How does structured adsorbent channel heterogeneity influence the efficiency of adsorptive CO_2 capture?

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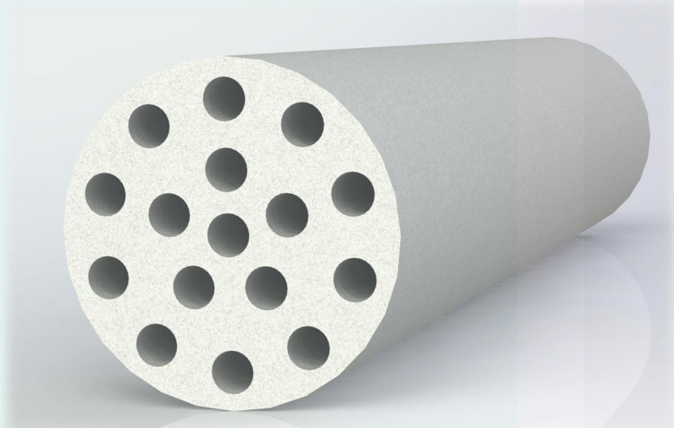
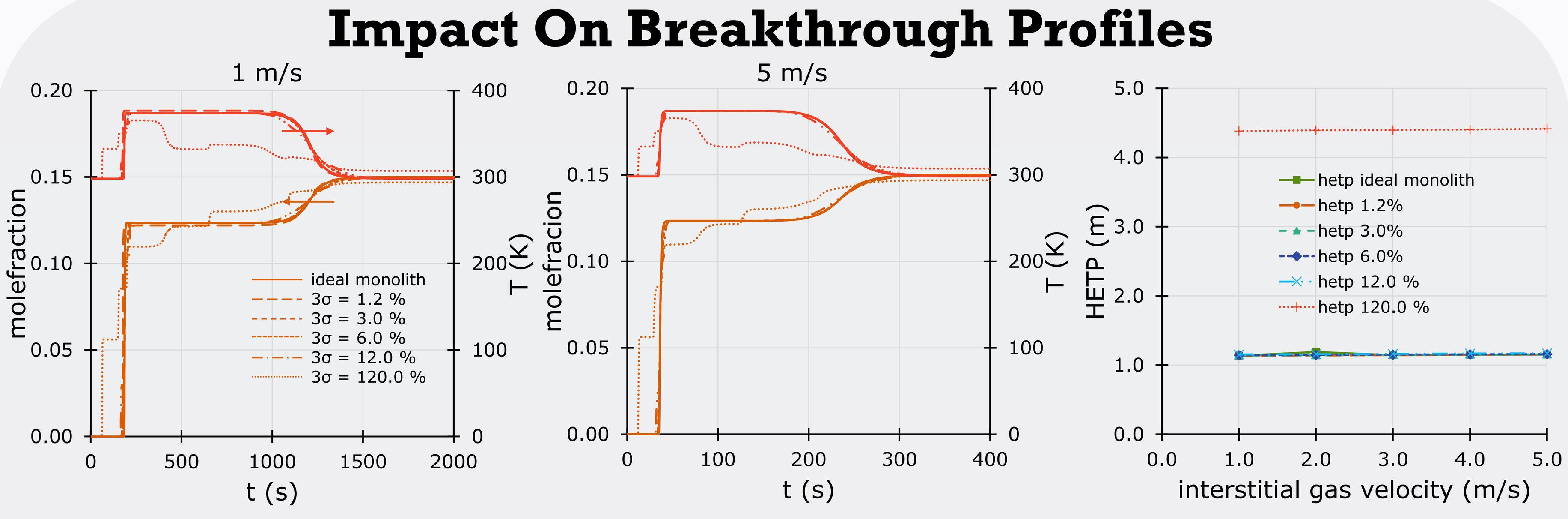
Adsorbent Monoliths are Heterogeneous

Adsorbent monoliths are often used in carbon capture applications, due to their favorable pressure drop and mass transfer characteristics. Monoliths are often imperfect: they contain a distribution of channels, due to defects in the die, inhomogeneous drying or variations in the deposition of the active coating layer.



