Session 3B – DAC
16/09/2025

Techno-Economic Analysis of Packed Bed and Monolithic Sorbent for Direct Air Capture

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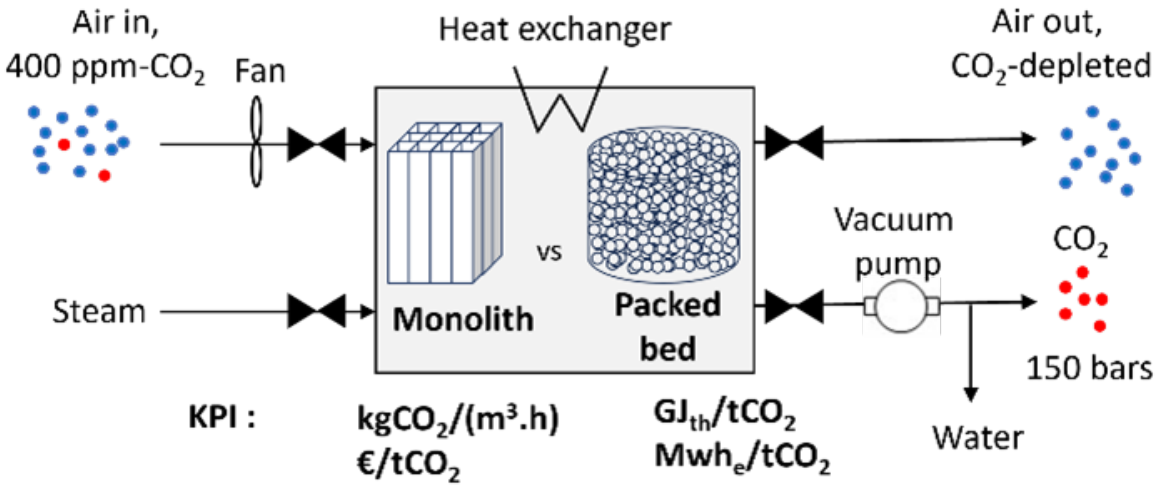
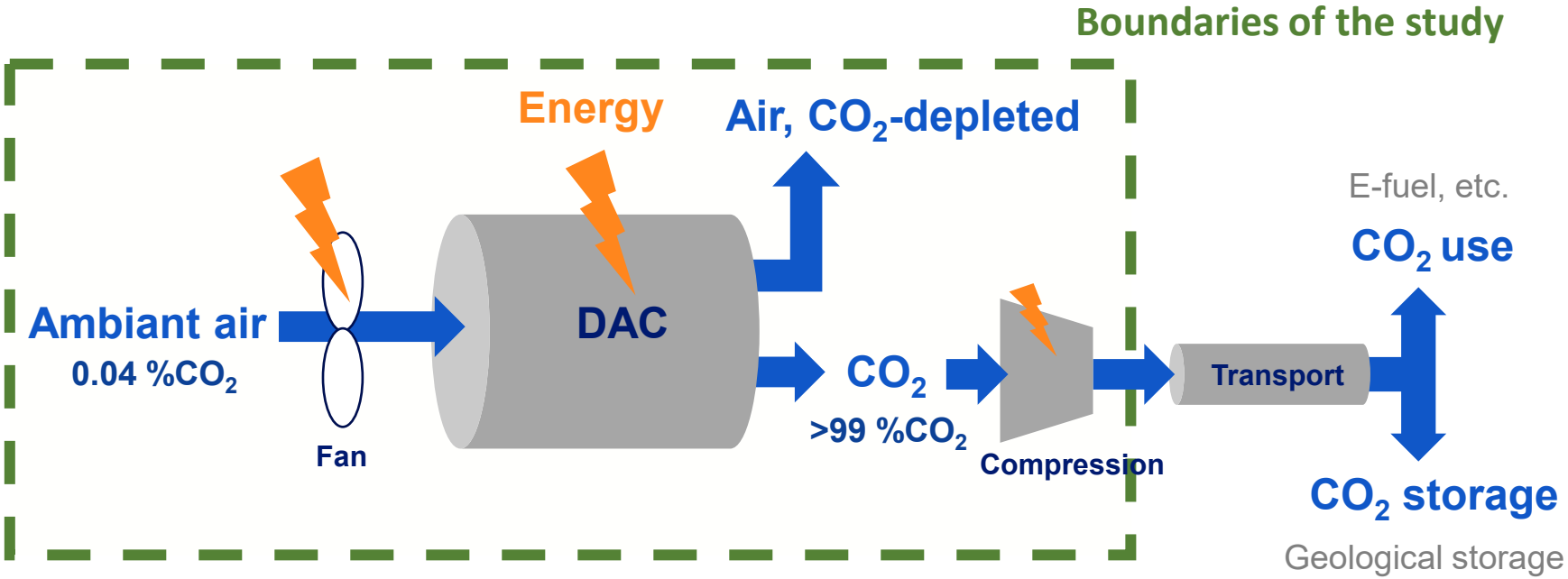


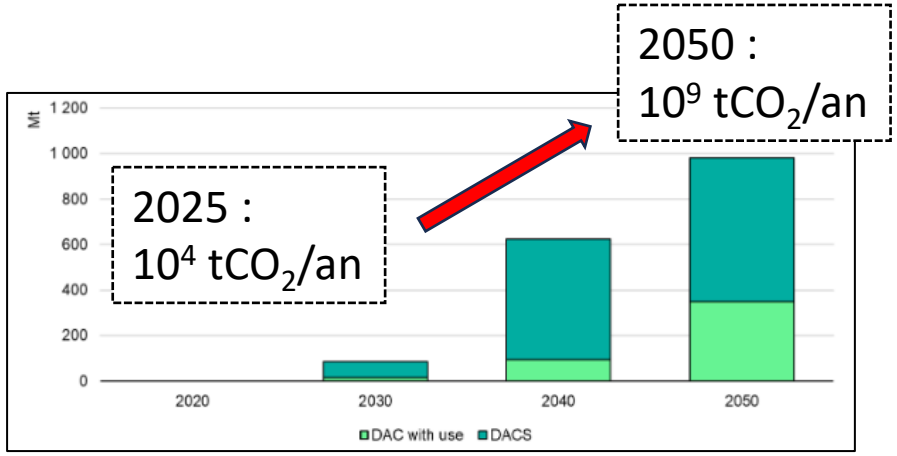
1. Context



Why ?

- ❑ For residual emissions : 1-20 Gt-CO_2/year
- ❑ Hard-to-abate, expensive-to-abate
- ❑ Agriculture, maritime and air transport

Need of a **massive deployment** if acceptable in terms of emission factor, impact, sustainability and cost



Net Zero Scenario of the International Energy Agency (IEA).

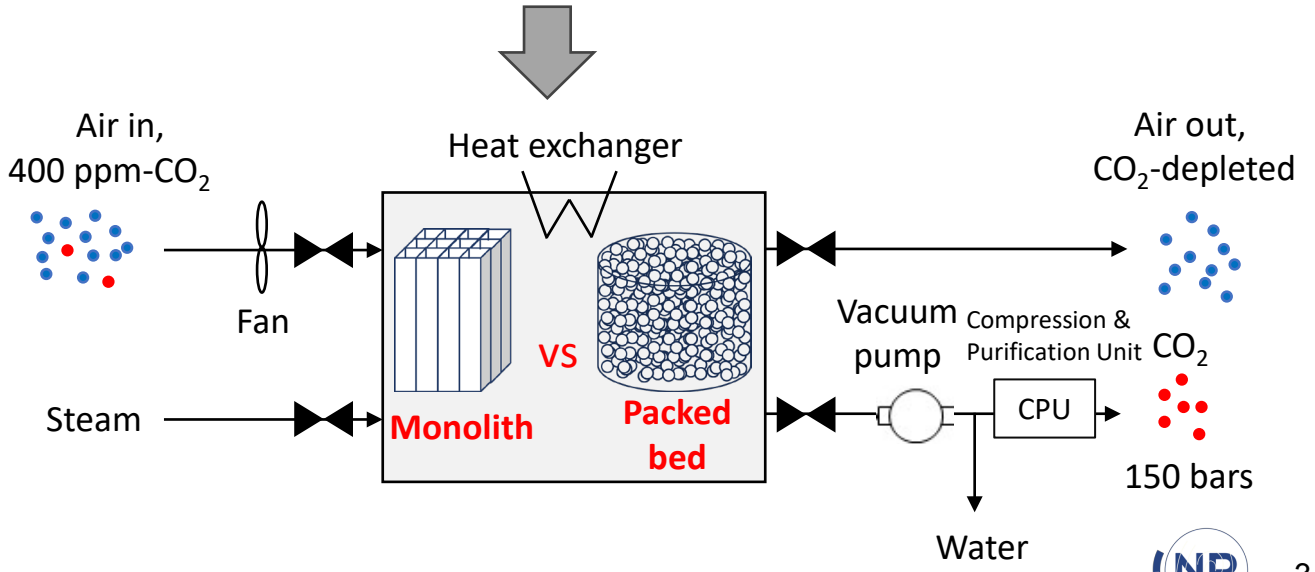
2. Objectives of the work

Observations :

