

Article

Comparative Evaluation of PSA, PVSA, and Twin PSA Processes for Biogas Upgrading: The Purity, Recovery, and Energy Consumption Dilemma

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Abstract: The current challenges of commercial cyclic adsorption processes for biogas upgrading are associated with either high energy consumption or low recovery. To address these challenges, this work evaluates the performance of a range of configurations for biogas separations, including pressure swing adsorption (PSA), pressure vacuum swing adsorption (PVSA), and twin double-bed PSA, by dynamic modelling. Moreover, a sensitivity analysis was performed to explore the effect of various operating conditions, including adsorption time, purge-to-feed ratio, biogas feed temperature, and vacuum level, on recovery and energy consumption. It was found that the required energy for a twin double-bed PSA to produce biomethane with 87% purity is 903 kJ/kg CH₄ with 90% recovery, compared to 961 kJ/kg CH₄ and 76% recovery for a PVSA process. With respect to minimum purity requirements, increasing product purity from 95.35 to 99.96% resulted in a 32% increase in energy demand and a 23% drop in recovery, illustrating the degree of loss in process efficiency and the costly trade-off to produce ultra-high-purity biomethane. It was concluded that in processes with moderate vacuum requirements (>0.5 bar), a PVSA should be utilised when a high purity biomethane product is desirable. On the other hand, to minimise CH₄ loss and enhance recovery, a twin double-bed PSA should be employed.

Keywords: carbon capture; biogas upgrading; pressure vacuum swing adsorption; PSA; PVSA; biomethane; polymeric sorbent



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1. Introduction

Biogas production from anaerobic digestion has sparked increasing interest over the past decade. Biogas production within the European Union was reported at a total of 16,838 ktoe in 2018, an 87% increase from 2000 [1]. Additionally, there has been a continuous increase in biogas-to-biomethane plants. In 2020, a survey by the Natural & bio Gas Vehicle Association (NGVA Europe) showed that 17% of gas used across 12 European countries consisted of biomethane [2]. Irrespective of this growth, in 2018, EurObserv'ER reported a total of 570 biomethane production plants within the EU, a value overshadowed by the 16,500 biogas plants due to the preferred use of biogas for electricity generation [1]. The traditional usage of biogas was mainly combustion in combined heat and power (CHP) units. However, biogas upgrading to biomethane is the current state-of-the-art application of biogas that leverages environmental benefits by reducing greenhouse gas (GHG) emissions as well as economic benefits due to the higher exploitation of energy contained in biomass [3,4].

The small number of biomethane production plants in comparison to biogas plants can be rationalised via an analysis of the current state-of-the-art technologies in biogas upgrading. Liquid amine scrubbing has been proven to provide the highest CH₄ purity of >99%, with the highest CH₄ recovery compared to other technologies [5–7]. Although amine scrubbing offers a promising performance, its required regeneration energy demand is high,

which has limited its further deployment [8,9]. Pressure vacuum swing adsorption (PVSA) is an alternative process that has been widely used for biogas upgrading and benefits from lowered energy penalties. However, conventional PVSA systems are associated with high CH_4 losses in the tail gas, reaching values of 11–14% [10], leading to low CH_4 recovery [10,11]. CH_4 recovery can be potentially enhanced, but at the cost of excess energy penalties, which in turn hinders their widespread roll-out for biomethane production.

Advancing the existing adsorbents and process configurations has been recognised as a potential solution to improve the performance of conventional PVSA systems. One strategy to advance state-of-the-art adsorbents is to develop materials with a lower required regeneration energy demand while retaining adequate working capacity and kinetics. Grande et al. [12] evaluated the performance of CMS-3K as a kinetic-based adsorbent versus zeolite 13X as an equilibrium-based adsorbent for biogas upgrading to produce ultra-pure biomethane (>98%) in a PVSA process, and under vacuum conditions of 0.1 bar. The required energy consumption for zeolite 13x and CMS-3K was 0.41 and 0.27 kW/mol CH_4 at low CH_4 recoveries of 60.7 and 79.7%, respectively. Wu et al. [13] developed a novel metal organic framework adsorbent (MOF 508b) and compared its performance with conventional carbon molecular sieve (CMS 3K) and zeolite 13X in a double-bed PVSA process. They found that to produce biomethane with a CH_4 purity of 98% and a recovery of 85%, the energy demand increases in the order of MOF 508b < CMS 3K < zeolite 13X with 0.185, 0.214, and 0.422 kWh/Nm³ CH_4 . The double-bed PVSA process is associated with high energy demand due to the vacuum requirement in both PVSA processes. Abd et al. [14] developed a novel plate PSA unit using a layered bed of UiO-66 and biomass-based adsorbent for biomethane production from a biogas mixture of 30:70 vol% CO_2 : CH_4 and could produce bio- CH_4 at 99.99% purity and 70% recovery at 2 bar adsorption pressure and atmospheric temperature. Rzepka et al. [15] developed nanosize zeolite $-\text{[Na}_{10}\text{K}_2]\text{-A}$ pellets, tested their performance in a single-bed VSA unit, and confirmed their superior performance by upgrading the biogas to biomethane with a purity > 99%.

There is a growing interest in utilising sustainable materials for biogas upgrading that take advantage of being low cost while reducing carbon emissions and addressing the circular economy context. Iens et al. [16] Developed an activated carbon adsorbent from pine sawdust that could produce biomethane with 95% purity and 60% recovery. Petracchini et al. [17] tested the performance of natural zeolites for biogas upgrading and produced biomethane with purity > 98% and recovery > 95%, while CO_2 purity in tail gas was <0.1%. They tested the performance of these sustainable adsorbents in an experimental rig for 3 months with no major reduction in their performance.

Sonneleitner et al. [18] assessed the performance of two commercial materials, Zeolite 13X and Lewatit®1065, for biogas upgrading using temperature swing adsorption (TSA) process and reported regeneration energy demands of 0.7–1.1 and 0.3–0.5 kWh/m³ for Lewatit®1065 and Zeolite, 13X respectively. The TSA processes suffer from low productivity and mechanical stability due to working at high temperatures for regeneration [18,19].