Such wall and channel heterogeneity can have a profound impact on breakthrough profiles,[1-5] since the gas flow in each channel becomes different. The effect on process performance, however, is not well understood. Here we performed a theoretical study of the effect of a wall size distribution in a vacuum swing adsorption cycle for CO_2 capture from a model flue gas (15% CO_2).

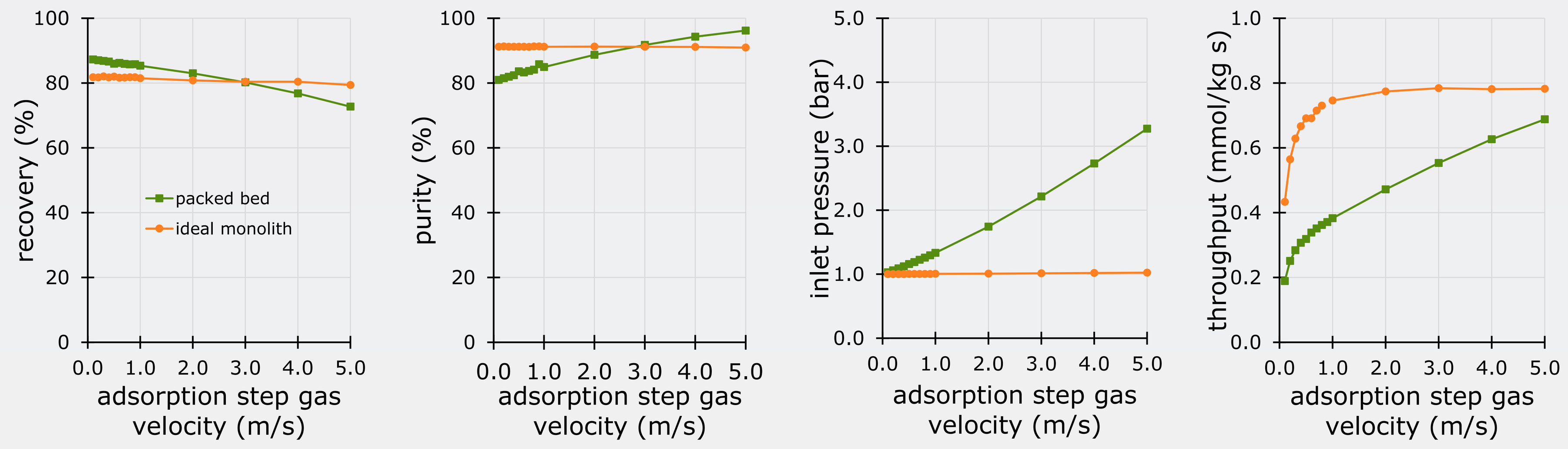
Impact On Breakthrough Profiles



15% CO_2/N_2 at different gas velocities

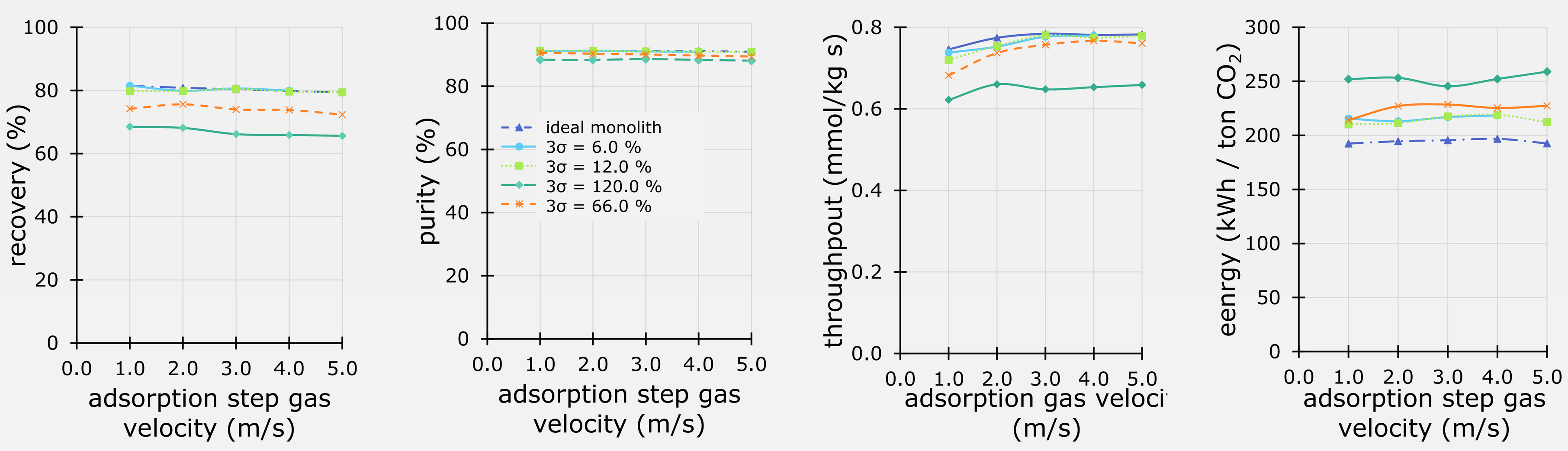
CO_2 breaks through in distinct steps, corresponding to each channel, different waves visible in temperature and concentration.
Only small effect of gas velocity in studied ranges.
HETP fairly independent of gas velocity.

Ideal Monolith vs Fixed Bed



With a monolith, less dependence on gas velocity, higher productivity.
Higher recovery & purity for a fixed bed is due to increase in pressure, increasing adsorbed amount.
Energy demand lies much lower for a monolith (190 kWh/ton) vs a fixed bed (320 kWh/ton).

