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Effect of pellet size on PSA performance: monolayer and multilayer bed case study for biogas upgrading

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Abstract

A demand-driven pressure swing adsorption (PSA) biogas upgrading application is modelled using monolayer and multilayered (bilayer) beds, to gain insight on the impact of the adsorbent pellet size on the overall performance of such processes. Pellet radii in the range of 0.1-2.4 mm were studied. Varying the pellet size influences the sorption kinetics and flow resistance, resulting in the existence of an optimum pellet size for monolayered beds. Small pellets may yield higher purities at low total productivities, yet show a more rapid decrease in product purity with increasing productivities due to the higher pressure drop. Furthermore, 18 configurations with beds containing a layer of larger pellets and a second layer of smaller pellets (bilayer) were investigated. Bilayered beds with 0.3 mm, 0.6 mm and 2.4 mm radius pellets were combined, with the first layer taking up 25, 50 or 75% of the bed. Such beds offer more freedom in influencing the overall kinetic mechanisms and pressure drop. With respect to upward flow in the adsorption step, beds with the smallest pellet size in the top layer (LS beds) offer higher product purity than beds with the smallest pellet in the bottom layer (SL beds). Compared to monolayer beds with a well-chosen pellet size, no significant performance gains for LS bilayer beds were noted in this study.

Keywords

Layered bed, pellet size, biogas, model, pressure swing adsorption

1 Introduction

Pressure swing adsorption (PSA) has become an established separation technique with widespread industrial use such as in hydrogen separation or oxygen separation, using an adsorbent bed selective

for target species within the feed gas mixture. Often unstructured (random) packed beds, containing beads or extrudates, are used. The size and shape of the adsorbent material play an important role in the performance of PSA units, impacting the adsorption kinetics, flow resistance, heat transfer and flow distribution. Alpay et al. studied the impact of particle size in the 0.15-0.71 mm range for a rapid PSA (RPSA) application using a two-step cycle with zeolite 5A. An optimum in particle size was found to exist depending on process conditions, limited by two key elements: (1) ineffective pressure swing and (2) diffusional limitations (Alpay, Kenney and Scott, 1994). For a larger PSA installation, Shigaki et al. examined 3.0 mm and 1.5 mm diameter cylindrical pellets of zeolite 13X in a CO_2 adsorption process (Shigaki *et al.*, 2018). The larger pellets were preferred due to the lower energy consumption because of the lower pressure drop they generate, whilst the kinetic mechanisms were such that the effect of the particle size was small. For kinetic based PSA separations, smaller particles bring the separation closer to equilibrium and can make the separation more difficult (Shirley and LaCava, 1995). In their simulation study, Nikolic et al., performed a parametric analysis of the particle size for two hydrogen recovery cases, and reported that a smaller particles size is preferred (Nikolic, Kikkinides and Georgiadis, 2009).

Adsorbent beds containing more than one adsorbent layer are also frequently encountered in PSA processes (Chlendi and Tondeur, 1995; Park, Kim and Cho, 2000; Rege *et al.*, 2001; Baksh and Ackley, 2002; Wilson and Webley, 2002; Ahn and Lee, 2003; Golden and Weist, 2004; Cavenati, Grande and Rodrigues, 2006; Grande and Rodrigues, 2007; Ribeiro *et al.*, 2008; Nastaj and Ambrozek, 2015; Sheikh Alivand and Farhadi, 2018; Xiao *et al.*, 2020). Such multilayered beds are typically comprised of layers having fundamentally different adsorption properties (e.g. selected specifically to adsorb a different target component). Perhaps the most well-known examples are guard beds (e.g. activated alumina) intended to protect the main adsorption layer (e.g. zeolite) from damage or decreased performance by strong adsorption of trace components such as water or carbon monoxide. On the other hand, layering can also be achieved by using the same material in a different size or shape. Such layered configurations allow for more flexibility in tuning the column dynamics and pressure drop. For

example, Mathews (Mathews, 2005) applied uniform as well as eight-layered activated carbon (AC) beds for the liquid-phase adsorption from phenol, with two column designs (cylindrical and tapered). The column using the smaller pellets at the exhaust (reverse stratified) showed steeper breakthrough profiles, beneficial for the separation process. The patent of Baksh and Simo (Baksh and Simo, 2012) describes the use of five-layered beds, combining AC, zeolite and activated alumina. The zeolite and AC layer consisted out of two sublayers, with different pellet size. Within such a subdivided layer, the first sublayer encountered by the feed flow had a smaller pellet size than the one above. This arrangement is proposed based on relative adsorption rates, with each of the sublayers intended to adsorb a target component as CO or nitrogen. Layers of different pellets size, with the larger pellets presented to the feed first, have also been reported in patents (Miller, 1990).