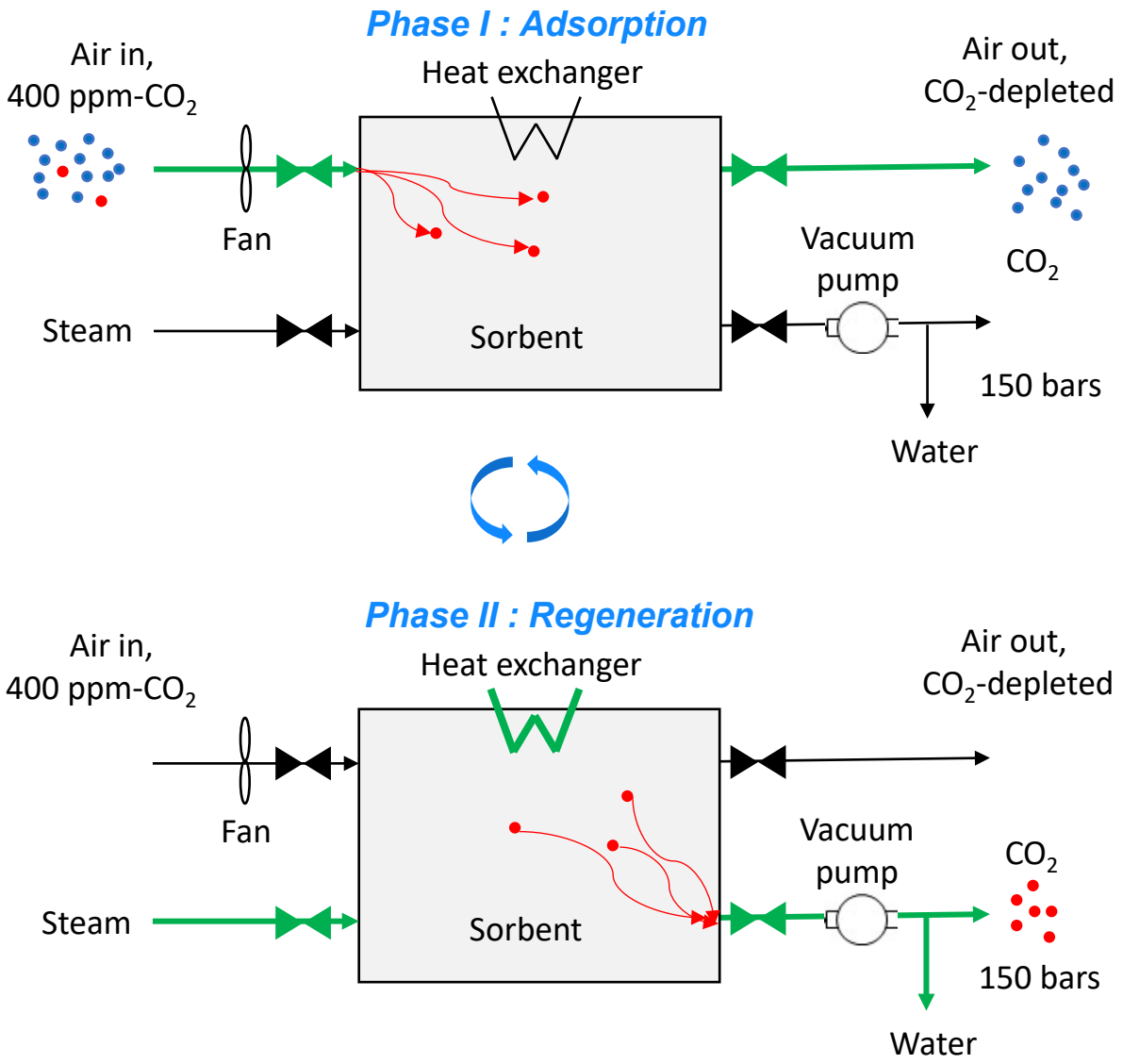
- ❑ Adsorption-based DAC processes are the most widely deployed
- ❑ Most of works are based on dry ambiant condition (without air humidity)
- ❑ Most of works are based on sorbent in pellet shape (packed bed), where the pressure drop for the air to pass trough the bed can be a bottleneck
- ❑ Simulation softwares allow studies to guide future process development

Objectives :

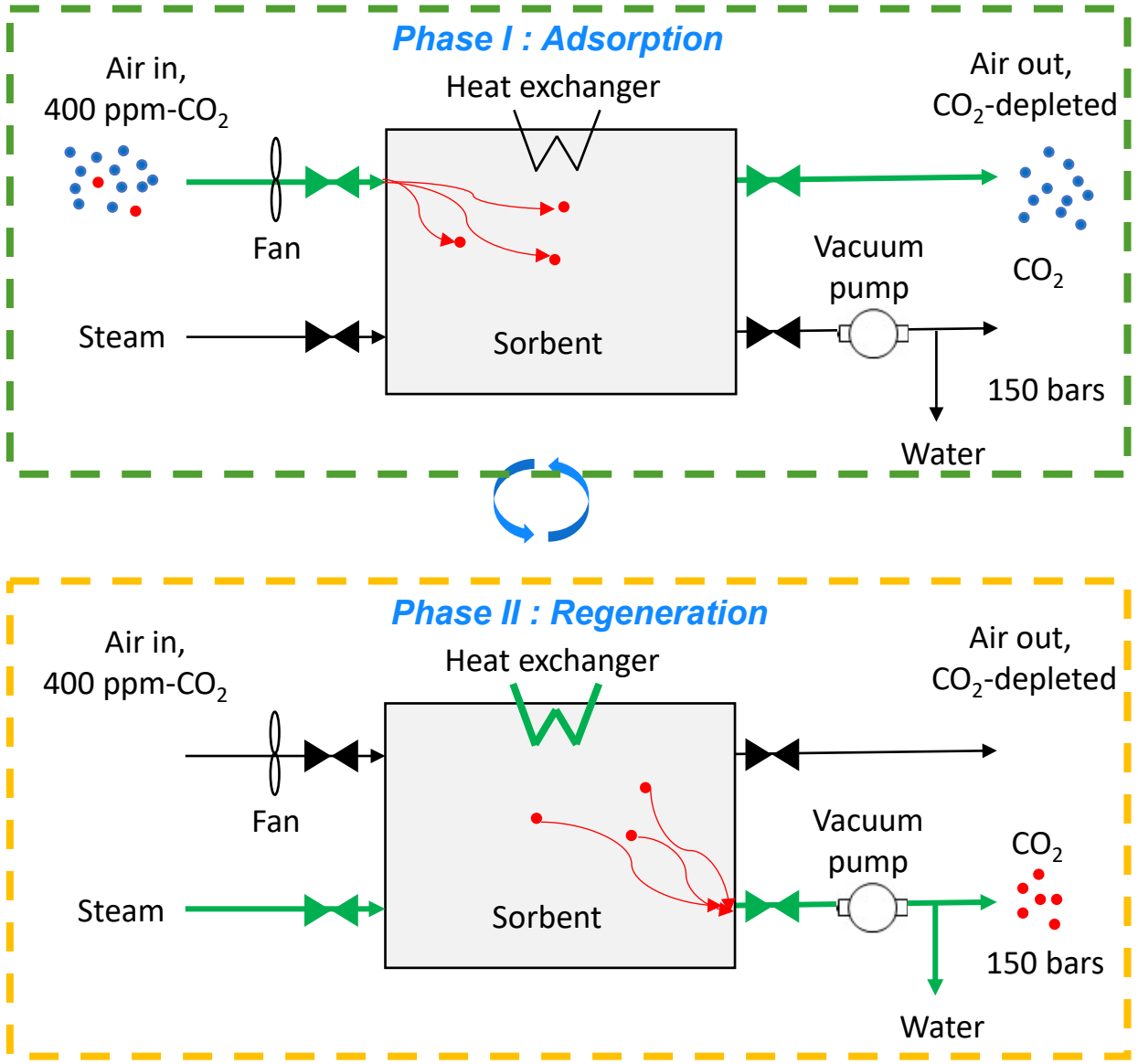
- ❑ Simulate an adsorption-based DAC process with a commercially available sorbent which isotherms for DAC conditions (low CO_2 pressure, humidity, temperature) that are available in literature
- ❑ Compared packed bed configuration to a structured sorbent shaping



3. S-VTSA (Steam-assisted Vacuum Thermal Swing Adsorption) process overview



4. S-VTSA (Steam-assisted Vacuum Thermal Swing Adsorption) cycle



	Step	Duration	Temperature (°C)	Pressure (bar)
1	Adsorption	Until $C_{CO_2}^{out}/C_{CO_2}^{in} = 50\%$	15	1.013
2	Evacuation	Until $P_{bed} = 0.35$ bar	15	0.3
3	Pre-heating	500 s	70	0.3
4	Pre-purge	50 s	70	0.3
5	**Desorption**	**Until $q_{CO_2} = 0.1$ molCO_2/kg**	**100**	**0.3**
6	Cooling	Until $T_{bed} = 70\ ^\circ$ C	70	0.3
7	Repressurization	Until $P_{bed} = 1.013$ bar	15	1.013

Oxydation

5. Sorbent material choice : Lewatit VP OC 1065

Lewatit VP OC 1065 (amine sorbent)

- ✓ Commercially available
- ✓ High CO_2 uptake in ambiant conditions (> 1 mol/kg)
- ✓ Available isotherms data in DAC conditions (low CO_2 pressure, humidity, temperature)
- ✓ Physico-chemical properties available (density, diameter, heat of adsorption, …)

Isotherms model:

- ❑ H_2O significantly enhances CO_2 adsorption.
  Co-adsorption CO_2 isotherm model:

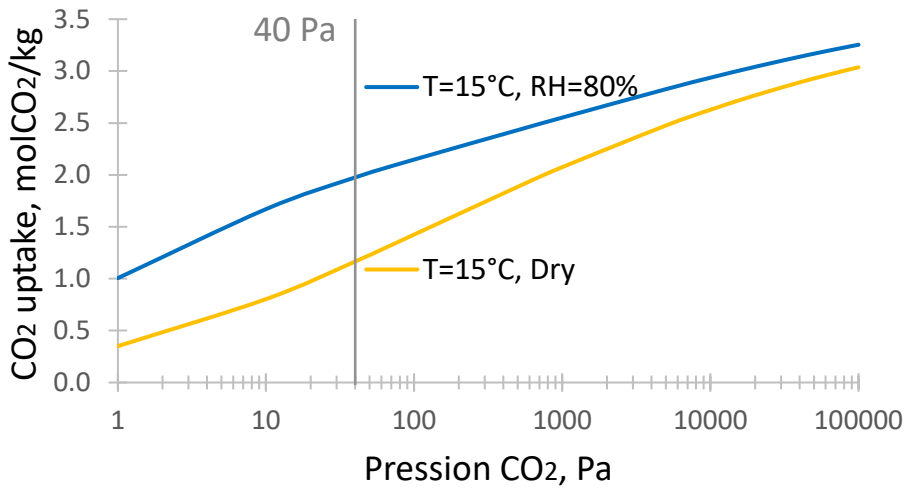
$$q_{CO2}^{wet}\left(q_{CO2}^{dry}, RH\right) = \left(b.e^{c.q_{CO2}^{dry}(T,p_{CO2})}.RH + 1\right).q_{CO2}^{dry}(T, p_{CO2})$$

with

RH relative humidity

$q_{CO2}^{dry}(T, p_{CO2})$ calculated from Toth isotherm in dry conditions

b and c fitted from data



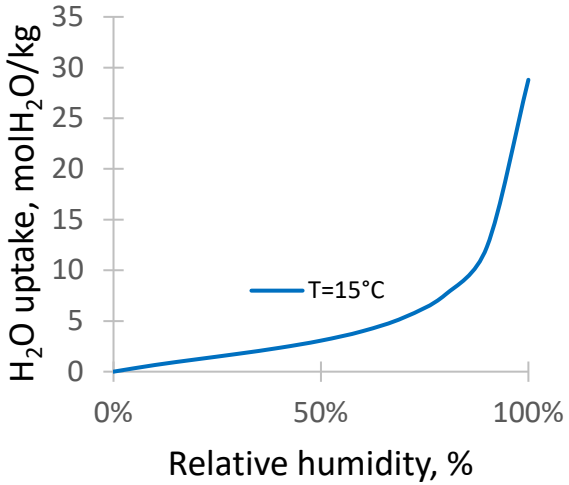
5. Sorbent material choice : Lewatit VP OC 1065

Isotherms model (continued):

- H_2O isotherm model: Guggenheim-Anderson-de-Boer (GAB) isotherm.