Golmakani et al. [20] utilised a combination of advanced adsorbent and a twin PSA configuration to address the low recovery and high energy consumption of biogas upgrading units. They used porous polymeric beads (PPB) adsorbents, which can be regenerated without a vacuum requirement, and used a twin double-bed PSA configuration to enhance the recovery while keeping the energy demand low. Their proposed process produced CH_4 with 90% recovery at an energy demand of 903 kJ/kg CH_4 . Although the twin double-bed PSA process enabled the enhancement of CH_4 recovery, the process required a compressor in the second unit to pressurise the feed, which caused an increase in energy demand, indicating a competition between recovery and energy consumption in such processes. Biomethane purity is another parameter that competes with recovery and energy consumption. Moreover, the minimum purity requirement varies in every country, from 85% for the Netherlands and France to 97% for countries like Sweden, Spain, and the UK [7], which accordingly needs to be met when designing the upgrading processes. Furthermore, operating conditions, including adsorption time, purge to feed ratio (PG/F), biogas feed

temperature, and vacuum level, are other parameters that can significantly affect purity, recovery, and energy consumption in each configuration. Therefore, to achieve an optimum biogas upgrading process with maximised biomethane purity and recovery and minimised energy consumption, firstly a comprehensive study of the most current state-of-the-art adsorption technologies for biogas upgrading was conducted, and it was concluded that PSA, PVSA, and twin PSA are more common due to low cost and performance. Then a systematic comparison of PSA, PVSA, and twin PSA over a wide range of operational conditions was studied. Accordingly, in this work, an analysis of the adsorbent's performance in PSA, PVSA, and twin double-bed PSA processes at identical inlet conditions (feed flowrate, composition, and temperature) was carried out to evaluate each process configuration's suitability for addressing the low recovery and high energy consumption of current adsorption technology for biogas upgrading. Consequently, the effect of operating conditions on the recovery, purity, and energy consumption of PPB adsorbent in pressure swing adsorption processes was studied. This included categorising the performance with respect to key process parameters, including adsorption time, PG/F, biogas feed temperature, and desorption vacuum level. Finally, the current framework of this paper aims to address the lack of a certain guideline for determining the minimum purity of biomethane to prevent extra energy penalties due to a trade-off between recovery and energy consumption with a minimum product purity requirement.

2. Materials and Methods

2.1. Process Description

The schematic of the explored PSA, PVSA, and twin double-bed PSA processes is provided in Figure 1. The bed diameter is 3 cm with 42.9 g of adsorbent. The PVSA process utilises vacuum conditions for the end-of-cycle bed regeneration step and is at the forefront of biogas applications [21–24]. During the vacuum desorption step in the PVSA process, the desorbed gas is treated as an off-gas due to the low CO₂ purity balanced by methane [10]. By implementing a twin double-bed PSA process (Figure 1b), further treatment of this off-gas is possible by enhancing overall CH₄ recovery and producing a CO₂ stream with storage requirement. The twin double-bed PSA's desorption step introduces a counter-current flow of a purge gas (PG) stream redirected from the PSA product stream. The PG and blow-down (BD) streams are both introduced to the second two-bed PSA, which concentrates the CO₂ desorbed into a high purity stream while recovering further CH₄ into the biomethane product stream.

Each cycle of a PVSA process contains eight steps (Figure 2a). Step 1 comprises feed introduction for CO₂ adsorption (AD) accompanied by a side-stream withdrawal from the product to purge Bed 2, designated as providing purge (PPG). This is followed by step 2, where pressure of both beds is equalised by opening the overhead interbed valve. Step 3 consists of the blowdown (BD) of Bed 1 and the repressurisation (RP) of Bed 2. BD ensures that Bed 1 is free of all adsorbates, while RP prepares Bed 2 to undergo the AD and PPG steps. In Step 4, Bed 1 is regenerated further by reducing the bed pressure below atmospheric levels with a vacuum pump, while Bed 2 is still undergoing the RP step. Steps 5–8 repeat steps 1–4 but in the alternate bed, with Bed 1 undergoing desorption and Bed 2 undergoing adsorption. Vacuum desorption is necessary for the PVSA process in preparing the bed for adsorption in the subsequent cycle and is facilitated by the BD and PG steps in desorbing all adsorbates. The twin double-bed PSA eliminates this step for both beds via an extended BD time interval of 20 s. Therefore, as illustrated in Figure 2b, one cycle contains only six steps. The feed portion used as purge or desorbent is then recovered in the second PSA unit, downstream of the first unit.

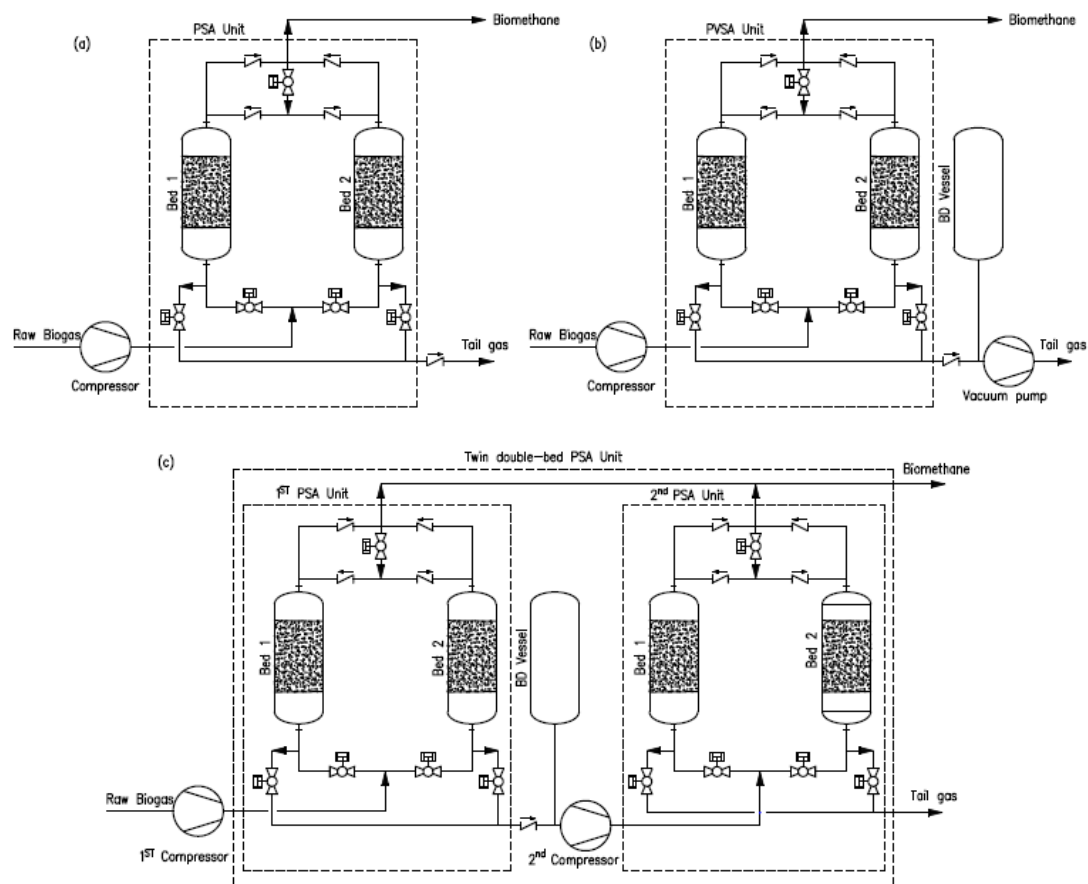


Figure 1. Schematic view of three cyclic adsorption processes for biogas upgrading: (a) PSA process, (b) PVSA process, (c) twin double-bed PSA process.