In this work we aim to gain insight on the impact of the pellet size, uniform and layered, on the PSA performance trends for a demand-driven application (De Witte, Denayer and Van Assche, 2021). To this purpose, a PSA model to upgrade CO_2/CH_4 (biogas proxy) using FAU zeolite was used as a case study, where the production rates were varied. First, a monolayer bed is discussed where the pellet radius is altered from 0.1-2.4 mm. In addition, PSA units with a bilayered bed of various pellet sizes (using 0.3 mm, 0.6 mm or 2.4 mm radius pellets) were simulated, and their performance in a PSA application compared to the monolayered beds.

2 Methods

A biogas upgrading PSA application was selected as a case study. In the system under consideration, carbon dioxide is adsorbed onto FAU (13X) zeolite from its gas feed of 33 mole% CO_2 and 67 mole% CH_4 to produce a methane rich product gas. The non-isothermal model is developed in Aspen Adsorption V9, based upon the model of De Witte et al. (De Witte, Denayer and Van Assche, 2021), and the cycle based upon the PSA application described by Santos et al. (Santos, Grande and Rodrigues, 2011) using 0.6 mm radius pellets. The PSA model of De Witte et al. (De Witte, Denayer and Van Assche, 2021) is characterized by the use of an extended Langmuir equilibrium model and the use of linear C_V valve models. The pressure drop over the bed was modelled using Ergun's equation

(Ergun, 1952), accounting for the impact of the pellets size on the flow and pressure profiles. Furthermore, the axial dispersion coefficient and heat transfer are affected by the pellet radius (supplementary information). In this work, the kinetic model of De Witte et al. (De Witte, Denayer and Van Assche, 2021) was adapted to a lumped linear driving force in the solid accounting for film, macropore and micropore diffusion. Two non-isothermal column calculation blocks, representing both columns of the dual bed unit, were solved simultaneously until convergence to cyclic steady state (CSS). Details on the model can be found in the supplementary information and previous work (De Witte, Denayer and Van Assche, 2021).

The PSA cycle is shown in Figure 1, consisting of eight timesteps: (**1**) a 50 s pressurisation step to 4 bara, (**2** and **3**) a 350 s production step where the purge valve is opened after 100 s, delivering a constant purge flowrate in step 3, (**4**) a 25 s top-top depressurization equalization step, (**5** and **6**) 150 s blowdown to 0.3 bara, subdivided here to synchronize with steps 1 and 2, (**7**) 250 s purge step and finally (**8**) a 25 s pressurization equalisation step.

First, monolayer beds with pellet sizes of 0.1 mm, 0.3 mm, 0.6 mm, 1.2 mm, 1.8 mm and 2.4 mm radius were tested. Except the pellet size, no alterations were made to the bed properties and therefor the simulated adsorbent mass within the units remains identical. Next, bilayered beds with layers of 0.3 mm, 0.6 mm or 2.4 mm radius pellets were tested in 0.3/0.6, 0.3/2.4 and 0.6/2.4 mm pairs. For these three pairs, the smallest pellet can be used in either the top (LS bed) or bottom layer (SL bed). Furthermore, the bottom layer height was set at 25, 50 or 75% of the column length. Accordingly, six multilayer configurations were tested for each of the three pellet pairs (Figure 2). In total, six monolayer bed and 18 multilayer (bilayer) bed configurations were simulated.