  → Isotherm model parameters are fitted from experimental data.

- N_2, O_2 and Ar adsorption neglected.



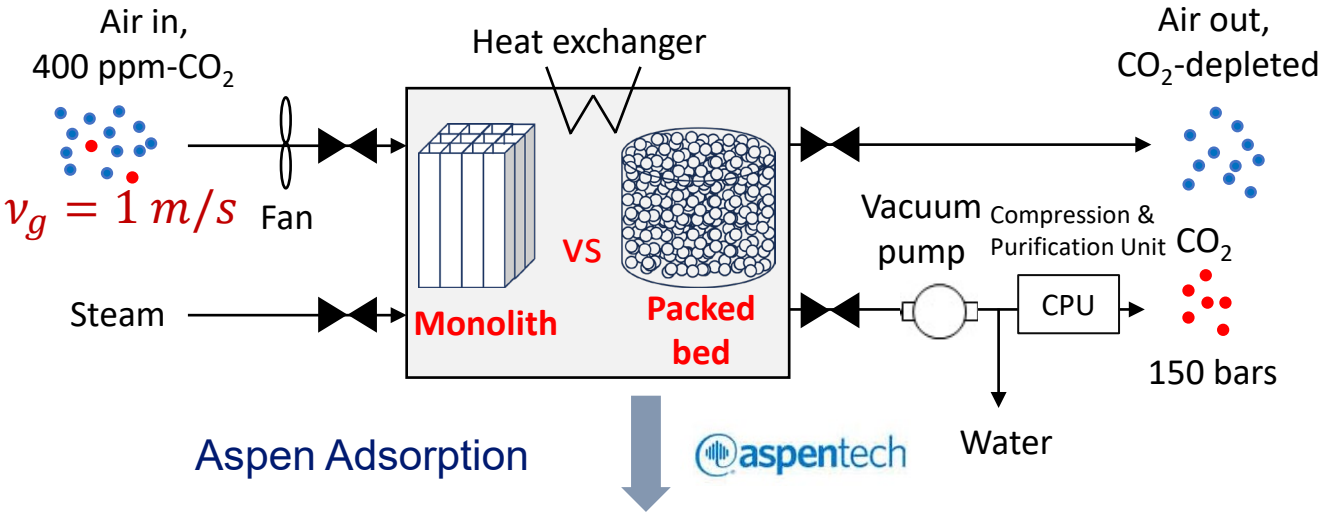
Heat of adsorption:

- $\Delta H_{CO2} = 73$ kJ/mol
- $\Delta H_{H2O} = 44$ kJ/mol

  → Clausius-Clapeyron fit from isotherms

$$\frac{dln(p_k)}{d\frac{1}{T}} = -\frac{\Delta H_k}{R}$$

7. Comparison between packed bed and monolith designs

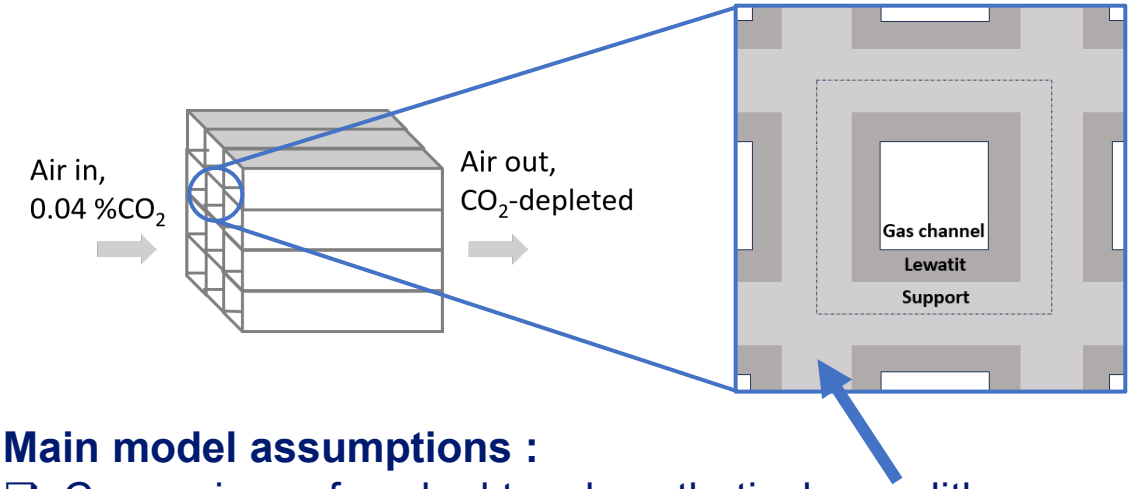


Aspen Adsorption → aspentech

Energy & Mass balance	Heat requirement (GJ_{th}/tCO_2) Elect. requirement (Mwh_e/tCO_2) Productivity ($kgCO_2/(h.m^3)$) Pressure drop (Pa) Water production ($kgH_2O/kgCO_2$)

Economic evaluation → Aspen Process Economic Analyzer

Cost of capture	$€_{2022}/tCO_2$



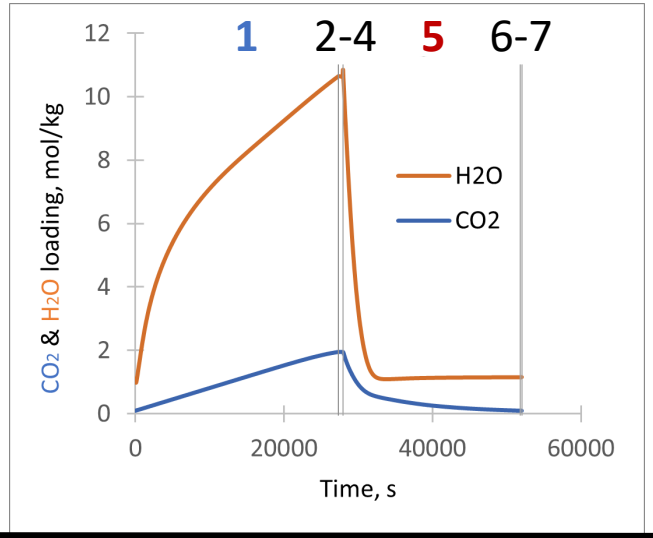
Main model assumptions :

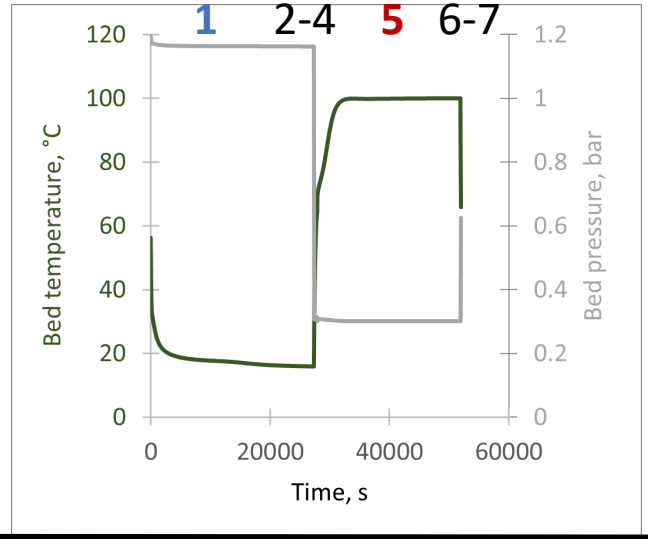
- ❑ Comparison of packed to a hypothetical monolith
  - ❑ Packed bed: filled with spherical particle
  - ❑ Monolith: sorbent coated on stainless steel support
- ❑ Small module dimension: H = 0.5 m ; D = 2 m
- ❑ 1-D model with axial dispersion and adiabatic bed, using linear driving force model for mass transfer
- ❑ Pressure drop:
  - ❑ Packed bed: Ergun equation
  - ❑ Monolith: Hagen-Poiseuille equation

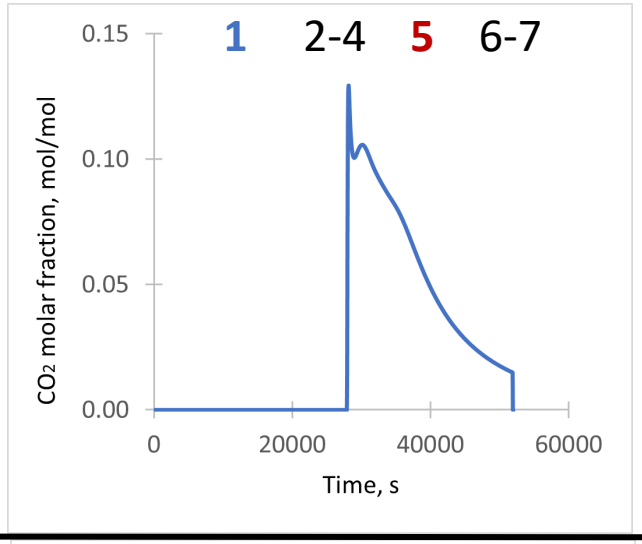
8. Results : Cycle behaviour

1 : Adsorption
2-4 : Intermediate steps
5 : Desorption
6-7 : Cooling-repressurization

