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How does structured adsorbent channel heterogeneity influence the efficiency of adsorptive CO₂ capture?

Benjamin Claessens,^{*} Emily Bloch, Sandrine Bourrelly

*52 Av. Escadrille Normandie Niemen
13013 Marseille
MADIREL laboratory
Aix-Marseille University / CNRS
benjamin.claessens@univ-amu.fr*

Abstract

The use of adsorbent monoliths allows to decouple the relationship between pressure drop and heat / mass transfer. This is especially important to intensify gas adsorption processes: larger gas flow rates can be treated, and shorter cycle times can be used. These aspects are crucial in carbon capture applications, where large gas flow rates must be treated, leading to short contact times in the adsorption column.

Commercial adsorbent monoliths consist of parallel-channel structures, which are extruded through a die. This extrusion process inevitably leads to defects, variations in channel size of 10-20 % are reported in literature.[1], [2] Experimental studies clearly show an effect on dynamic breakthrough curves.[1], [3] The impact of these imperfections on cyclic process performance, however, is not well understood.

In this work, process simulations were performed to link a distribution in channel sizes to process performance variables in a cyclic vacuum swing adsorption (VSA) process for carbon capture. In a first step an ideal adsorbent monolith was compared with a fixed bed. Using a monolith allows for a higher productivity (0.78 mmol/kgs) compared to a fixed bed (0.68 mmol/kgs) and a lower energy demand (190 kWh /tonne CO₂ versus 320 kWh/tonne CO₂). Furthermore, process performance for monoliths is much less dependent on interstitial gas velocity, allowing a more stable operation with a varying feed gas flow rate.

In a next step, simulations were performed taking into account a Gaussian distribution of monolith channel sizes. Simulations were performed with 3σ ranging from 1.2 % to a large variation of 120 %. Starting from a variation of 12 %, a real impact on process performance can be expected. In the worst-case scenario, with a very large channel distribution, recovery drops from 81 % to 68 %, throughput from 0.78 mmol/kg s to 0.65 mmol/kg s and energy demand from 190 kWh / tonne CO₂ to 320 kWh / tonne CO₂ (Figure 1). Interestingly, even for very large wall size distributions, the performance of monoliths remains better than a fixed bed of spherical particles.

In conclusion, this work shows the promising properties of adsorbent monoliths for CO₂ capture and offers guidelines

for practical defect management in structured adsorbents.[4]

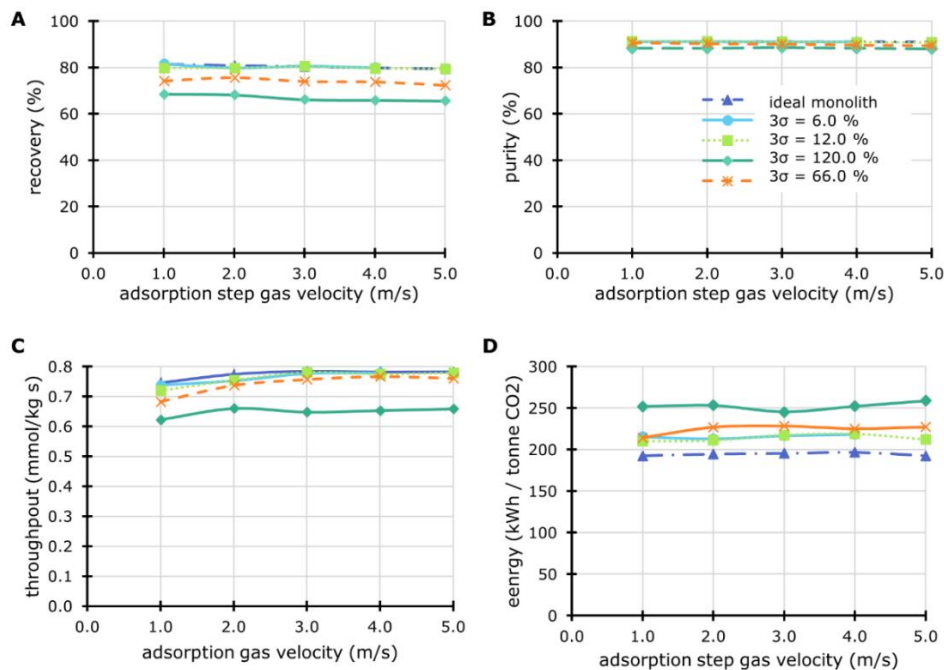


Figure 1. Comparison between the recovery (A), purity (B), throughput (C) and energy demand (D) of a monolith without wall size distribution and a monolith containing a wall size distribution.

References

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