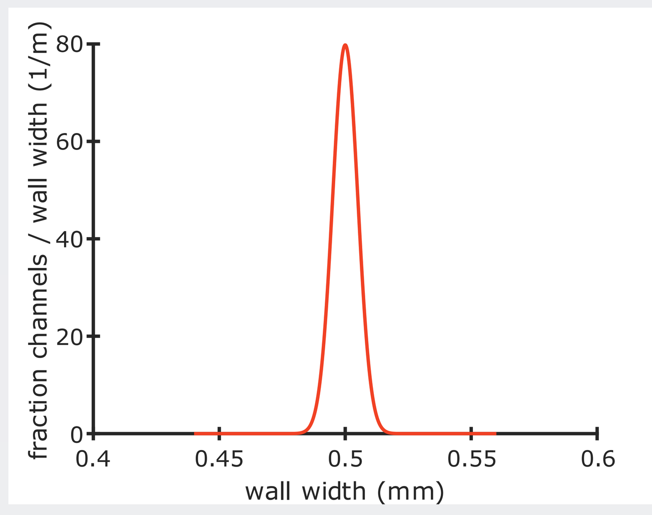
Ideal Monolith vs Imperfect System



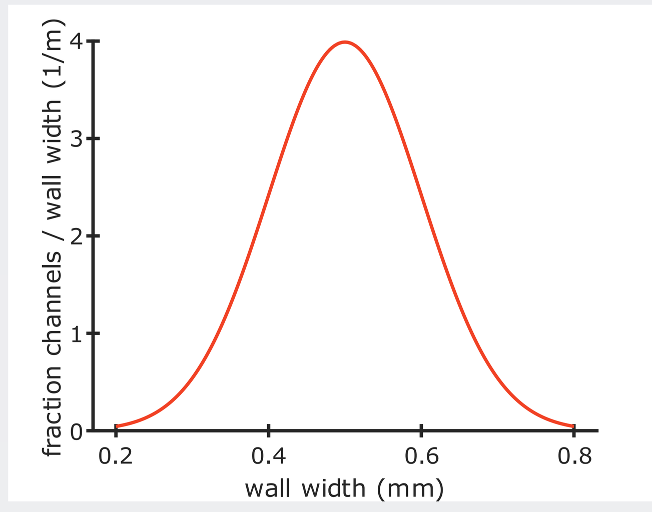
All performance parameters decrease with broader distribution.
Decrease in purity due to the larger amount of N_2 entrained in larger channels.
Decrease in recovery and productivity due to the more difficult desorption of CO_2 from the smallest walls.

Methodology
Incorporating Channel Heterogeneity

3σ = 6.0 %



3σ = 120 %



A Gaussian distribution of wall sizes was assumed, with standard deviation $3\sigma = 6\%, 12\%, 66\%, 120\%$. Each monolith was cut into 5 channels, with a wall size equal to the average of a 20% part of the distribution. Square channels were assumed.

Methodology
Mass & Energy Balances

$$\frac{\partial(y_i)}{\partial t} = -\frac{(1-\epsilon)}{\epsilon}\frac{\rho_p}{\rho_f}\frac{\partial q_i}{\partial t} - \frac{1}{\rho_f}\frac{\partial(u\rho_f y_i)}{\partial x}\bigg|_x + \frac{D_{ax}}{\rho_f}\frac{\partial \rho_f \frac{\partial y_i}{\partial x}}{\partial x}\bigg|_x - \frac{y_i}{\rho_f}\frac{\partial(\rho_f)}{\partial t}$$ component mass balance

$$\frac{\partial \rho_f}{\partial t} = -\frac{(1-\epsilon)}{\epsilon}\rho_p\sum_{i=1}^{n_c}\frac{\partial q_i}{\partial t} - \frac{\partial(\rho_f u)}{\partial x}\bigg|_x$$ total mass balance