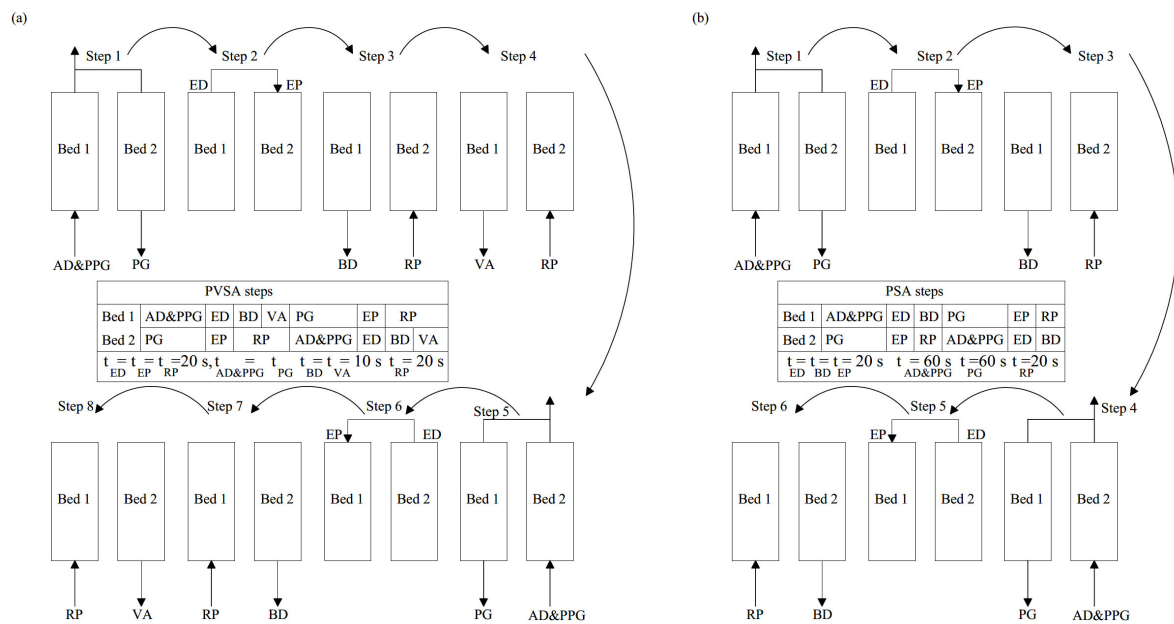


Figure 2. Breakdown of the steps in each cycle for 2 cyclic adsorption process: (a) PVSA process, (b) twin double-bed PSA process. AD: adsorption, PPG: providing purge, ED: equalisation depressurisation, BD: blow down, PG: purge, EP: equalisation pressurisation, RP: repressurisation, PSA: pressure swing adsorption, PVSA: pressure vacuum swing adsorption.

2.2. Mathematical Model

PSA, PVSA, and twin double-bed PSA processes described in the previous section were simulated via the process simulation software Aspen Adsorption[®], with the default Implicit Euler integrator used. The design of the processes followed the process flow diagram illustrated in Figure 1. The cyclic nature of the adsorption process was modelled and included all the cycle steps outlined in Figure 2. The numerical equations used to predict process performance are outlined in Table 1. The Partial Differential Equations (PDEs) applied to the adsorbent bed were discretized into 40 nodes via the Aspen Adsorption[®] Upwind Differencing Scheme 1 (UDS1). The software's Implicit Euler integrator was used to solve the resulting ordinary differential equations (ODEs). The convergence criteria were the product mole fractions and the tolerance set at below 10^{-6} for two consecutive cycles.

Table 1. Partial differential equations (PDEs) of the cyclic adsorption processes.

Description	Formulation
Gas phase mass balance for component i	$\frac{\partial}{\partial z} \left(\varepsilon D_z c_{gT} \frac{\partial y_i}{\partial z} \right) - \frac{\partial}{\partial z} \left(u_s c_{g,i} \right) - \varepsilon \frac{\partial c_{g,i}}{\partial t} - (1 - \varepsilon) \rho_p \frac{\partial q_i}{\partial t} = 0 \quad (1)$
Linear driving force (LDF) model for mass transfer	$\frac{\partial q_i}{\partial t} = MTC_i (q_i^* - q_i) \quad (2)$
The heat balance formula for bed wall	$\rho_w C_{pw} A_w \frac{\partial T_w}{\partial t} = 2\pi r_{bi} h_w (T_g - T_w) - 2\pi r_{bo} h_o (T_w - T_{atm}) \quad (3)$
The heat balance formula for solid adsorbent	$(1 - \varepsilon) \left[\rho_p \sum_{i=1}^n q_i C_{v,ads,i} + \rho_p C_{ps} \right] \frac{\partial T_p}{\partial t} = \rho_b \sum_{i=1}^n (-\Delta H_{ads})_i \frac{\partial q_i}{\partial t} \quad (4)$
The heat balance formula for gas phase	$\frac{\partial}{\partial z} \left(\lambda \frac{\partial T_g}{\partial z} \right) - c_{gT} C_p \frac{\partial (u_s T_g)}{\partial z} - \varepsilon C_v T_g \frac{\partial c_{gT}}{\partial t} - (1 - \varepsilon) a_p h_f (T_g - T_p) - \frac{4h_w}{2r_{bi}} (T_g - T_w) - \varepsilon c_{gT} C_v \frac{\partial T_g}{\partial t} = 0 \quad (5)$
Formula for heat transfer from gas phase to bed [25–28]	$Nu_w = \frac{h_w 2r_{bi}}{k_g} = 12.5 + 0.048 Re \quad (6)$
Formula for effective axial heat dispersion coefficient [29,30]	$\frac{\lambda}{k_g} = 7 + 0.5 Re Pr \quad (7)$
Formula for heat transfer from bed wall to atmosphere [31,32]	$Nu_o = \frac{h_o 2r_{bo}}{k_a} = 0.1 Ra^{1/3} \quad (8)$
Formula for heat transfer from solid adsorbent to gas phase [29,33]	$Nu_f = \frac{h_f d_p}{k_g} = 2 + 1.1 Pr^{1/3} Re^{0.6} \quad (9)$
Pressure drop calculation by Ergun equation [34,35]	$-\frac{\partial P}{\partial z} = \frac{150 \mu (1 - \varepsilon)^2}{d_p^2 \varepsilon^3} u_s + \frac{1.75 (1 - \varepsilon) \rho}{d_p \varepsilon^3} u_s u_s \quad (10)$

Initial conditions ($t = 0$) are: $y_{CO_2} = 0$; $c_{g,CO_2} = 0$; $q_{CO_2} = 0$; $y_{CH_4} = 1$; $T_g = T_p = T_w = T_{inlet}$.