For all bed configurations, multiple CSS runs were performed by imposing various total product flowrates withdrawn from the methane product buffer tank. This method mimics a demand driven PSA application (De Witte, Denayer and Van Assche, 2021). Three key performance indicators are monitored: methane product purity, methane recovery and total productivity.

$$Purity = \frac{N_{CH_4,prod}}{N_{CH_4,prod}+N_{CO_2,prod}} = \frac{N_{CH_4,prod}}{N_{prod}} \qquad \text{eq. 1}$$

$$Recovery = \frac{N_{CH_4,prod}}{N_{CH_4,feed}} = \frac{Purity}{y_{CH_4,feed}} \left(\frac{N_{prod}}{N_{feed}}\right) \qquad \text{eq. 2}$$

$$Total\ productivity = \frac{N_{prod}}{t_{cycle}W_{ads}} \qquad \text{eq.3}$$

The total productivity is a controlled parameter and calculated using the cycle time t_{cycle} and adsorbent mass W_{ads} in the unit. The total productivity is defined here for the total amount of product mixture leaving the buffer tank, rather than considering only the light component fraction (methane productivity), which is more often used in literature (Li *et al.*, 2008; Santos, Grande and Rodrigues, 2011). The purity of the product flow is calculated based on the total moles N_{prod} of product obtained per cycle, and the recovery is calculated based on the total moles of product and feed per cycle. All bed configurations were simulated using the same cycle including the cycle step times and valve settings.

3 Results and discussion

3.1 Pressure drop and mass transfer in monolayered beds

It is well established that the pellet size and shape have a profound impact on the flow resistance in packed beds (Allen, von Backström and Kröger, 2013). Figure 3(left) shows the average pressure in the bed during the adsorption step (step 3), in function of total productivity. Total productivity is controlled by setting the product flow rate from the methane buffer tank. The average pressure inside the columns lowers with decreasing pellet radius and increasing total productivity (indicative of higher feed flow rates) in a nearly linear fashion. The velocities in the bed are well below 1 m/s, yielding a near linear dependency of the pressure gradient with gas velocity according to Ergun's equation. Overall, the pressure drop remains relatively small, even for the 0.3 mm radius pellets: the column reaches a final average pressure differing less than 0.1 bar from the 4.0 bara feed pressure applied on the feed valve. To explore the effect of larger pressure drops, smaller pellets (0.1 mm radius) were tested (not shown), although these small pellets are typically only applied in RPSA (Alpay, Kenney and Scott, 1994; Moran, Patel and Talu, 2018). Such small pellets yield a more pronounced pressure drop

(>0.15 bar), with the average pressure in the column at 3.82-3.62 bara over the reported productivity range.

Such pressure effects also play during the low-pressure purging step (step 7, Figure 1), increasing the average column pressure above the 0.3 bara blowdown pressure. Accordingly, the beds with augmented pressure drop do not experience the full theoretical pressure swing from 4.0 to 0.3 bara applied on the valves. This yields detrimental effects as a lower uptake capacity in the production steps and a reduction of purging efficiency in the purge step. Often, the effect of pressure drop is only considered during specific PSA steps rather than the cumulative effect on the PSA performance (Moran and Talu, 2017). In this work, we will explore the impact of the pellets size (and related pressure drop) on global PSA performance.

Regarding adsorption kinetics, a linear driving force (LDF) in the solid film is assumed, where the mass transfer coefficient is lumped considering the gas film, macropore and micropore mass transfer resistances. For methane adsorption on zeolite 13X, crystalline diffusion coefficients in the order of 10^{-9} m^2/s are anticipated (Guo, Shah and Talu, 2007). Considering zeolite crystallites of 2 μm diameter, the mass transfer equations for methane are expected to be dominated by film and/or macropore diffusion and thereby impacted by the pellet size. With respect to CO_2 diffusion, Silva et al. found crystalline diffusion (Silva, Schumann and Rodrigues, 2012) to be dominant in binderless 13X pellets. They performed Zero-Length Column (ZLC) experiments and obtained a crystalline diffusion coefficient in the order of 10^{-15} m^2/s at 373 K, also used in this work. It was also demonstrated that the size of the beads did not influence the rate of desorption, indicating that pellet size does not influence this process. Still, Hu et al. performed ZLC experiments on commercial 13X pellets and showed the diffusion mechanism of CO_2 to be macropore diffusion controlled with an impact of the pellet size (Hu *et al.*, 2014). Using a frequency response technique, Hossain et al. indicate the dominance of macropore diffusion for carbon dioxide in commercial 13X pellets (Hossain *et al.*, 2019). In this work, based on the lumped model estimations (supplementary information), we anticipate the

dominance of CO_2 macropore diffusion for larger pellets (roughly a radius >1 mm), and micropore diffusion dominance for smaller pellets (roughly a radius <0.1 mm).