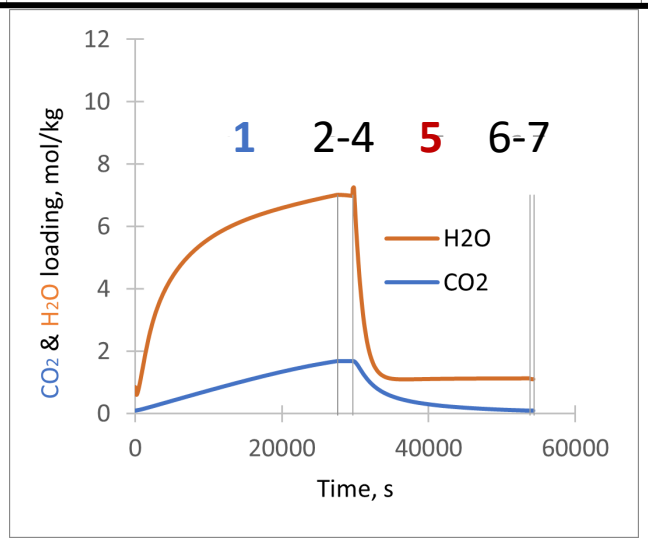
Packed bed
$v_g = 1\ m/s$

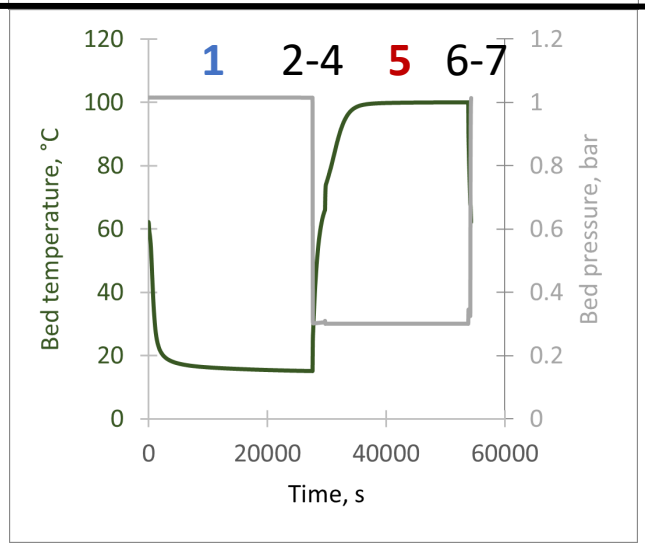


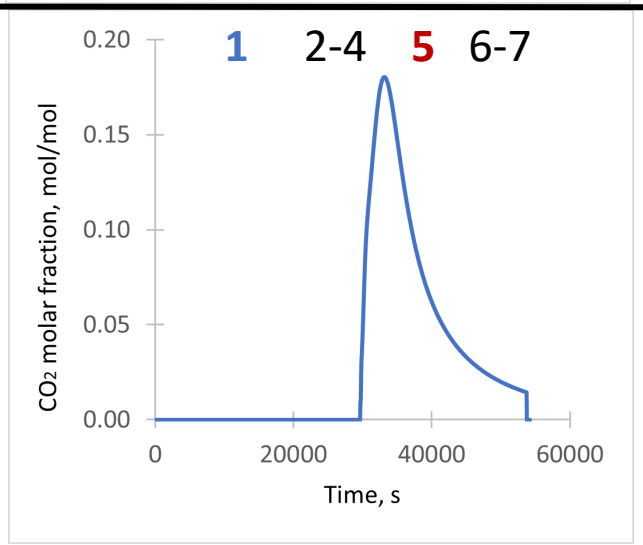




Monolith
$v_g = 1\ m/s$

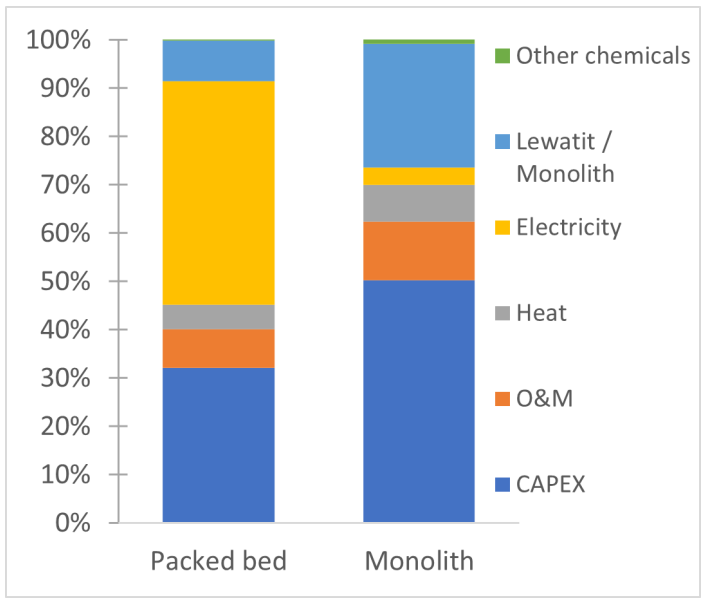




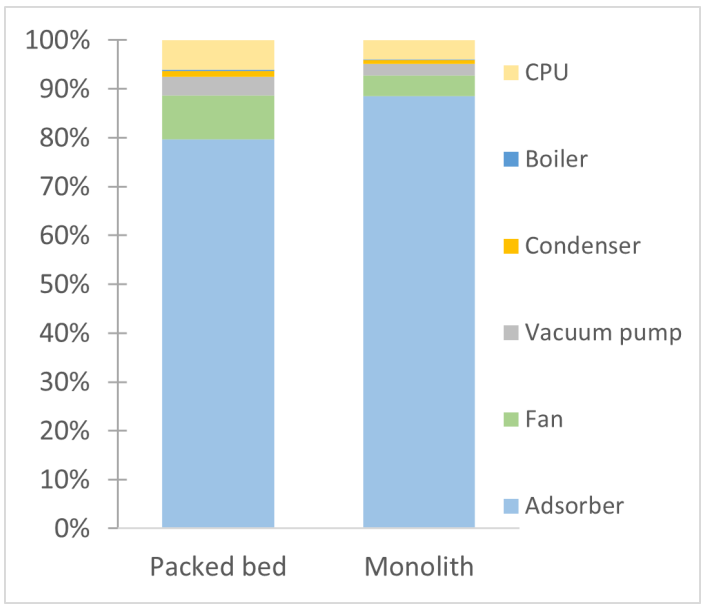


8. Results : Main results (reference case)

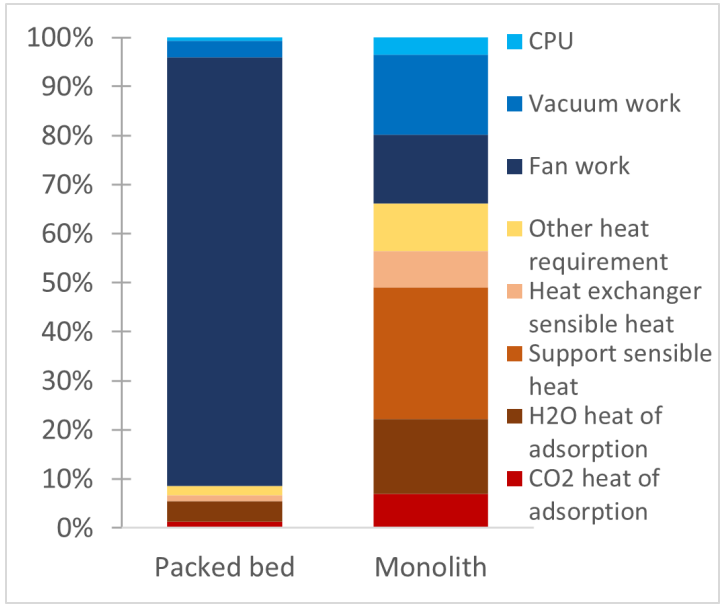
	Design	Packed bed	Monolith
CO_2 productivity	$kgCO_2/(h.m^3)$	2.39	1.17
H_2O production	tH_2O/tCO_2	2.10	1.50
Pressure drop	Pa	26.5×10^3	0.66×10^3
Total heat. energy	GJ/tCO_2	10.7	15.9
Total elec. energy	MWh/tCO_2	13.8	0.98
Total primary energy	GJ/tCO_2	124.9	24.0
Cost of capture	$€/tCO_2$	**2102**	**2090**