The following assumptions were made: the ideal gas law applies to CO_2 and CH_4 , a constant heat of adsorption, a non-isothermal energy balance within the vessel, and a uniform packing density and void fraction in the adsorbent bed. The energy balance accounts for heat diffusion between adsorbate and adsorbent. Additionally, a variation in axial dispersion along the bed under plug flow conditions was assumed. The radial variations were considered negligible, and the velocity distribution was related to a momentum balance. The porous polymeric beads used as adsorbent and the linear driving force (LDF) model with experimental mass transfer coefficients (MTCs) reported in [20] were used to predict mass transfer from gas to particle.

The purity and recovery of CH_4 in the product stream are calculated using Equations (11) and (12).

$$Purity_{CH_4}(\%) = \frac{\int_0^{t_{AD}} c_{CH_4 P} u_s dt}{\sum_{i=1}^n \int_0^{t_{AD}} c_{i P} u_s dt} \times 100 \quad (11)$$

$$Recovery_{CH_4}(\%) = \frac{\int_0^{t_{AD}} c_{CH_4 P} u_s dt - \int_0^{t_{PG}} c_{CH_4 P} u_{sPG} dt}{\int_0^{t_{AD}} c_{CH_4 f} u_s dt + \int_0^{t_{RP}} c_{CH_4 f} u_{sf} dt} \quad (12)$$

where, $c_{CH_4 P}$, and $c_{i P}$ are the concentration in the product stream of CH_4 and component i , respectively, $c_{CH_4 f}$ is the concentration of CH_4 in the feed stream, and u_s is the superficial

velocity with the subscripts P, PG, and f representing the product, purge gas, and feed streams, respectively.

The purity and recovery of biomethane by a twin double-bed PSA process were calculated using Equations (13) and (14)

$$\text{Twin bed purity}_{\text{CH}_4}(\%) = \frac{\frac{r_{\text{PSA1}}}{100} \times \text{Purity}_{\text{PSA1}} + \frac{(100-r_{\text{PSA1}})}{100} \times \frac{r_{\text{PSA2}}}{100} \times \text{Purity}_{\text{PSA2}}}{\frac{r_{\text{PSA1}}}{100} + \frac{(100-r_{\text{PSA1}})}{100} \times \frac{r_{\text{PSA2}}}{100}} \quad (13)$$

$$\text{Twin bed recovery}_{\text{CH}_4}(\%) = r_{\text{PSA1}} + (100 - r_{\text{PSA1}}) \times r_{\text{PSA2}} \quad (14)$$

where r represents recovery, and the subscripts PSA1 and PSA2 designate it to the 1st PSA process or the 2nd downstream PSA process.

The power of the inter-unit compressor in the twin double-bed PSA is calculated by Equation (15) [36]:

$$\text{Power} = \dot{n} \times Mw \left(\frac{n_s}{n_s - 1} \right) f \left(\frac{P_1}{\rho_1} \right) \left[\left(\frac{P_2}{P_1} \right)^{\frac{n_s-1}{n_s}} - 1 \right] \quad (15)$$

The compressor inlet gas molar flow rate, pressure, and density are represented by $\dot{n}$, P_1 , and ρ_1 , respectively. The term n_s is the isentropic expansion factor and is broken down in Equation (16). The term f is calculated by Equation (17) and is used to correct the compressor ‘head’ with the variation of n_s [36]:

$$n_s = \frac{\ln \left(\frac{P_2}{P_1} \right)}{\ln \left(\frac{\rho_2'}{\rho_1} \right)} \quad (16)$$

$$f = \frac{h_2' - h_1}{\left(\frac{n_s}{n_s-1} \right) \left(\frac{P_2}{\rho_2'} - \frac{P_1}{\rho_1} \right)} \quad (17)$$

The compressor is modelled as isentropic and is therefore a constant entropy process. As a result, the enthalpy and density of the exit stream, h_2' and ρ_2' , are defined as functions of the inlet stream entropy. The energy consumption of vacuum pumps during the VA and PG steps is calculated using Equation (18) [19,37].

$$\text{Power} = \left(\frac{n_s}{n_s - 1} \right) RT_{gf} \left[\left(\frac{P_{\text{atm}}}{P_{\text{va}}} \right)^{\frac{n_s-1}{n_s}} - 1 \right] u_{sf} c_{gf} \pi r_{bi}^2 \quad (18)$$

u_{sf} and c_{gf} are superficial velocity and concentration of gas at bed inlet.

3. Results and Discussion

This paper sets out to systematically perform a sensitivity analysis on the impact of numerous parameters, including PG/F, adsorption time, temperature, and vacuum level, on the performance of cyclic adsorption processes (CAP). Furthermore, considering the state-of-the-art studies, it can be concluded that PSA, PVSA, and twin double-bed PSA are the most common adsorption processes to upgrade the biogas; therefore, this paper aims to compare their performance for producing biomethane by means of recovery and energy consumption. Finally, a certain guideline for specifying the minimum purity requirement of biomethane for every adsorbent will be presented.

3.1. Effect of Purge to Feed Ratio

The portion of product recycled back to the bed undergoing the regeneration step was designated as the PG/F ratio. This step was necessary to recover the adsorption capacity of the saturated adsorbent bed. Table 2 summarises the performance of a PSA process

producing biomethane using PPB adsorbent over a range of PG/F at 25 °C, 6 bar, and an adsorption time of 45 s, with a step breakdown depicted in Figure 2b. It was observed that an increase in PG/F resulted in a drop in recovery (Figure 3a) due to the reduction in the total product flow. This is aligned with Lee et al.'s [38] work, which experimentally investigated the effect of the purge-to-feed ratio on biomethane purity. This was caused by the higher fraction of product rerouted to the PG stream, regenerating the parallel bed. To investigate the reason for this trend, the loading after reaching cyclic steady state (CSS) conditions is depicted in Figure 3b. The CO₂ loading at the vessel inlet during the AD step was unaffected by the PG/F decrease and was approximately identical in both Case PG1 and Case PG4. However, a lower CO₂ loading at the bed median at the start of the AD step was observed in Case PG1, which employs a higher PG/F of 20%. This lower level of saturation in the bed median at higher PG/F flow was thus beneficial to the AD step and ensured an optimised level of active sites for larger adsorption of CO₂. This indicated that by increasing the PG/F ratio, CO₂ loading at the end of the regeneration step is lower in the bed (Figure 3c). Accordingly, a larger amount of CO₂ is adsorbed during the adsorption step, resulting in enhanced biomethane product purity. During the purge step, since the gas stream enters from the bed end, the local sorbent adsorbed less impurity. Thus, sorbents adsorb impurities from the product stream rather than desorbing and being regenerated. This is indicated by the presence of a peak in CO₂ loading at a distance of ~0.13 m.