$$\rho_f = \frac{P}{RT_g}$$ ideal gas EOS

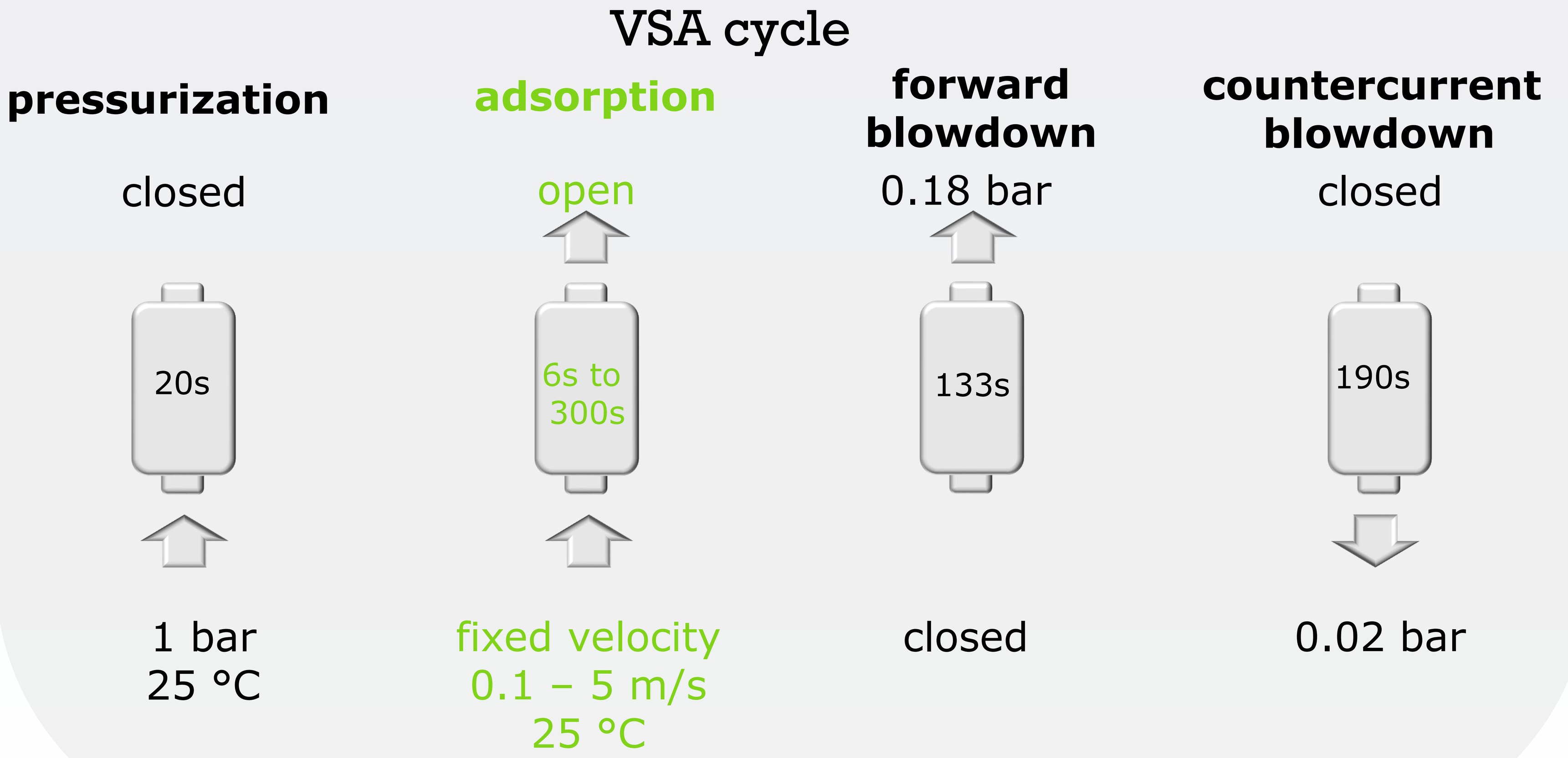
$$\frac{\partial(q_i)}{\partial t} = k_{ldf,i}(q_{eq,i} - q_i)$$ linear driving force kinetics

$$\frac{\partial T_g}{\partial t} = -\frac{1}{\rho_f C_v}\frac{\partial(u\rho_f H_g)}{\partial x} - \frac{k_{heat}a'(T_g - T_s)}{\epsilon\rho_f C_v} - \frac{E_g}{\rho_f C_v}\frac{\partial \rho_f}{\partial t}$$ gas phase energy balance

$$\frac{\partial T_s}{\partial t} = \frac{k_{heat}a'(T_g - T_s)}{C_{v,s}\rho_p(1-\epsilon)} + \frac{1}{C_{v,s}}\sum_{i=1}^{n_c}\Delta H_i\frac{\partial q_i}{\partial t}$$ solid phase energy balance

With parameters for zeolite 13X from the work of Wilkins *et al.*[6] and Brandani *et al.*[7] Solved using an *in-house* developed process simulator in MATLAB.

Methodology
VSA cycle



Cycle parameters from the work of Farooq.[8]

Conclusions

Using a monolith allows for a higher productivity (0.78 mmol/kg s) compared to a fixed bed (0.68 mmol/kg s) and a lower energy demand (190 kWh/ton CO_2 versus 320 kWh/ton CO_2). When the monolith contains a distribution of wall sizes, the process performance decreases.
The process recovery drops from 81% to 68%, the throughput drops from 0.78 mmol/kg s to 0.65 mmol/kg s and the energy demand increases from 190 kWh/ton CO_2 to 251 kWh/ton CO_2 for the widest distribution. Large defects can thus severely impact performance.

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