Cost distribution



CAPEX distribution



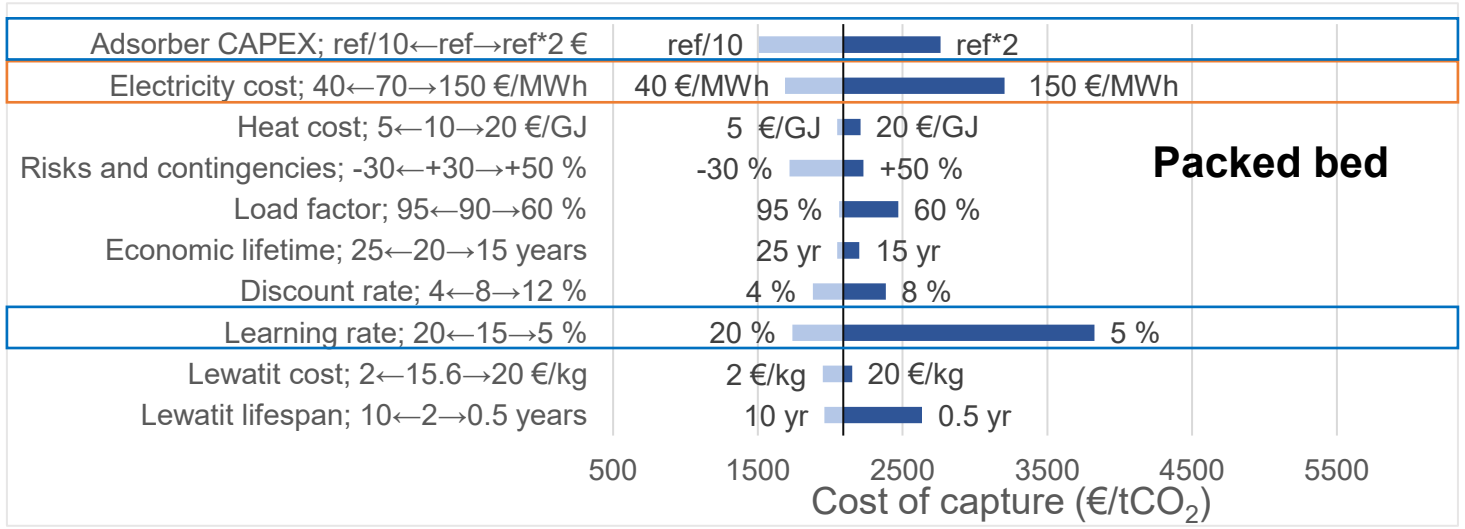
Primary energy distribution

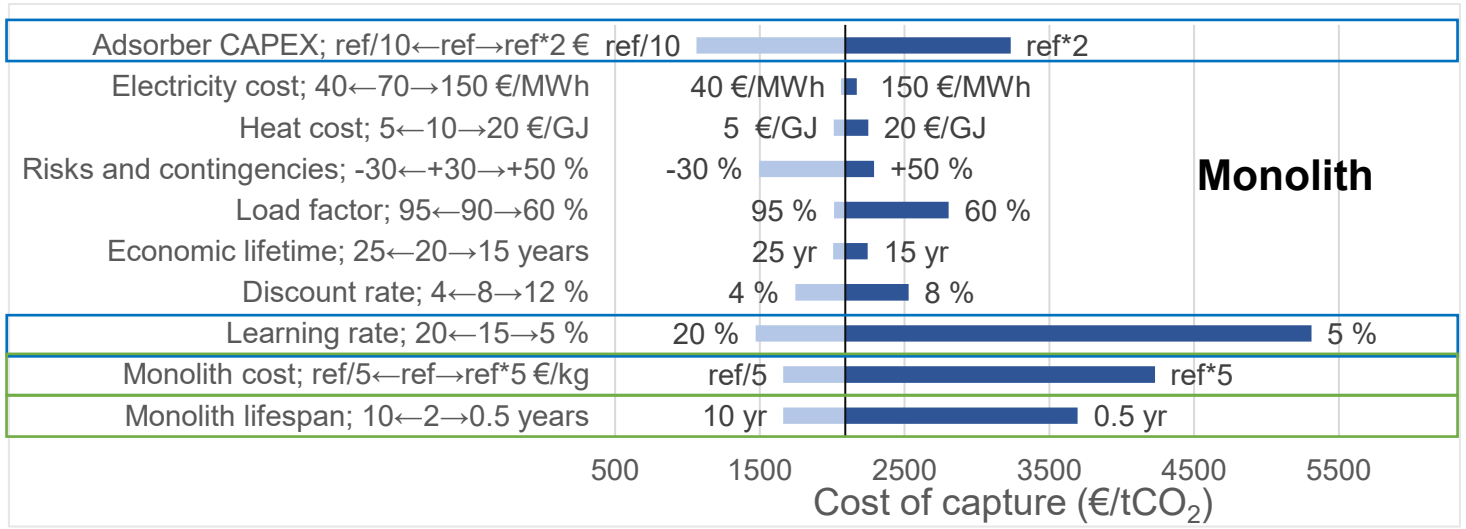
(Primary energy = heat. energy + elec. energy * 2.3)

Key message:

- High pressure drop of packed bed → high elect. requirement
- High cost of capture (high adsorber cost)
- Packed bed: OPEX intensive
- Monolith: CAPEX intensive

9. Results : Economic sensibility





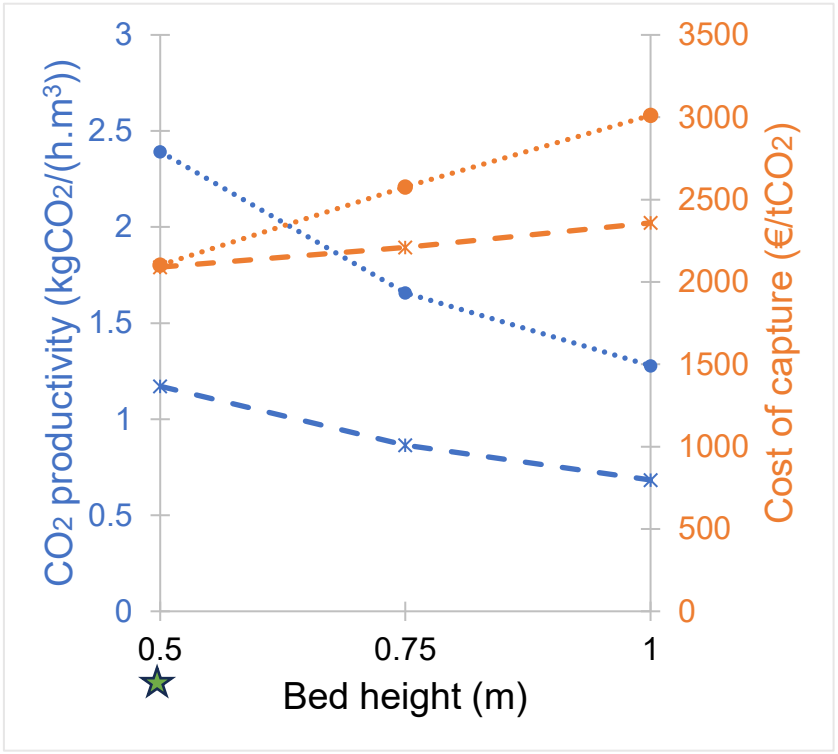
- Cost of capture influenced by the economic and technical assumptions.
- Process data from simulations = inputs for Aspen Process Economic Analyzer (APEA).
- Estimation for feasibility studies: -30% to +50% accuracy.

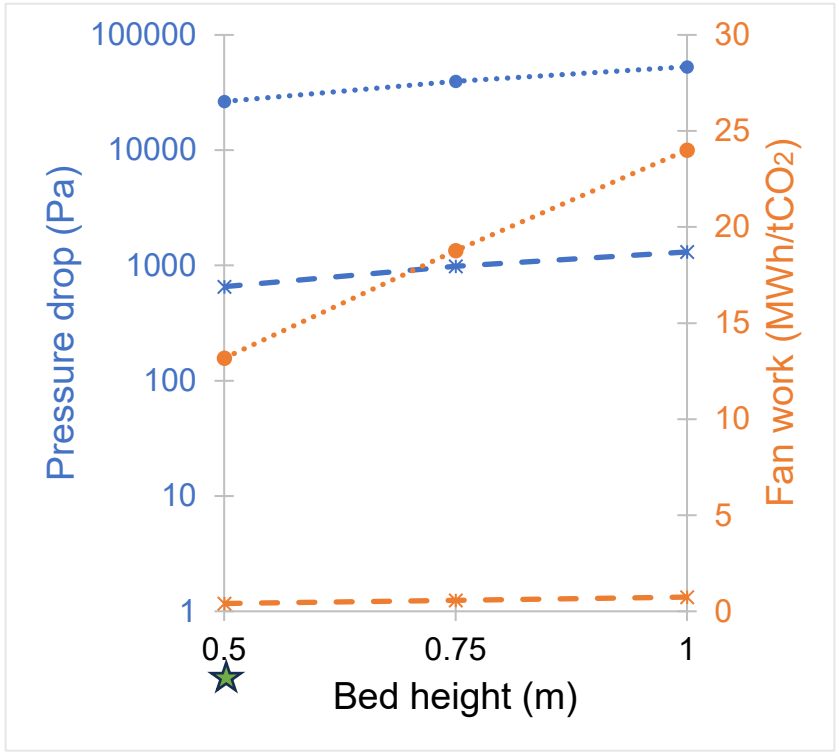
→ Economic sensitivity

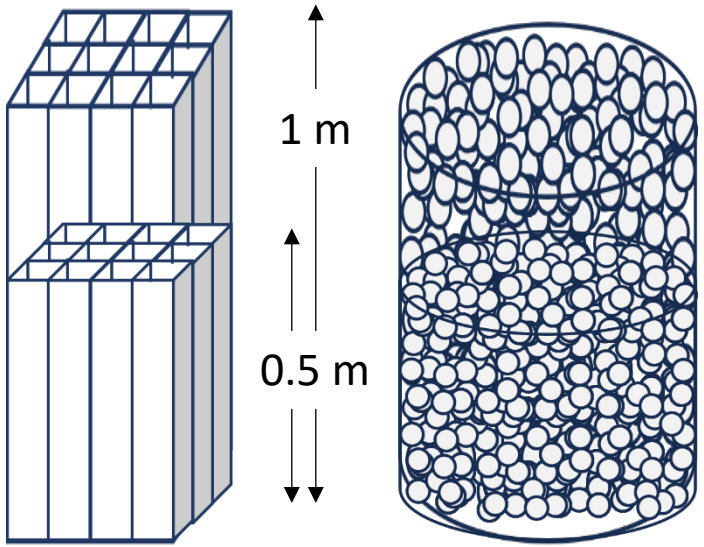
Key message:

- High influence of adsorber cost and learning rate (high CAPEX & modularity)
- Packed bed cost influenced by electricity cost (due to high requirement)
- Monolith cost more influenced by CAPEX parameter and by monolith cost and lifetime