Table 2. Effect of PG/F on performance of PSA process for biogas upgrading, $t_{\text{ads}} = 45$ s, $T = 25$ °C.

Case No.	PG/F (%)	CH ₄ Purity (%)	CH ₄ Recovery (%)
PG1	20	98.8	49.6
PG2	15	97.9	55.3
PG3	10	95.5	61.1
PG4	5	92.4	70.2

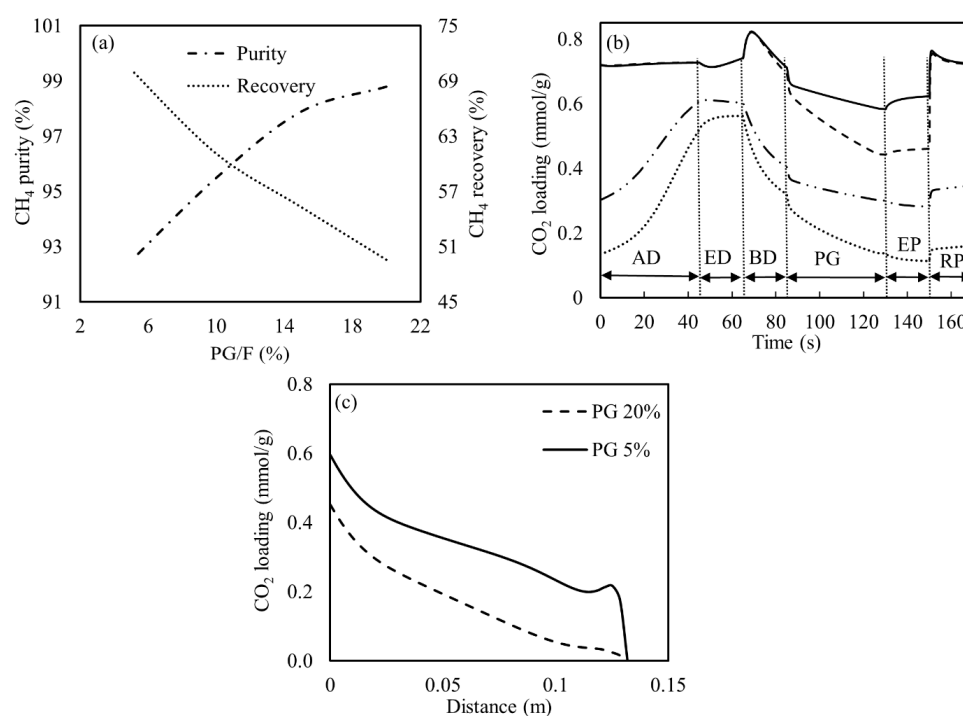


Figure 3. Investigating the effect of purge to feed (PG/F) on: (a) CH₄ purity and recovery, (b) CO₂ loading at bed inlet and bed median after CSS conditions for Case PG1 (PG/F = 20%) and Case PG4 (PG/F = 5%) Legend: CO₂ loading at --- bed inlet for Case PG1 (PG/F = 20%), — bed inlet for Case PG4 (PG/F = 5%), bed median for Case PG1 with PG/F = 20%, — · — · — bed median for Case PG4 with PG/F = 5%, and (c) CO₂ loading along the bed at end of PG step after CSS conditions.

3.2. Effect of Adsorption Time

Table 3 lists a PVSA process's performance parameters as a function of adsorption time at CSS conditions as per the steps outlined in Figure 2a. Results for purity and recovery were recorded at a fixed temperature of 25 °C, PG/F of 5%, and vacuum desorption pressure of 0.1 bar.

Table 3. Effect of adsorption time on purity, recovery, and energy consumption in a PVSA process with: T = 25 °C, PG/F = 5%, Vacuum = 0.1 bar.

Case No.	t_{ads} (s)	CH ₄ Purity (%)	CH ₄ Recovery (%)	Feed Compressor (kJ/kg CH ₄)	Vacuum Compressor (kJ/kg CH ₄)	Total Energy (kJ/kg CH ₄)
t1	45	99.96	58.78	1168	475	1643
t2	65	99.81	67.24	1009	386	1395
t3	85	98.99	72.79	928	384	1312
t4	105	95.35	76.18	920	355	1275
t5	115	92.06	78.13	868	346	1214
t6	125	89.05	79.58	852	339	1191
t7	145	83.99	81.87	828	308	1136

Note: t_{ads} : adsorption time.

Referring to Figure 4a, as adsorption time increased, the biomethane purity dropped but led to a higher recovery. During the adsorption stage, more mass of adsorbent is saturated with time, which leads to a drop in purity. However, a longer adsorption time leads to more product, resulting in a higher recovery. A trade-off between purity, recovery, and energy consumption was observed in such a manner that by targeting a higher purity, the recovery decreased and more energy was required, Figure 4b. This observation is aligned with Witte et al.'s [39] work on the impact of adsorption duration on PSA performance. Since countries vary in the minimum specifications attributed to commercial biomethane purity, it was necessary to determine the impact a certain guideline for a minimum biomethane purity has on energy consumption. This was achieved by correlating energy consumption at various adsorption times (Figure 4b). A decrease in adsorption time from 65 (Case t2) to 45 s (Case t1) led to a negligible increase in purity from 99.81% to 99.96%, while the required energy consumption increased by 18%, from 1395 to 1643 kJ/kg CH₄. In comparison, the decrease in adsorption time from 105 s (Case t4) to 85 s (Case t3) resulted in an increase in product purity from 95.35% (Case t4) to 98.99% (Case t3) while raising the compressor duty by only 2.9%. Point I in Figure 4b showed that at purity levels above 98%, there was a considerable drop in recovery, while point II showed that at purity levels above 99%, the energy consumption increased considerably. As a result, achieving CH₄ purity of >98% becomes less feasible for commercial applications due to the high energy consumption accompanied by a steep recovery drop; thus, a minimum target of 98% is recommended due to the involved cost-benefit implications.

