption and desorption.
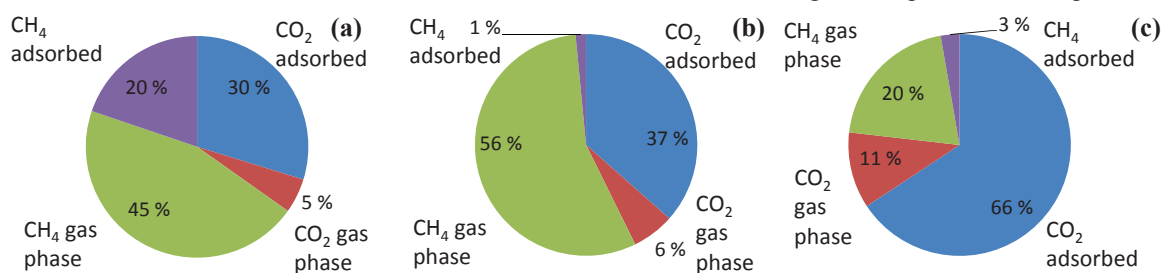

Figure 2. Percentage of CO_2 and CH_4 in the column: (a) after column finished feed step using adsorption equilibrium isotherms; (b) after column finished feed step with a kinetic adsorbent; (c) after the third pressurization step before blowdown.

In order to reduce the methane content in the column prior to the blowdown step, we have specified a "kinetic" adsorbent where actually the methane molecules have a very small diffusion coefficient and thus take a very long period of time to reach equilibrium (and we can assume that almost no methane is adsorbed per cycle). The amount of CO_2 and CH_4 assuming that the adsorbent kinetically limits methane adsorption is shown in Figure 2b. However, even considering that the adsorbent is very selective to CO_2; more than 50% of the total moles inside the column correspond to methane in the gas phase. For this reason, if we are interested in recovering CO_2 at high purity, a large portion of this methane should be removed from the column before the blowdown step.

Assuming that the cycle has three equalization steps that proceed to nearly completion and that no methane is adsorbed or desorbed and that very little CO_2 is moved between columns, then the final pressure of the column before blowdown will be close to 19 bars. The total number of moles before the blowdown step considering that the cycle has three pressure equalizations is shown in Figure 2c. Since the pressure before the blowdown is 19 bars, the content of methane in the product is still high (20%). However, this analysis was a very initial approach and such high CO_2 purity cannot be achieved due to thermal effects, unfavorable and incomplete desorption of CO_2, mass transfer resistances, etc.

The introduction of a rinse step with CO_2 can also be an alternative. However, if the rinse step is placed before the blowdown step, the column pressure is 19 bars. At this pressure the CO_2 will also be

adsorbed (releasing more heat and significantly reducing the productivity of the unit). In order to use a rinse step, a different cycle should be designed, possibly by the introduction of a "provide purge" step.

The next step in the design is to evaluate the thermal effects due to adsorption. Since we are considering the utilization of a kinetic adsorbent, the heat of adsorption released due to methane adsorption can be neglected. On the other side, since the application we are studying involve a large amount of gas, then the column will operate very close to adiabatic regime. We have performed the simulation of a breakthrough curve assuming that the column is nearly adiabatic (U = 2 W/(m^2.K)) and for comparison purposes, one simulation assuming that the column is isothermal. The parameters used for the simulation are listed in Table 2.

Table 2. Column and adsorbent properties used in the simulation of PSA process for CH_4-CO_2 separation

Adsorbent density [kg/m^3]	1060
Column porosity	0.38
Rectifying PSA and breakthrough column simulation	
Column length [m]	6.0
Column radius [m]	1.94
Feed pressure [bar]	70
Feed temperature [K]	313
Feed time [s]	300
Blowdown time [s]	150
Purge time [s]	100
Equalization steps [s]	60
Stripping PSA	
Column length [m]	6.0
Column radius [m]	2.50
Feed pressure [bar]	5
Feed temperature [K]	313
Feed time [s]	250
Blowdown time [s]	150
Rinse time [s]	50
Equalization step [s]	40

The molar flowrates of CH_4 and CO_2 obtained in the simulated breakthrough curves are shown in Figure 3. In the case of the isothermal breakthrough curve, the CO_2 breakthrough is quite steep (due to low mass transfer resistance and favourable isotherm). However, when the column is nearly adiabatic, the breakthrough time is significantly reduced by a temperature increase of over 40 K due to CO_2 adsorption.