10. Results : Sensitivity on bed height







★Reference case: $H = 0.5$ m

Packed bed

Monolith

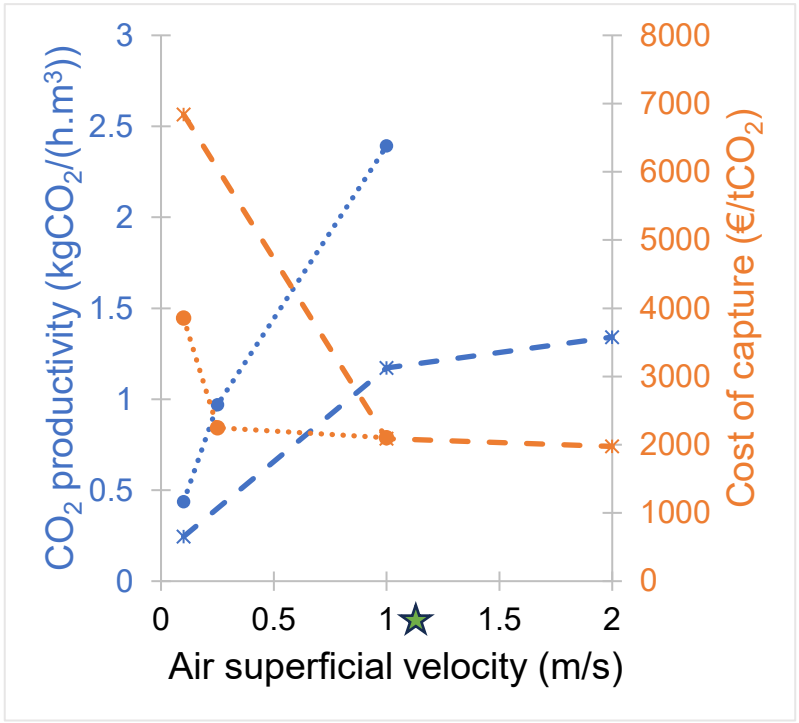
Key message:

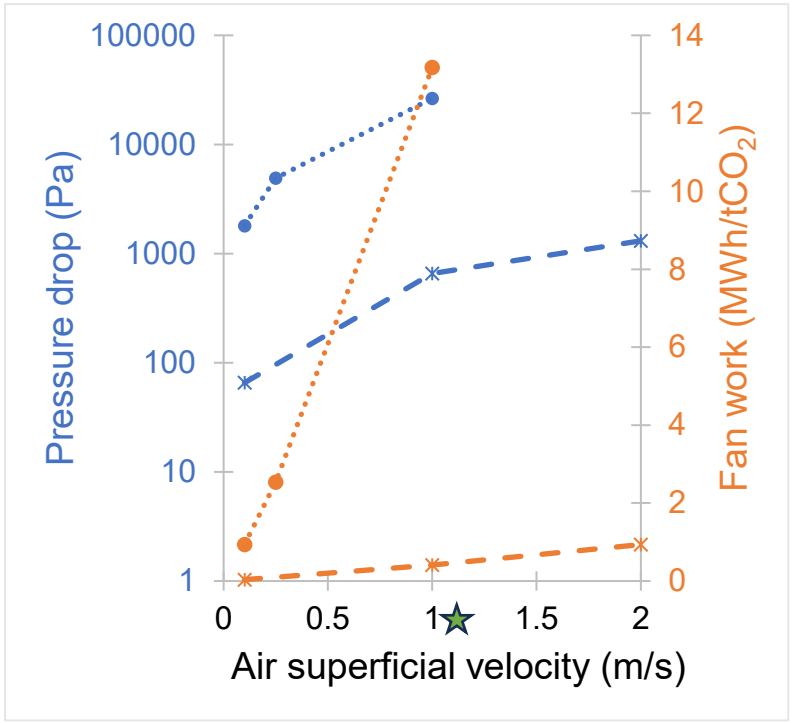
Smaller height are favorable in term of :

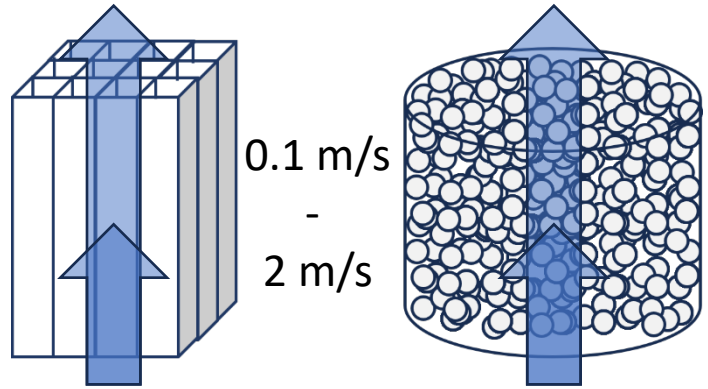
- Productivity
- Energy requirement (pressure drop)
- Cost

→ Small height can be limited by technical feasibility

11. Results : Sensitivity on air velocity during adsorption phase







★ Reference case: $v_g = 1$ m/s

Pressure drop:

- ❑ Packed bed: Ergun equation

$$\frac{\partial P}{\partial z} = -\left(\frac{150(1-\varepsilon_i)^2}{(2r_p)^2 {\varepsilon_i}^3}\,\mu\, v_g + 1.75\,\rho_g \frac{(1-\varepsilon_i)}{2r_p {\varepsilon_i}^3}\, {v_g}^2\right)$$

- ❑ Monolith: Hagen-Poiseuille equation

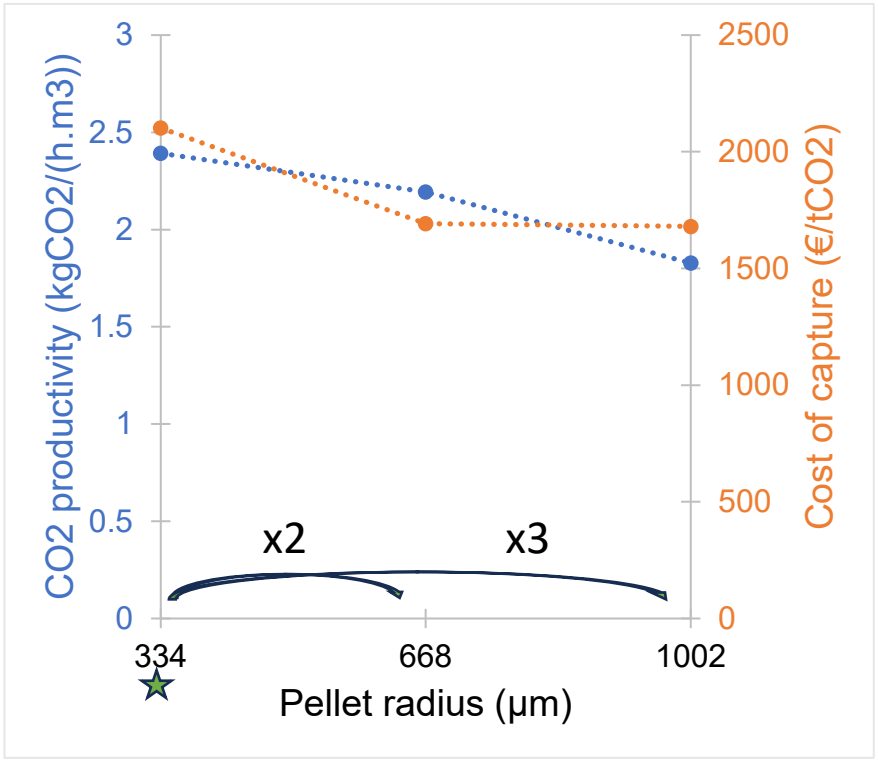
$$\frac{\partial P}{\partial z} = -\frac{28.4\,\mu}{\varepsilon_i\, a^2} v_g$$

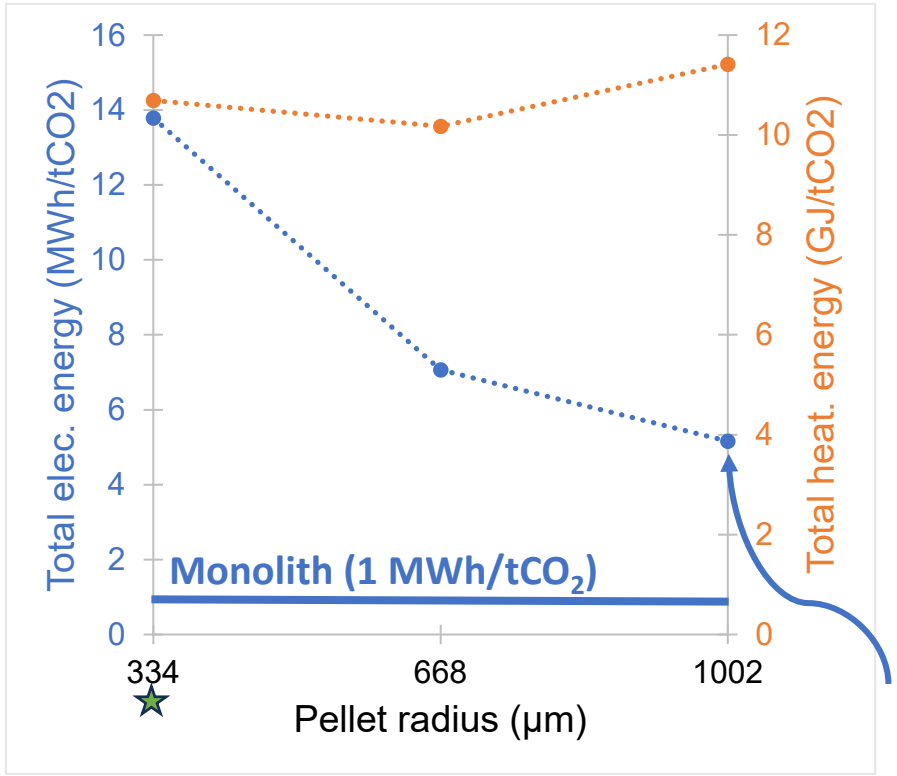
Key message:

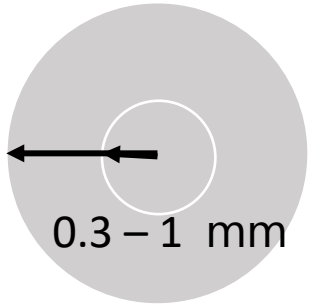
Higher air velocity:

- Productivity improved → until residence time becomes to short vs mass transfer from gas bulk to sorbent.
- Too high pressure drop for packed bed

12. Results : Sensitivity on pellet radius for packed bed







Sensitivity on pellet radius for packed bed

★Reference case: $r = 0.3$ mm

Elec. energy remains high (> 5 MWh/tCO_2) compared to monolith (1 MWh/tCO_2)

Key message:
Larger pellet radius:
- Productivity & cost reduces
- Pressure drop reduces → fan work reduces (remains high)

13. Conclusion and perspectives

Conclusion :

- ❑ Very high pressure of packed bed → High elec. energy (> 5 MWh/tCO_2) compared to monolith (ca. 1 MWh/tCO_2)
- ❑ Packed bed more OPEX intensive vs. Monolith more CAPEX intensive
- ❑ High cost of capture (~2000 €/tCO_2) with high adsorber cost → expected to decrease with further development and economies of scale

Perspectives :

- ❑ Sensitivity analysis on process parameter for monolith ★
- ❑ Monolithic design development
- ❑ Cheaper adsorber module
- ❑ Experimental to reinfor simulation results & economic estimation

★ P. de Joannis, C. Castel, M. Kanniche, E. Favre, and O. Authier, "**Techno-Economic Analysis of Packed Bed and Structured Adsorbent for Direct Air Capture**," *Carbon Capture Science & Technology*, p. 100518, Sep. 2025, doi: 10.1016/j.ccst.2025.100518.