To characterize the mass transfer process within the PSA application, an average mass transfer coefficient (MTC) was calculated at CSS by considering the time and node averaged MTC over the PSA cycle. The results are shown in Figure 3(right) for CO_2. Overall, a slight impact of the total productivity can be noticed, and this reflects mostly the mild impact of the Reynolds number and film transfer coefficient on the overall mass transfer. The inverse of the average CO_2 MTC in Figure 3 (Fig. S1 supplementary information) is nearly independent (order 0.1) on the pellet radius for smaller pellet sizes (0.1-0.6 mm). At larger pellet sizes, this inverse MTC dependency on the pellet radius is near the order 1.4. This is in rough correspondence with the expected transition from a micropore diffusion dominated mechanism (order 0) to macropore diffusion (order 2 dependency on pellet radius), here calculated over a wide range of conditions encountered over the column and throughout the cycle. For methane, this order is more constant near a value of 1.9 (Fig. S1 supplementary information), in good correspondence to the expected macropore diffusion order 2.

The lumped kinetics model may also be compared to other studies and kinetic models for 13X. Rodrigues et al. (Grande and Rodrigues, 2007; Santos, Grande and Rodrigues, 2011) used a bilinear driving force model. Film, macropore and micropore transfer were considered, with CO_2 intracrystalline diffusion coefficients in the same order of magnitude as used in this study. Webley et al. used a constant LDF value for CO_2 capture using a VSA unit (Webley *et al.*, 2017).

Besides the impact of the pellet size on the pressure drop and kinetics, also axial dispersion is significantly influenced by the particle size (Moran, Patel and Talu, 2018). In the employed model, a larger particle size will enlarge the dispersion coefficient (eq. S.4 supplementary information), further broadening the mass transfer zone.

3.2 PSA performance of monolayered beds

Figure 4 shows the obtained methane product purity from monolayer PSA systems at CSS, in function of the total productivity. For systems with pellet radii of 1.2-2.4 mm, a higher purity is obtained for

the smaller pellet irrespective of the total productivity tested. In this 1.2-2.4 mm range, the PSA system benefits from more favourable kinetics with smaller pellet size. For the systems with pellet radii of 0.1-0.3 mm, a higher purity is obtained for the larger pellet irrespective of the total productivity tested. The impact of pressure drop is more significant in this 0.1-0.3 mm radii range, and large pellets may be preferred here to reduce the negative impact on the experienced pressure swing. An optimum (maximized purity for a given productivity level) pellet size thus emerges near a 0.6 mm radius. However, as pressure drop increases with total productivity, the pellet size resulting in the highest purity increases with total productivity. It is anticipated that just below the smallest total productivity simulated (<5.0 mol/h/kg), the highest purity is achieved by the 0.3 mm radius pellets. From the lowest value of 5.0 mol/h/kg the 0.6 mm pellets achieve the highest purity. At higher productivity values near 8.7 mol/h/kg total productivity, the 1.2 mm radius pellets take this role. Just above the largest total productivity used here (>11.2 mol/h/kg), this is anticipated to be the 1.8 mm radius pellets.

To investigate the result of Figure 4 in more detail, the CO_2 loading profiles for the 0.6 mm (Figure 5, top rows A and B) and 2.4 mm (Figure 5, bottom rows C and D) radii pellet beds are shown. The profiles are shown at a low 5.0 (row A and C) and higher 9.9 mol/kg/h (row B and D) total productivity, at the beginning (50 s) and the end (400 s) of the adsorption steps.

With increasing time, the CO_2 front penetrates deeper into the bed, where for the higher productivity (row B and D) CO_2 breaks through the bed at 400 s, thereby causing a significantly decreased methane purity with rising productivity. Generally, the concentration fronts for the 0.6 mm radius pellets are steeper than the 2.4 mm pellets (e.g. row A and C at 400 s) due to the increased mass transfer. For a lower total productivity setting (row A and C), the smaller pellets are favoured with respect to a high purity product. However, at higher total productivity (row B and D), the pressure drop also plays a more significant role. Here, a tail can be noticed in the profile at 50 s in the cycle (row B and D) , located at the top of the column. This indicates the fouling of the product end caused by the equalization pressurization step (step 8, Figure 1), as discussed previously (De Witte, Denayer and Van Assche, 2021). The tail is more pronounced for the 0.6 mm pellets (row B). For these smaller pellets, the higher

pressure drop results in a lowered uptake capacity and a faster moving concentration front. It can be noticed that the 0.6 mm bed is more saturated at 400 s, compared to the 2.4 mm radius pellet bed (row B and D).