4. Dual Pressure Swing Adsorption

One possible alternative to obtain methane and carbon dioxide with high purity is to use the Dual Pressure Swing Adsorption concept earlier proposed for binary gas mixture separation [11]. A similar dual PSA concept is employed commercially to produce high purity nitrogen [12]. The commercial

process use two PSA columns in series to improve the purity of N_2 while the unit proposed in this work aim to purify the two gaseous streams.
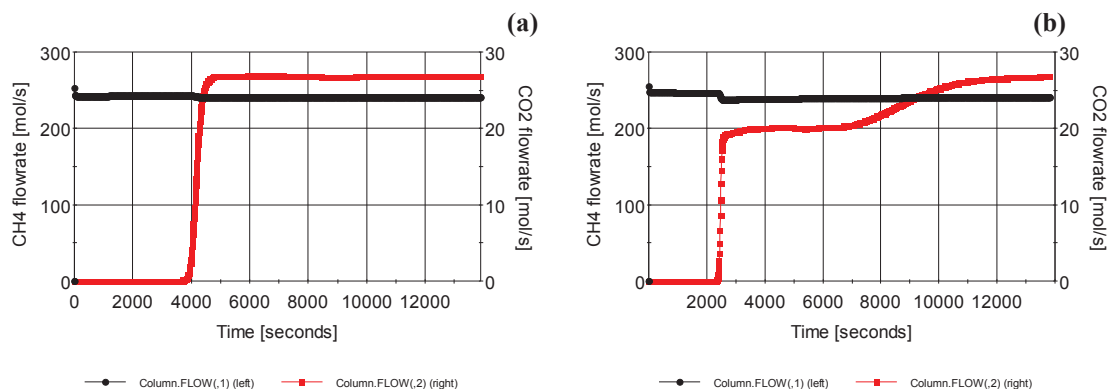

Figure 3. Simulated breakthrough curve of 10% CO_2 – 90% CH_4 stream at 70 bar and 313 K; (a) exit molar flowrate with isothermal operation; (b, c) exit molar flowrate and column temperature in different positions with nearly adiabatic regime. Simulation parameters are listed in Table 5.

The design of the Dual-PSA resembles distillation: one rectifying section for purification of the light component and one stripping section for the purification of the most adsorbed gas. For this mixture using the pre-defined operating conditions, the Dual-PSA scheme can be observed in Figure 4.The benefits of the Dual PSA concept are related to the utilization of simple cycles with targeted steps: light recycle in the rectifying section and heavy recycle in the stripping section. The materials employed in the rectifying and stripping sections can also be different.
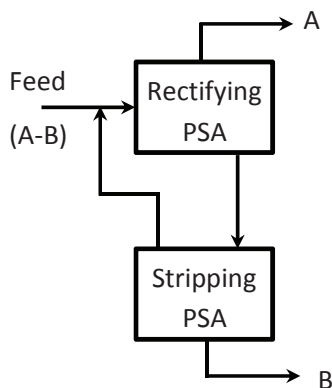

Figure 4. Dual PSA scheme used to obtain purified streams of methane and carbon dioxide.

4.1. Dual PSA concept: Rectifying section

The purpose of the rectifying section is to purify the less adsorbed component. The largest complication of the cycle used in this section is associated to the very high pressure of the feed gas. A cycle with three pressure equalizations is used to reduce the content of CH_4 that will be transferred to the

stripping section. The scheme of the cycle is presented in Figure 5. A cycle with 10-steps (including 3-equalization steps) can be accommodated in a multi-column scheme with more than 6 columns