Thank you

Any questions ?

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Assumptions

2.3.1. Main assumptions of packed bed model

The packed bed model makes the main following assumptions:

1. Low *et al.* have shown from experimental adsorption isotherms of N_2, O_2 and Ar on Lewatit that their loadings are very small (<< 0.1 mol/kg) compared to CO_2 and H_2O loading in ambient air conditions (Low et al., 2023). So N_2, O_2 and Ar adsorption loadings are neglected in the study. CO_2 and H_2O are the only adsorbed substances.
2. 1-D spatial dimension model with axial dispersion is considered.
3. The Aspentech's PENG-ROB method is used, with Peng-Robinson equation of state for compression to supercritical CO_2.
4. The packed bed of spherical particles is non-isothermal and is considered adiabatic with radial uniform (i.e. 1-D) heating of the gas phase.
5. The gas mass transfer of component to adsorbent is expressed using the linear driving force (LDF) model, considering reaction kinetics instantaneous.

Equations

2.3.2. Mass balance

The overall mass balance for a multi-component gas phase considers axial dispersion, convection, accumulation, and gas-solid mass transfer. Each substance k in the gas phase is governed by the equation below:

$$-\varepsilon_i E_{z,k} \frac{\partial^2 c_k}{\partial z^2} + \frac{\partial (v_g c_k)}{\partial z} + \varepsilon_B \frac{\partial c_k}{\partial t} + J_k = 0 \qquad (2)$$

Where ε_i is the interparticle voidage, $E_{z,k}$ is the axial dispersion coefficient of component k (m^2/s), c_k is the concentration of component k (mol/m^3), v_g is the superficial gas bed velocity (m/s), and $\varepsilon_B = \varepsilon_i + (1-\varepsilon_i)\varepsilon_p$ is the total bed voidage with ε_p the intraparticle voidage.

The axial dispersion is given by the relation below (Kast, 1981):

$$E_{z,k} = 0.73\, D_{m,k} + \frac{v_g r_p}{\varepsilon_i \left(1 + 9.49 \frac{\varepsilon_i D_{m,k}}{2 v_g r_p}\right)} \qquad (3)$$

Where $D_{m,k}$ is the molecular diffusion coefficient of k in the gas (m^2/s), and r_p is the particle radius (m).

The rate of adsorption of k ($mol/(m^3.s)$) on adsorbent is expressed using LDF model:

$$J_k = -\rho_B MTC_k (q_k^* - q_k) \qquad (4)$$

Where q_k^* is the equilibrium adsorbed quantity (mol/kg) (i.e. given by the isotherm), q_k is the loading of k on the adsorbent (mol/kg), ρ_B is the sorbent bulk density (kg/m^3), and MTC_k is the lumped mass transfer coefficient (1/s) (Rezaei and Webley, 2009):

$$MTC_k = \frac{\varepsilon_i K_k}{\rho_B \frac{dq_k^*}{dP_k} RTZ} \qquad (5)$$

Where $\frac{dq_k^*}{dP_k}$ (mol/(kg.Pa)) is numerically derived from the isotherms, R is the universal gas constant (J/(mol.K)), Z is the compressibility factor, and the K_k (1/s) is given as a lumped term comprising the external film resistance term and the pore diffusion term:

$$\frac{1}{K_k} = \frac{r_p}{3(1-\varepsilon_i)\, k_{f,k}} + \frac{r_p^2}{15(1-\varepsilon_i)\, K_{p,k}} \qquad (6)$$

The film transfer coefficient $k_{f,k}$ (m/s) is obtained from the Sherwood Sh_k number:

$$k_{f,k} = Sh_k \frac{D_{m,k}}{2r_p} \qquad (7)$$

Where $Sh_k = 2.0 + 1.1\, Sc_k^{1/3} Re^{0.6}$, for a Reynolds number $Re = \frac{v_g 2 r_p \rho_g}{\mu}$ in the range of [3:10000] (Wakao and Funazkri, 1978) with $Sc_k = \frac{\mu}{D_{m,k}\rho_g}$ the Schmidt number.

The pore diffusion coefficient $K_{p,k}$ (m^2/s) is obtained from:

$$\frac{1}{K_{p,k}} = \frac{\tau}{\varepsilon_p}\left(\frac{1}{D_{n,k}} + \frac{1}{D_{m,k}/p}\right) \qquad (8)$$

Where τ is the tortuosity, p is a parameter used to consider the apparent diffusion of the CO_2 into the pore of the sorbent (estimated from breakthrough curves in wet conditions, see **Section 2** of the Supplementary Materials for model validation), and $D_{n,k} = 97 r_{pore} \left(\frac{T}{M_k}\right)^{0.5}$ is the Knudsen coefficient (m^2/s) , with r_{pore} the pore radius (m) and M_k the molar mass (kg/mol).

2.3.3. Gas energy balance

The energy balance of the gas phase includes gas conductivity, convection, accumulation of heat, compression, and heat transfers with the solid (i.e. sorbent) and with the intern heating:

$$-\varepsilon_i k_g \frac{\partial^2 T_g}{\partial z^2} + C_{v,g} v_g \rho_g \frac{\partial T_g}{\partial z} + \varepsilon_B C_{v,g} \rho_g \frac{\partial T_g}{\partial t} + P \frac{\partial v_g}{\partial z} + HTC a_p (T_g - T_s) + a_H Q_H = 0 \qquad (9)$$

Where $C_{v,g}$ is the specific capacity of gas (J/(K.kg)), P the pressure (Pa), and T_g and T_s the gas and solid temperature (K), respectively.

The gas thermal conductivity k_g (W/(m.K)) is based on axial dispersion (Froment et al., 2011):

$$k_g = C_{v,g} \rho_g \sum_k (E_{z,k} y_k) \qquad (10)$$

Equations

With y_k the molar fraction of substance k in gas.

The heat transfer coefficient HTC (W/m² K) between gas and solid is calculated from (Froment et al., 2011):

$$HTC = jC_{p,g}v_g\rho_g Pr^{-\frac{2}{3}} \quad (11)$$

With Pr the Prandtl number and the j-factor are:

$$Pr = \frac{\mu C_{p,g}}{k_g} \quad (12)$$

$$j = 1.66Re^{-0.51} \; if \; Re < 190 \quad (13)$$

$$j = 0.983Re^{-0.41} \; otherwise \quad (14)$$

The specific surface area of the bed a_p (m²/m³) is calculated as:

$$a_p = (1-\varepsilon_i)\frac{3}{r_p} \quad (15)$$

An internal heat exchanger is used to heat/cool the bed according to:

$$a_H Q_H = a_H U_H (T_g - T_H) \quad (16)$$

With a_H the heat exchanger specific surface (m²/m³), U_H the heat exchanger coefficient (W/m²) and T_H the heating/cooling media temperature (K).

Pressure drop across the packed bed considers Ergun equation for ideal sphere:

$$\frac{\partial P}{\partial z} = -\left(\frac{150(1-\varepsilon_i)^2}{(2r_p)^2\varepsilon_i^3}\mu v_g + 1.75\,\rho_g\frac{(1-\varepsilon_i)}{2r_p\varepsilon_i^3}{v_g}^2\right) \quad (17)$$

2.3.4. Solid energy balance

The energy balance of the solid phase includes thermal conduction, accumulation of heat of sorbent and heat of adsorbed phase, heat of adsorption, and heat transfer with the gas:

$$-k_s\frac{\partial^2 T_s}{\partial z^2} + \rho_B C_{p,s}\frac{\partial T_s}{\partial t} + \sum_k \rho_B C_{pa,k}\,q_k\frac{\partial T_s}{\partial t} + \rho_B\sum_k\left(\Delta H_k\frac{\partial q_k}{\partial t}\right) - HTC\,a_p(T_g - T_s) = 0 \quad (18)$$

Where k_s is the solid thermal conductivity (W/(m.K)), $C_{pa,k}$ the specific heat capacity of adsorbed substance (J/(K.kg)), and $C_{p,s}$ (J/(kg.K)) the specific heat capacity of sorbent valid from ambient conditions up to 120 °C (Low et al., 2023):

$$C_{p,s} = (-32300 \times T_s^{-2} + 0.00227 \times T_s + 0.994).10^3 \quad (19)$$

The heat of adsorption ΔH_k (J/mol) of CO_2 and H_2O is estimated using Clausius-Clapeyron equation with the fitted isotherms of adsorption:

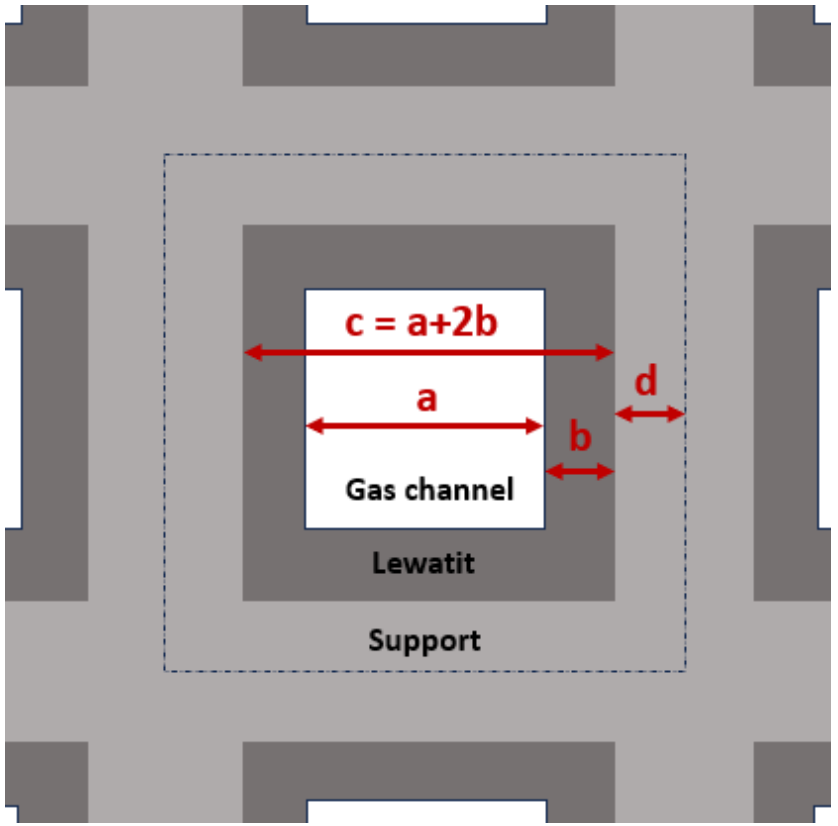
$$\frac{d\ln(p_k)}{d\frac{1}{T}} = -\frac{\Delta H_k}{R} \quad (20)$$

The estimation of ΔH_k is detailed in **Section 3** of the Supplementary Materials.