Beyond the product purity and total productivity, the recovery is another key performance indicator. The recovery in function of total productivity is shown in Figure 6. The curves for the different 0.3 to 2.4 mm radii pellets are very similar (some sizes are omitted for clarity), yet minor differences may be observed. The calculated recovery can be scrutinized (eq. 2) in terms of a combination of three factors, (1) the molar product-to-feed ratio and (2) product purity, with the third factor (3) being a constant methane feed fraction. Advantageously, the product-to-feed ratio is slightly lower for the smaller pellets (0.3 mm and especially 0.1 mm) as a result of the lower averaged pressure in the columns and reduced blowdown losses. It should be remarked that the employed cycle has a fixed purge flowrate, unaffected by column pressures (in contrast to a purge orifice). Secondly, the purities are high for the small pellets at low productivities, thus yielding two small advantages adding up to a slightly higher recovery for the 0.1 and 0.3 mm radii pellets. At higher total productivity these small pellets suffer a stronger decrease in purity, resulting in a slightly lower recovery than the larger pellets.

3.3 Pressure drop in multilayered beds

From the above monolayer bed analysis, three pellet sizes were selected to build bilayered bed configurations: small 0.3 mm, intermediate 0.6 mm, and the largest 2.4 mm radius pellets. A distinction can be made whether the smallest or largest pellets are placed in the top layer. The configurations with the small pellets layer at the bottom and large pellets layer on top are denoted as SL beds. Similarly, layered beds with the large pellets at the bottom are denoted as LS beds (Figure 2). For multilayered beds, the flow resistance of a layer impacts the pressure experienced further along the flow path. Figure 7 gives the pressure profiles inside a column after 400 s in the PSA cycle, for two monolayered beds (0.3 mm or 2.4 mm radius) as well as two bilayered beds (0.3/2.4 mm pair) with 75% of bed taken by the 2.4 mm radius pellets. For the SL bed made up out of a 25% length 0.3 mm pellets and a top layer of 75% length with 2.4 mm pellets, the smaller pellets are first in the upwards

flow path. This results in a lower, near constant, pressure of 3.94 bara for the remaining top layer. On the other hand, for the LS bed, the pressure drop over the first large pellet layer is very small even though it takes up 75% of the bed length, yielding a higher average pressure than in the SL bed.

The cycle has both upward as downward flow steps (Figure 1). Regarding the downward flow purge step, a similar consideration as in Figure 7 can be made. The average purge pressure will be disadvantageously higher for SL beds. Both the high (adsorption) as low (purge) average column pressure are thus negatively impacted to a larger extent by SL beds than LS beds. Besides these effects, the flow behaviour in the other steps of the cycle is also influenced (such as pressurization rates). Here, we again note that the valve settings were not varied upon varying the pellet size.

3.4 Performance of multilayered beds

3.4.1 Multilayer bed using the 0.3 mm / 2.4 mm pair

First the effect of the bilayered configuration is studied by combining (non-optimal) small 0.3 mm radius pellets with significant pressure drop, and (non-optimal) larger 2.4 mm pellets showing more severe kinetic limitations. The performance of the 0.3/2.4 mm pair beds is shown in Figure 8 (left and right). These are divided in configurations with the smallest pellets (SL) or largest pellets (LS) in the first (bottom) layer. The length of the bottom layer takes up 0% (monolayer), 25%, 50%, 75% or 100% (monolayer) of the total bed length. Focusing on the SL beds first, Figure 8 (left) shows these beds do not yield purities in excess of those obtained with a 0.3 mm monolayer bed at low productivities. Only the 75%-0.3 mm/25%-2.4 mm layered bed is able to achieve similar purities of the 0.3 mm monolayer bed. The concentration front does not penetrate the last 25% (2.4 mm layer) of the bed to a large extent. At the largest total productivities near 11 mol/h/kg, the pressure drop plays a larger role in the performance and the purities drop monotonically with a rising fraction of the 0.3 mm bottom layer, from 0% (monolayer 2.4 mm) to 100% (monolayer 0.3 mm). Overall, the highest purities are always obtained by the 0.3 mm or 2.4 mm monolayer beds, and not the SL multilayered beds.