Note that in this cycle, half of the steps recycle the light product in order to displace CO_2 from the top of the column (and simultaneously to increase the methane recovery in the case of the pressure equalizations). However, the large amount of light product in the column has a negative effect on the CO_2 purity.
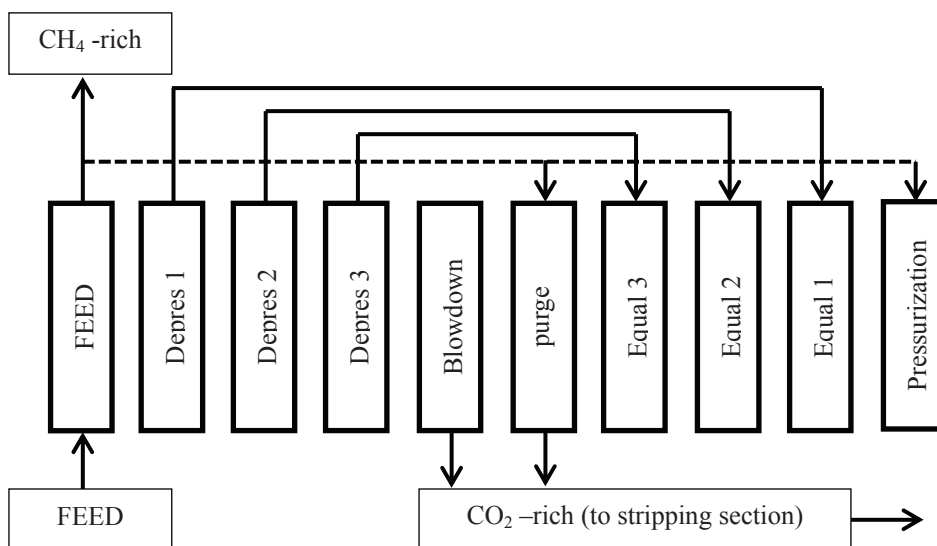
Figure 5. Scheme of the cycle steps employed in the rectifying section of the PSA unit.

Results of a simulation of the rectifying PSA unit are shown in Figure 6 (pressure and exiting molar flowrate in a cycle in cyclic steady state). The parameters used in the simulation are listed in Table 2. The methane purity obtained is 98% with a total CH_4 recovery of 85%. The contamination of 2% of CO_2 is mainly due to some CO_2 recycled in the pressure equalization steps (particularly in the third one). Due to the size of the columns, the pressure equalization steps were allowed to last 60 seconds which also promotes some CO_2 desorption. The CO_2 purity of the streams taken from the blowdown and purge steps is 39%. The difference between 39% and the maximum purity shown in Figure 2c is that the adsorbent is not completely regenerated (some CO_2 is not desorbed) and also by the thermal and mass transfer effects that limit the amount of adsorbed CO_2 when compared to the ideal case. In fact, the effective utilization of the adsorbent is less than 40% of its total capacity. In cyclic steady state (after cycle 50), the temperature variation within a cycle is around 30 K. The stream with 39% purity obtained in the blowdown and purge steps has to undergo further purification. This is done in the stripping section described in the next section.

4.2. Dual PSA concept: Stripping section

The objective of the stripping section in the dual PSA is to further purify the CO_2-rich stream to comply specifications. Furthermore, a CH_4-containing stream can be recycled to the rectifying section. Alternatively, to save some compression energy, the recycled stream can be used as fuel locally to produce electricity or steam. The main feature of the stripping section is the lack of a purge step with light

compound. In a similar manner to distillation, the stripping section should be rich in the heavy component and thus, only a heavy recycle is considered. The step where the heavy component is recycled is termed rinse. The steps used in the stripping section are (see Figure 7):

- co-current pressurization and feed with the stream taken from rectifying section,
- depressurization,
- rinse with purified CO_2 stream,
- blowdown step (where purified CO_2 is obtained)
- pressure equalization.

The cycle described in Figure 7 can be fitted in a three column system. Note that also to increase the content of heavy component in the column, the pressurization is also carried out with feed step. The parameters used for the simulation are reported in Table 2.
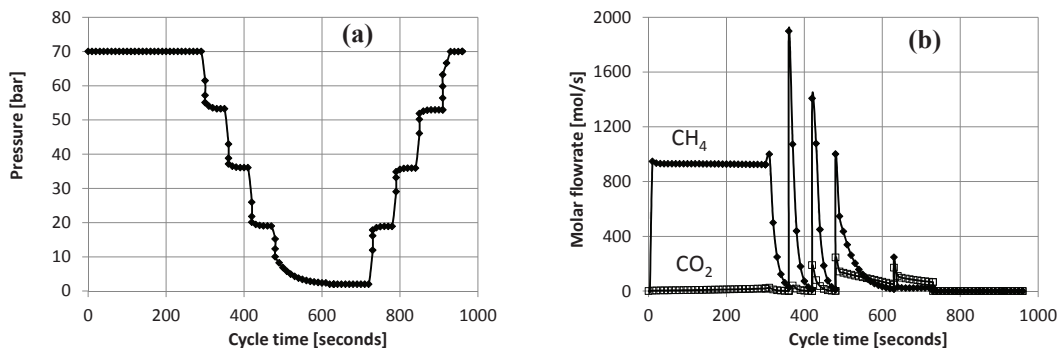

Figure 6. Pressure (a) and CH_4 and CO_2 molar flowrate exiting the column (b) during one cycle under steady state in the rectifying section of the dual PSA unit.