Figure 5 maps the CO₂ and CH₄ loading along the adsorbent bed for Cases t1 and t7 with adsorption times of 45 and 145 s, respectively. An analysis of the CO₂ loading profile (Figure 5a) shows that at 45 s AD time, the CO₂ loading in the bed decreases sharply beyond 0.05 m down to a value of zero, indicating the high purity achieved under this condition. On the other hand, the longer adsorption time of 145 s saturates the full bed length at an overall moderate rate. The higher CO₂ loading at the bed end causes the possibility of CO₂ carrying over into the product stream and thus lowers overall biomethane purity. For insight on the effect of adsorption time on product recovery, Figure 5b depicts the CH₄ loading along the bed at the end of the AD step for Cases t1 and t7 with adsorption times of 45 s and 145 s, respectively. The high CH₄ loading for Case t1 with a lower adsorption time shows that much of the input CH₄ is adsorbed; thus, less CH₄ is leaving the bed, leading to a considerable reduction in CH₄ recovery.

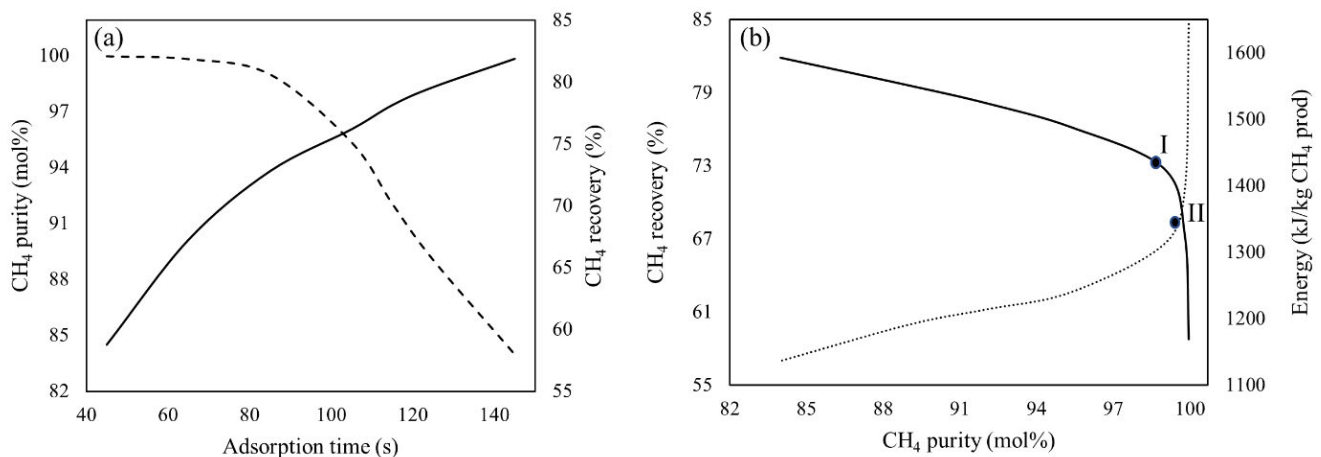


Figure 4. (a) Effect of adsorption time on purity and recovery. (b) Effect of purity on recovery and energy consumption. Legend: — recovery, --- purity, and energy consumption.

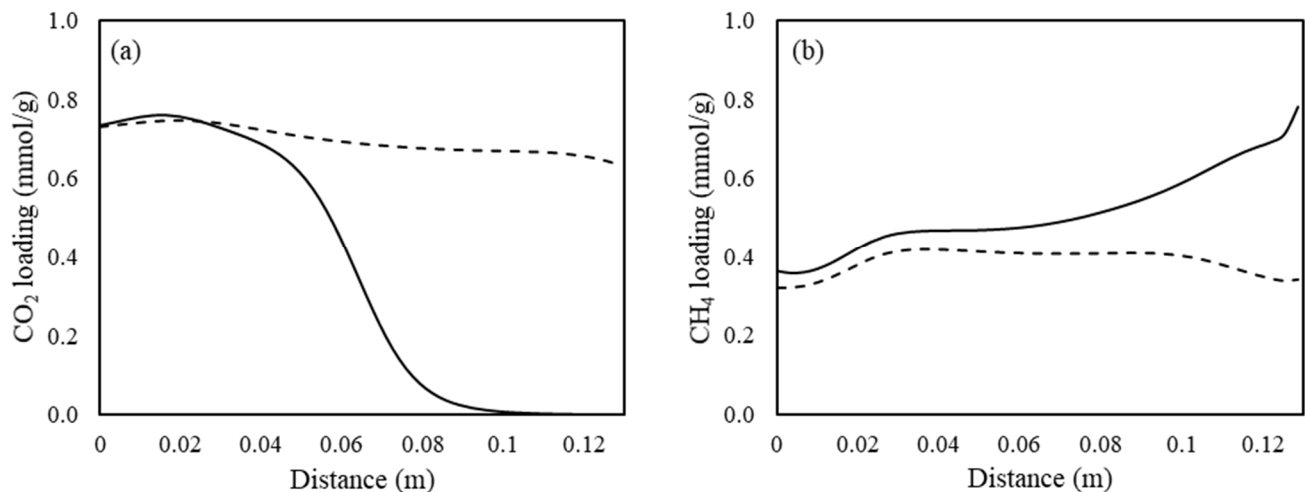


Figure 5. Loading along the bed at end of AD step for Cases t1 and t7 with adsorption times of 45 s and 145 s, respectively. (a) CO₂ (b) CH₄. Legend: — Case t1 at AD time of 45 s, --- Case t7 at AD time of 145 s.