The results are different for the LS beds, as shown in Figure 8 (right). At low total productivities the LS beds with the 0.3 mm layer at the top obtain similar purities to a monolayer 0.3 mm bed. Even a top

layer of only 25% 0.3 mm pellets is able to increase the methane purity over the 2.4 mm monolayer bed (at given total productivity). At the largest total productivities, the pressure drop starts to dictate the obtained purity and the purity drops roughly with a rising fraction of the 0.3 mm layer, similar to the SL beds. Remarkably, the LS multilayer beds achiever higher purities than the (0.3 or 2.4 mm) monolayer configurations for intermediate total productivities (~6-8 mol/kg/h). Under such settings, the concentration front penetrates all the last (0.3 mm) layers of the LS beds to a significant extent. Here, the separation is able to benefit from the increased kinetics, whilst the first (2.4 mm) layer does reduce the impact on the experienced pressure swing. When considering the recovery, the impact of the multilayer beds is quite small. Also the differences between monolayer beds were shown to be small (Figure 6).

3.4.2 Multilayer bed using the 0.6 mm / 2.4 mm pair

Extending the study above (0.3 mm and 2.4 mm), multilayered beds were built using a performant 0.6 mm radius pellet layer, first combined with large 2.4 mm radius pellets. Considering the monolayer beds in this 0.6 mm-2.4 mm range, the improvement in purity by increased kinetics of smaller pellets is anticipated to mostly outweigh the negative impact of increased pressure drop. As above, the LS beds can be favoured over the SL beds (supplementary information). The SL layered beds (2.4 mm top layer) have almost similar purity-productivity curves as the 2.4 mm monolayer bed. Similar, LS beds (0.6 mm top layer) show very similar purity-productivity curves as the 0.6 mm monolayer bed. Only at low product flowrates from the methane buffer tank, do the LS beds with a 75% bottom (2.4 mm) layer tend more to their (2.4 mm) monolayer counterpart in terms of purity. Here, the concentration fronts do not penetrate deep in the last 25% of the bed. At the same imposed total productivities, LS beds can achieve (or even mildly surpass) the attained purities of the 0.6 mm monolayer configuration. This is especially the case at higher total productivities (>6 mol/h/kg), where the 1.2 mm monolayer bed was shown to yield higher purities than the 0.6 mm monolayer bed (Figure 4).

3.4.3 Multilayer bed using the 0.6 mm / 0.3 mm pair

Further extending the studies above (0.3/2.4 mm and 0.6/2.4 mm pairs), the multilayered beds are now built using 0.6 mm radius pellets, combined with small 0.3 mm radius pellets. Considering the monolayer beds in this 0.3 mm-0.6 mm range, the improvement in purity by increased kinetics of smaller pellets is anticipated to be largely offset by a larger negative impact of increased pressure drop. Again, the LS beds outperform the SL beds (supplementary information). Overall, the purity-productivity curves for the LS beds (0.6 mm bottom layer) are roughly similar to the curve of the performant 0.6 mm monolayer bed. However, at larger productivities the pressure drop plays a more prominent role and a higher purity is obtained with an increasing fraction of 0.6 mm bottom layer, avoiding the negative pressure effect of the 0.3 mm layer. For the SL beds (0.3 mm bottom layer), the purity-productivity curves are roughly similar to the curve of the 0.3 mm monolayer bed. Similar to the SL beds, at larger productivities the pressure drop plays a more prominent role and a higher purity is obtained with an increasing fraction of 0.6 mm top layer.

3.5 Analysis of multilayer bed effects

For this case study, the recovery is most strongly impacted by the total productivity and far less by the bed configuration (Figure 6). The achievable total productivity at a set 97% methane purity will be used as a single measure for the performance of the PSA units. These values are obtained by interpolation from the purity-productivity curves above (Figure 4 and Figure 8, supplementary information). The values are shown in Figure 9, for the investigated 0.3/0.6, the 0.6/2.4 and the 0.3/2.4 mm radius pellet pairs.