Results of a simulation of the stripping section are shown in Figure 8. Note that the feed pressure used in the stripping cycle is 5 bar. The pressure was increased to increase the CO_2 adsorption capacity in this column. Also note that the column diameter is larger than in the rectifying section, even when there is much less gas to treat. The main reason for this is the lower total pressure of the gas which results in higher gas velocity if the same diameter is kept. Due to the lack of a purge step with the light gas, there is some CO_2 leaving the column in the whole duration of the feed step. Some CO_2 is also recycled in the pressure equalization step and in the rinse step (no major CO_2 breakthrough during this step). In this initial simulation, the CO_2-rich stream has a purity of 91%. Also, 94% of the CH_4 entering the cycle is recovered as top product and recycled to the rectifying section.
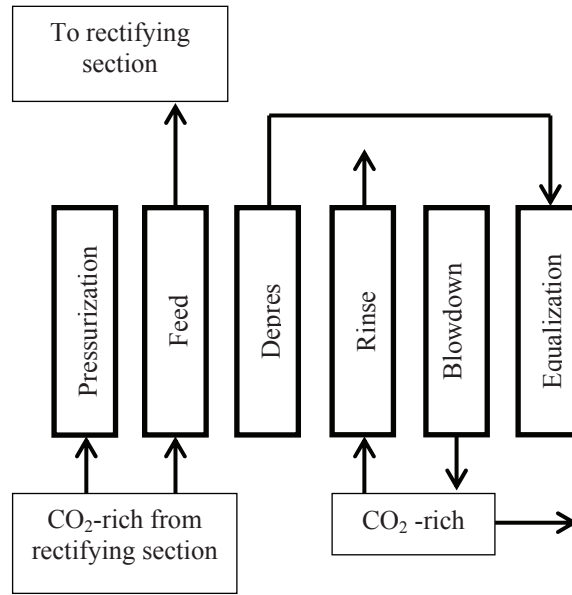
Figure 7. Scheme of the cycle steps employed in the stripping section of the PSA unit.

Integrating the rectifying and stripping sections, the mass balance of the dual PSA unit (in cyclic steady state) is shown in Figure 9. The overall purity of methane is 98% satisfying the pipeline quality specification. Furthermore, the purity of the CO_2-rich stream is 91% satisfies the project requirements. The overall recovery of methane is higher than 99% and the recovery of CO_2 is 82%. The overall unit productivity is 3.3 mol CH_4/ (kg_{ads}.h). The productivity is nearly 50% lower than the one obtained in PSA units for biogas upgrading and the reason is the existence of a stripping section for CO_2 purification, which is actually not done in commercial units. The productivity of the stripping section is comparable to the state-of-the-art units for biogas upgrading [7], but working on much higher flowrates. Due to lack of literature regarding high-pressure PSA units for CH_4-CO_2 it is not possible to compare unit productivity of the rectifying section. Furthermore, we believe it is possible to improve the results of this cycle (either in CO_2 purity and in unit productivity). The energetic consumption of this unit is used entirely in the stripping section: recompression from 2 to 5 bars, blowdown at 0.5 bar and recompression of the top-product stream to 70 bar for recycle.
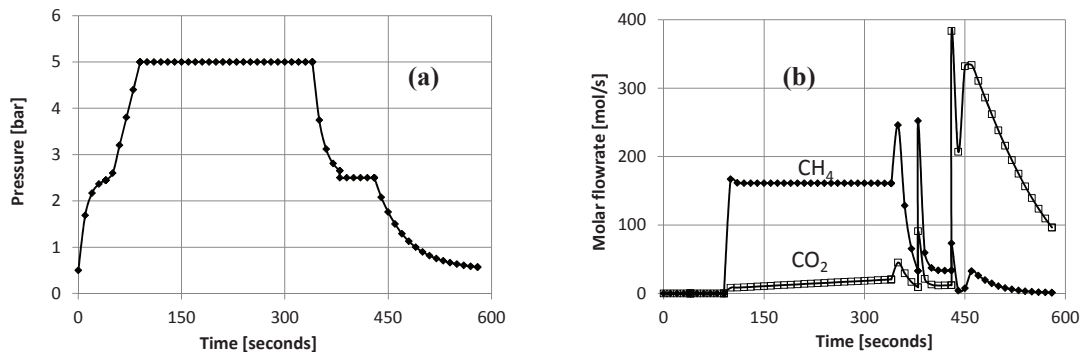