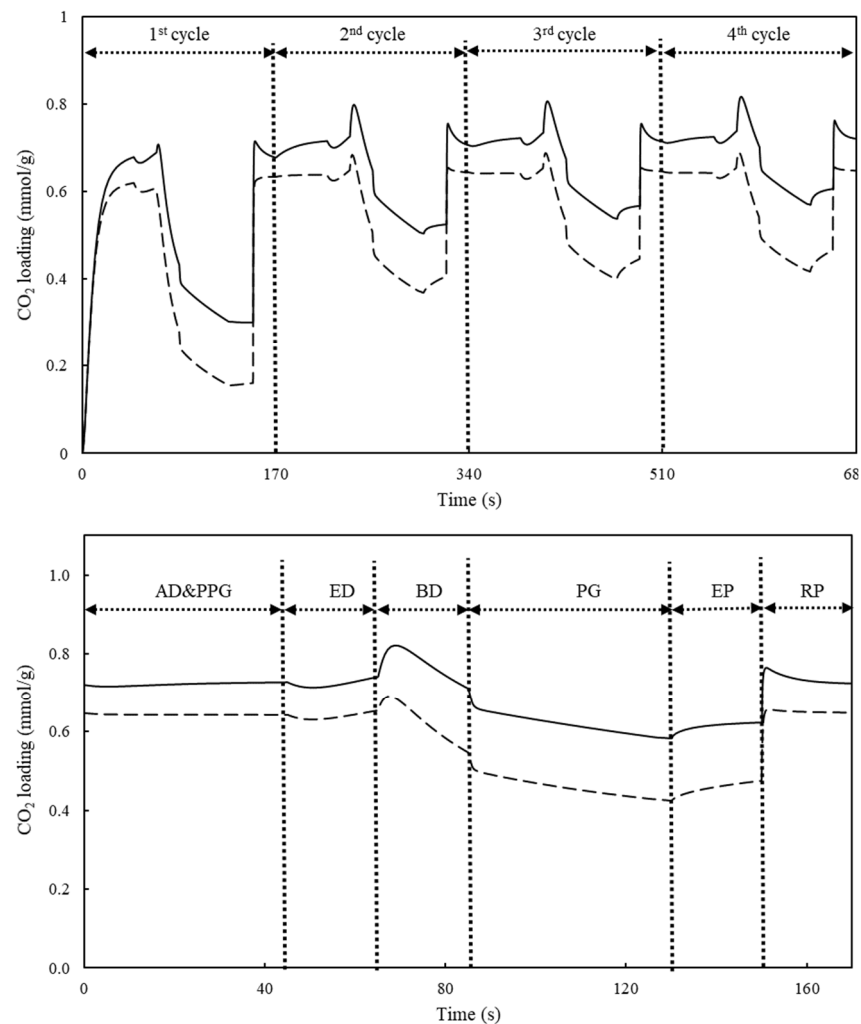
3.3. Effect of Temperature

The effect of temperature on the performance parameters of a PSA process was investigated (Table 4). All results are reported at CSS conditions and follow the steps described in Figure 2b. Case T1 presents the results of a PSA run at process conditions of 25 °C, 6 bar, an adsorption time of 45 s, and a PG/F ratio of 5%. Case T2 was allocated the same conditions as Case T1, except with a higher biogas feed temperature (35 °C). This led to a 9.2% and 14% drop in purity and recovery, respectively. This can be caused by the inverse relation of temperature to adsorption capacity, a characteristic of traditional adsorbents employing separation by physisorption [40].

To investigate the cause of the drop in purity and recovery as a result of a temperature increase, the CO₂ loading of unsteady-state cycles 1 to 4 (Figure 6 top) and under CSS conditions (Figure 6 bottom) was investigated at a 45 s adsorption time. Figure 6a shows that the first two cycles have a very different pattern. However, in subsequent cycles, the patterns become more similar until CSS conditions are reached. At a higher temperature, an improved regeneration of the adsorbent bed is observed but a lower overall CO₂ loading in the AD step. This confirms the dependency of PPB adsorbent-adsorbate interactions on the temperatures studied, with lower temperatures favouring the adsorption of CO₂ rather than the desorption.

Table 4. Effect of temperature on purity and recovery, PG/F = 5%.

Case No.	t_{ads} (s)	Temp. ($^{\circ}\text{C}$)	CH_4 Purity (%)	CH_4 Recovery (%)
T1	45	25	92.5	70.2
T2	45	35	83.96	60.2
T3	40	35	87.39	57.5
T4	35	35	91.17	54.3
T5	33	35	93.32	47.8

**Figure 6.** Temperature effect on CO₂ loading at 25 $^{\circ}\text{C}$ (Case T1) and 35 $^{\circ}\text{C}$ (Case T2) at bed inlet for a PSA process at (top) various cycles, (bottom) cyclic steady state (CSS) conditions; Legend: — Case T1 with biogas feed temperature of 25 $^{\circ}\text{C}$, - - - Case T2 with biogas feed temperature of 35 $^{\circ}\text{C}$.

The effect of temperature on the performance of a PSA process at a given purity of ~93% was explored. The product purity in Case T2 was increased to 93.3% by decreasing the adsorption time from 45 s to 33 s (Case T5), allowing a reasonable comparison with the performance of Case T1. The resulting recovery at 35 $^{\circ}\text{C}$ to achieve a ~93% purity dropped to 47.8% compared to the 70.2% obtained at 25 $^{\circ}\text{C}$ for Case T1. Hence, a lower temperature favours a higher recovery in a PSA process due to an increase in adsorption time and higher adsorption capacity. As adsorption temperature increases, the diffusion rate of CO₂ into the adsorbent pores increases; however, weak Van der Waals intermolecular forces, which characterise physisorption, may not overcome the increase in internal energy of CO₂, resulting in a drop in overall loading capacity [41].

3.4. Comparison of PSA, PVSA, and Twin Double-Bed PSA

The performance of a PSA, PVSA, and twin double-bed PSA unit was compared based on the required energy consumption and maximum achievable recovery and purity. The process parameters used for the results in Table 5 are a biogas feed composition of 40:60 (vol%) CO₂/CH₄, 25 °C temperature, and a PG/F of 5%. The components considered for energy consumption include the compressor for biogas feed pressurisation, the PVSA-specific vacuum pump for evacuation, and the double PSA-specific second feed compressor. Referring to Case V1 and Case V4 in Table 5, a higher CH₄ purity is achievable in a PVSA at any identical adsorption time, considering Section 3.2, which demonstrated that in a PVSA process, the product purity decreases as adsorption time increases. Comparing the PSA (Case V1) and twin double-bed (Case V2) processes, it can be seen that the introduction of a secondary PSA unit in tail gas significantly enhanced the recovery from 70.0% to 90.0%, with a slight increase in required energy from 869 to 903 kJ/kg CH₄. However, the purity drops by almost 6%. This observation is aligned with the work conducted by Augelletti et al. [10].

Table 5. Effect of vacuum on purity, recovery, and energy consumption of twin double-bed PSA and a PVSA process with: T = 25 °C, PG/F = 5%.