When transitioning from a monolayer (100%) small pellet bed to a monolayer larger pellet bed, via a bilayered bed configuration, an SL configuration (orange) or LS configuration (blue) approach can be applied. Overall, LS beds yield higher 97% purity productivities than the SL beds. When combining two non-0.6 mm pellets (monolayer performant), as the 0.3/2.4 mm pair, the achievable 97% productivity of both monolayer beds can be clearly surpassed by using LS beds. However, if one of the layers is already built using the (monolayer performant) 0.6 mm radius pellets, the achievable 97% productivity gains over the 0.6 mm monolayer bed are very mild (<1.5%) here. The best LS bed improvements over

the 0.6 mm monolayer bed are even within the error margins (considering interpolation and CSS material balance closure tolerances) of this study. In contrast, the SL beds yield low 97% purity productivities (up to 10% lower than 0.6 mm monolayer beds).

Beyond these theoretical aspects, the practice of selecting the particle size also depends on important aspects as the crush strength, the tendency for fluidization and abrasion, and pellet cost. Furthermore, the pellet size will also impact flow distribution and operators may alter their cycle and valve settings accordingly, thus adapting the cycle to the used bed. Creating a multilayered bed involves more complex filling procedures and the use of layer strainers. On the other hand, layered beds can be used to partially circumvent limitations of monolayer beds for small pellets (e.g. crush strength or pressure drop).

4 Conclusion

The effect of pellet size was investigated for various bed configurations for a modelled PSA biogas upgrading application. Monolayered beds with pellet sizes in the range of 0.1-2.4 mm radius were studied, where a balance between pressure drop and kinetic effects should be considered. For this example, a performant monolayer bed configuration emerged by using 0.6 or 1.2 mm radius pellets. Overall, the balance between improved kinetics and increased pressure drop is affected by the imposed total productivity in the demand driven application considered here. Furthermore, 18 different configurations of bilayered beds were tested with 0.3/0.6 mm, 0.3/2.4 mm and 0.6/2.4 mm radius pellet pairs. The bottom layer was 25, 50 or 75% of the length of the bed. For the tested example, LS beds where the smallest pellet layer is placed at the column outlet (top, outlet during adsorption step) outperform SL beds. The pressure swing is maintained to a larger extent for LS beds. LS bed performance gains over the monolayer beds are possible considering the 0.3/2.4 mm pair. Still, the gains in this study appear limited with respect to a well-chosen pellet size for the monolayer bed (0.6 mm in this case). Finally, some critical notes are made with respect to the practice of selecting the pellet size.

Supplementary information

Supplementary information is available.

Statements and Declarations

The authors have no relevant financial or non-financial interests to disclose.

Joeri F.M. Denayer is an editorial board member of Adsorption journal.

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Figure captions

Figure 1: PSA cycle scheme for the dual bed unit comprising eight steps

Figure 2: Graphical representation of pellet sizes, and bed configurations for monolayer and bilayer beds where small and large pellets are combined in six combinations with a layer taking up 25, 50 or 75% of the bed length

Figure 3: Average pressure in monolayer bed of varying pellet radius during PSA cycle step 3 (left), averaged linear driving force CO_2 mass transfer coefficient throughout PSA cycle (right)

Figure 4: Methane product purity in function of total productivity for monolayered beds with different pellet radii at CSS

Figure 5: CSS CO_2 solid loading profile (full line) and CO_2 fluid phase fraction (dashed lined) at the start (50 s) and end (400 s) of the production steps within the cycle. The 0.6 mm radius pellets are shown at (row A) 5.0 mol/h/kg and (row B) 9.9 mol/h/kg total productivity. The 2.4 mm radius pellets at (row C) 5.0 mol/h/kg and (row D) 9.9 mol/kg/h total productivity

Figure 6: Methane recovery in function of total productivity for monolayered beds with different pellet radii at CSS

Figure 7: Pressure profile at 400 s in the PSA cycle at 11.2 mol/h/kg total productivity for monolayered beds (0.3 and 2.4 mm radii pellets) and multi-layered beds (75% of the bed 2.4 mm) with a 0.3 mm layer on top or bottom

Figure 8: Methane product purity in function of imposed total productivity for monolayered and multilayered beds with pellets of 0.3 and/or 2.4 mm radius at CSS. (Left) Small pellet layer at the bottom SL beds, (right) small pellet layer at the top LS beds

Figure 9: Interpolated total productivity values at 97% methane purity for 18 multilayer configurations and three monolayer beds using 0.3, 0.6 and 2.4 mm radius pellets. These are given with the relative bed length of the layer with the largest pellet size