Figure 8. Pressure (a) and CH_4 and CO_2 molar flowrate exiting the column (b) during one cycle under steady state in the stripping section of the dual PSA unit.

The dual PSA concept has proved to be efficient in separating and purifying CH_4 and CO_2 for natural gas applications. The dual PSA concept can be used provided that a selective material can be employed. The adsorbent has to be selective to CO_2 and furthermore, CH_4 has to be kinetically excluded from the micropores.
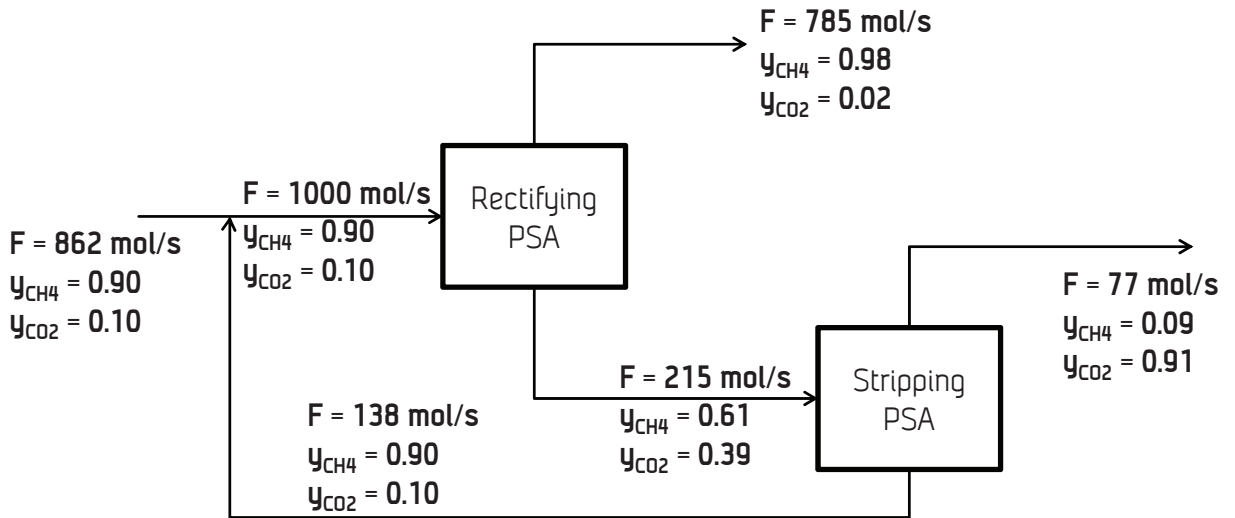

Figure 9. Mass balance in cyclic steady state of the dual PSA unit for CH_4-CO_2 separation.

5. Conclusions

In this work we have demonstrated the feasibility to use the dual-PSA concept for separation and purification of CO_2 from natural gas streams. The unit is composed of a rectifying section operated at 70 bars and having three pressure equalizations. Methane with 98% purity is obtained from the rectifying section. The gas exiting the blowdown and purge steps is treated in a stripping section which is another PSA unit with a completely different cycle where CO_2 is recycled in a rinse step. Carbon dioxide with a purity of 99.1% is obtained and the top product is recycled as feed gas to the rectifying section. The overall recovery of methane is higher than 99%. The overall dual PSA unit productivity is 3.3 mol CH_4/ (kg_{ads}.h). Requirements for the adsorbent material to be employed in this unit is: selective to CO_2 and kinetic control of CH_4 adsorption by micropore size.

6. Acknowledgements

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