Case No.	t _{ads} (s)	Type	P _{VA} (bar)	CH ₄ Purity (%)	CH ₄ Recovery (%)	1st Compressor (kJ/kg CH ₄)	2nd Compressor (kJ/kg CH ₄)	Total Energy (kJ/kg CH ₄)
V1	45	PSA	1	92.5	70.2	869		869
V2	45	TD PSA	1	87	90	869	34	903
V3	55	PVSA	0.6	93.1	71.2	948	56.5	1005
V4	55	PVSA	0.5	95.4	67.5	1008	83	1091
V5	65	PVSA	0.5	93.1	71.1	958	82	1040
V6	85	PVSA	0.5	87.8	75.6	897	77	974
V7	90	PVSA	0.5	86.5	76.6	886	75	961
V8	55	PVSA	0.1	99.9	63.5	1074	387	1461
V9	65	PVSA	0.1	99.8	67.2	1009	386	1395
V10	85	PVSA	0.1	98.9	72.8	928	384	1312
V11	105	PVSA	0.1	95.3	76.2	920	355	1275
V12	115	PVSA	0.1	92.0	78.1	868	346	1214
V13	125	PVSA	0.1	89.0	79.6	852	339	1191
V14	132	PVSA	0.1	87.0	80.4	842	327	1169
V15	132	PVSA	0.01	87.9	80.4	842	874	1716

Note: T_{ads}: adsorption time, P_{VA}: vacuum pressure, TD PSA: twin double-bed PSA.

Comparing PVSA (Case V3) and PSA (Case V1) to achieve almost similar purity and recovery in both systems, a vacuum pressure of 0.6 bar is required, and the adsorption time should increase from 45 to 55 s. This results in a ~16% increase in energy requirement. Although the energy consumption of PSA was significantly lower, the process suffers from providing a higher recovery. On the other hand, it was found that a pressure vacuum of 0.6 bar was not adequate to provide acceptable recovery. In Case V4, to achieve a higher purity, the vacuum pressure was reduced to 0.5 bar. Accordingly, purity from the produced CH₄ increased to 95.4%. On the other hand, the recovery dropped to 67.5%. It is not reasonable to compare the PVSA results (Case V4) with the double PSA values (Case V2) since the output CH₄ purity is different. Hence, it was necessary to increase the adsorption time of the PVSA Case up to 90 s (Case V7) to achieve a purity equivalent to the double PSA Case of 87%. The CH₄ recovery of a double PSA for producing biomethane with 87% purity is 90%, while a PVSA with a vacuum level of 0.5 bar has a 76.6% recovery and a 961 kJ/kg CH₄ energy consumption, approximately the same value compared to the 903 kJ/kg for the twin double-bed PSA Case. By reducing the vacuum level from 0.5 (Case V7) to 0.1 (Case V14) or even 0.01 bar (Case V15), the adsorption time for producing biomethane with 87% purity increases, but the recovery increases from 76.6 to 80.4%, which is only a 5% rise. Therefore, vacuum levels below 0.5 bar do not have a considerable effect on recovery for

producing the same purity of CH_4 , but the required energy demand increases exponentially, Figure 7. Accordingly, when a high purity biomethane stream is required, the PVSA's ability to provide purity at moderate recovery and energy requirements justifies its widespread use within the industry for biogas upgrading. However, the double PSA arrangement minimises CH_4 loss and provides enhanced recovery at a reasonable energy demand that is unmatched by conventional PSA arrangements, making it a feasible alternative under low-purity biomethane regulations. The conventional PVSA processes with zeolitic materials require vacuum levels below 0.2 bar [10]. Thus, the recovery and energy demand of a twin double-bed PSA with 90% and 903 kJ/kg CH_4 , respectively, are superior to the corresponding PVSA process with 80% and 1169 kJ/kg CH_4 recovery (Case V14). In summary, shifting toward a lower vacuum level causes a negligible improvement in PVSA recovery and purity, but a significant increase in energy requirements.

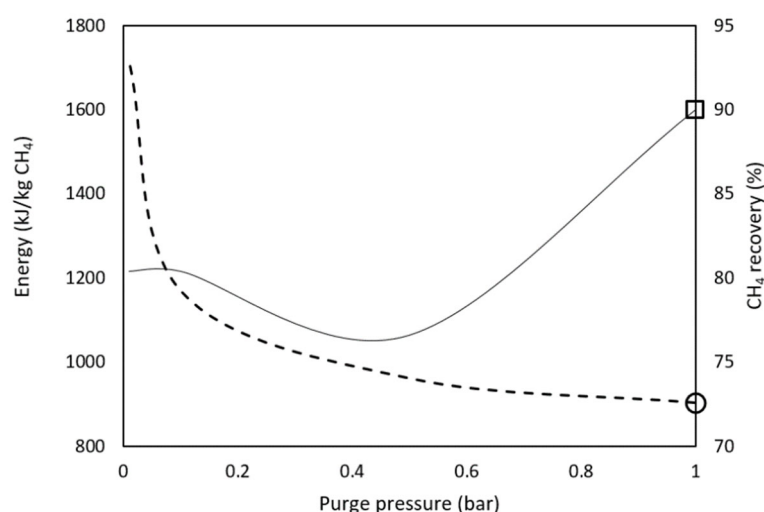


Figure 7. Comparison of twin double-bed PSA ($P = 1$ bar) with PVSA processes at different vacuum levels for producing 87% CH_4 purity. Legend: — CH_4 recovery of PVSA, --- energy consumption of PVSA. ○ energy consumption of twin double-bed PSA, □ recovery of twin double-bed PSA.

4. Conclusions

The high energy consumption and/or low recovery are the main obstacles to the widespread rollout of cyclic adsorption processes for biogas upgrading to commercial-grade biomethane. The high energy consumption is mainly due to the vacuum requirement in the pressure vacuum swing adsorption (PVSA) processes that are used for the current state-of-the-art biogas upgrading units. Accordingly, this study evaluated PSA, PVSA, and twin double-bed PSA processes over a wide range of operating conditions to map the optimum configuration and operating parameters for balancing purity, recovery, and energy consumption. It was found that at similar conditions, the energy consumption and recovery of a twin double-bed PSA and conventional PVSA process were 903 and 961 kJ/kg CH_4 , accompanied by a recovery of 90% and 76%, respectively. The effect of operating conditions, including adsorption time, PG/F ratio, biogas feed temperature, vacuum level, and process configuration, was studied to determine the significant parameters for enhancing the recovery and lowering energy consumption of either the PSA or PVSA processes. It was observed that an increase in adsorption time from 45 s to 145 s caused the purity to drop by ~16% from 99.96% to 83.99%, while recovery increased ~39% from 58.78% to 81.87%. The increase in temperature from 25 °C to 35 °C resulted in a ~32% drop in recovery for producing biomethane with ~93% purity. It was concluded that there is a more significant trade-off between recovery and energy consumption for units loaded with adsorbents with a moderate vacuum requirement (>0.5 bar). A higher recovery with reasonable energy demand is more achievable by a twin double-bed PSA process, while a higher purity at moderate recovery is better attained by a PVSA process.

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