Bed density

	Design	Pellet	Monolith
Particle radius / Coating thickness	mm	0.334	0.3
Gas channel a	mm	-	1
Support half thickness d	mm	-	0.25
Active bed volume voidage ε_i	m^3/m^3	0.40	0.39
Bed volume voidage ε	m^3/m^3	0.40	0.23
Sorbent volume fraction	$m^3_{sorbent}/m^3$	0.60	0.35
Support volume fraction	$m^3_{support}/m^3$	0	0.42
Bed density (i.e. sorbent)	kg/m^3	476	281
Support density	$kg_{support}/m^3$	0	3301
Apparent sorbent interfacial area	m^2/m^3	5389	907

Table 5. Compacity differences between packed bed and monolith.



Parameters

Parameter	Symbol	Value	Unit	Source
Interparticle voidage	ε_i	0.40	-	(Scott and Kilgour, 1969; Weitz, 2004)
Intraparticle voidage	ε_p	0.34	-	(Low et al., 2023)
Sorbent bulk density	ρ_B	476	kg/m^3	(Low et al., 2023)
Particle radius	r_p	334	µm	Average particle radius from(Driessen et al., 2020)
Pore radius	r_{pore}	28.5	nm	(Lanxess, 2023)
Particle tortuosity	τ	2.5	-	Order of magnitude for sphere (Rezaei and Webley, 2010)
Adsorbed CO_2 heat capacity	$C_{pa,CO2}$	88	kJ/(kmol.K)	Estimated from liquid CO_2 (Wurzbacher, 2015)
Adsorbed H_2O heat capacity	$C_{pa,H2O}$	75	kJ/(kmol.K)	Estimated from liquid water (Peter Stephan et al., 2010)
Heat of adsorption CO_2	ΔH_{CO2}	73	kJ/mol	Clausius-Clapeyron estimation using CO_2 isotherms
Heat of adsorption H_2O	ΔH_{H2O}	44	kJ/mol	Clausius-Clapeyron estimation using H_2O isotherms
Sorbent thermal conductivity	k_s	0.43	W/(m.K)	(Bos et al., 2019)
Heat exchanger specific area	a_H	100	m^2/m^3	Order of magnitude (Schellevis and Brilman, 2024)
Heat transfer coefficient	U_H	50	W/(m^2.K)	Order of magnitude (Peter Stephan et al., 2010)
Heat capacity of indirect heating media	$C_{p,w}$	4.19	kJ/(kg.K)	Water as media

Table 3. Adsorbent properties and model parameters. Intraparticle voidage, pore radius, density, and sorbent thermal conductivity values are in inert conditions.

Clausius-Clapeyron

By plotting $ln(p_k) = f(\frac{1}{T})$ for a constant loading, the heat of adsorption ΔH_k (J/mol) of CO_2 and H_2O can be estimated using Clausius-Clapeyron equation. The slope of the linear curve gives the heat of adsorption at one loading:

$$\frac{dln(p_k)}{d\frac{1}{T}} = -\frac{\Delta H_k}{R} \qquad (12)$$

This was done for five different loadings in the case of CO_2 and H_2O, using the dry isotherms. As can be seen in **Figure S 4**, the heat of adsorption is dependent on the loading, as observed in previous literature (Low et al., 2023; Sonnleitner et al., 2018; Sutanto et al., 2017). For CO_2, the decision was to consider a typical median value (using the fitting curve equation) at a loading of 1.1 $molCO_2/kg$ for the CO_2 heat of adsorption implemented in the model, which gives $\Delta H_{CO_2} = 73$ $kJ/molCO_2$. For H_2O, the dependence is low, so it was decided to consider à reasonable value of $\Delta H_{H_2O} = 44$ $kJ/molH_2O$. These values are in the range of the values from literature (Low et al., 2023; Sonnleitner et al., 2018; Sutanto et al., 2017).

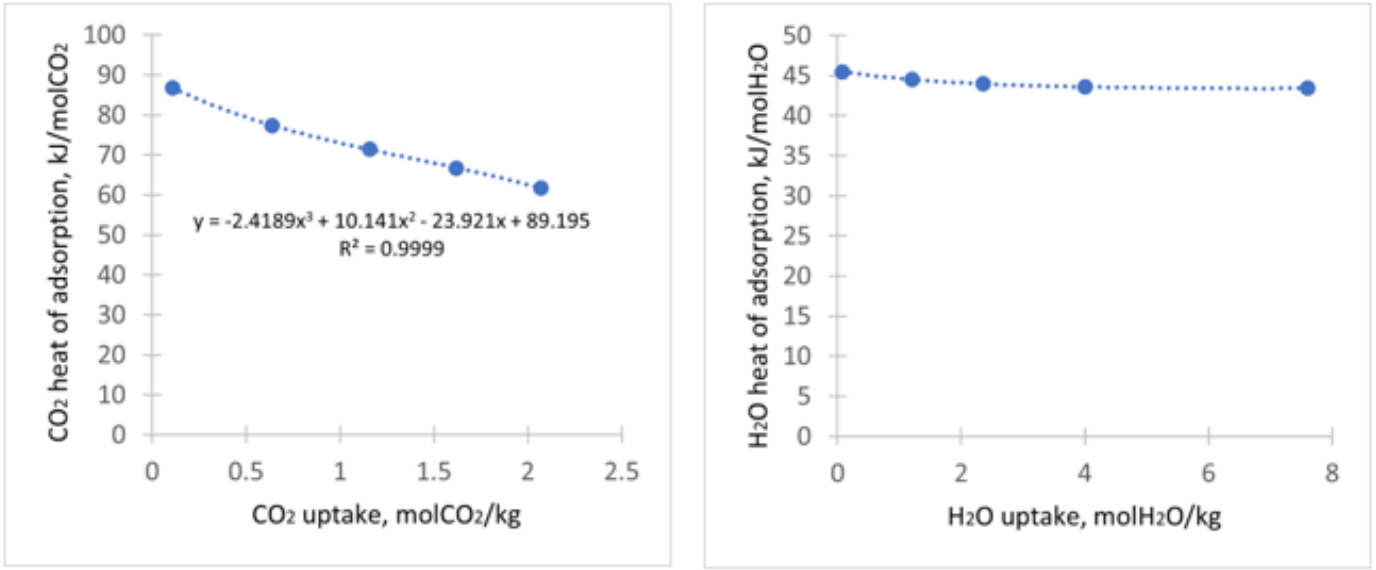


Figure S 4. CO_2 heat of adsorption (left) and H_2O heat of adsorption (right) as a function of the loading.

KPI

CO_2 working capacity	$molCO_2/kg$	CO_2 captured (after CPU) per mass of sorbent
$q_{CO2}^{max}/q_{CO2,amb}^{+}$	-	Fractional loading: Ratio of CO_2 uptake at the end of adsorption per equilibrium CO_2 uptake in ambient air
Capture rate	%	CO_2 capture rate, including losses in adsorption, evacuation and pre-purge steps
CO_2 productivity	$kgCO_2/(h.m^3)$	Ratio of the flux of CO_2 captured per bed volume, adjusted with a load factor of 0.9
H_2O productivity	$kgCO_2/(h.m^3)$	Ratio of the flux of H_2O captured per bed volume, adjusted with a load factor of 0.9
H_2O production	$kgH_2O/kgCO_2$	Ratio of the mass of H_2O captured per mass of CO_2 captured
Pressure drop	Pa	Air pressure drop in the bed during adsorption
Lineic pressure drop	Pa/m	Pressure drop divided by bed height
Indirect heat	GJ/tCO_2	Indirect heating requirement for regeneration
Steam heat	GJ/tCO_2	Steam heating requirement for regeneration
Fan work	MWh/tCO_2	Fan work
Vacuum work	MWh/tCO_2	Vacuum pump work
CPU work	MWh/tCO_2	Work of the compressors in the CPU
Total heat. energy	GJ/tCO_2	Sum of steam and indirect heating
Total elec. energy	MWh/tCO_2	Sum of electrical work of fan, vacuum pump and CPU compressors
Total primary energy	GJ/tCO_2	Total energy requirements using a primary energy factor of 2.3 for electricity (= thermal energy + electrical energy x 2.3)(Ministère de la transition écologique et de la cohésion des territoires, 2024)
Specific CAPEX	€/tCO_2	Specific capital cost
Specific OPEX	€/tCO_2	Specific operational cost
Cost of capture	€/tCO_2	Cost of capture (CAPEX + OPEX)

Table 6. Key performance indicators.

Economic estimation

Capital cost	
Components capital costs of Adsorber, Fan and Vacuum pump at unit scale	APEA estimation
Components capital costs of Adsorber, Fan and Vacuum pump at plant capacity scale (100 $ktCO_2/yr$)	15% learning rate on above cost
Components capital of Boiler, Condenser, Flash vessel and CPU	APEA estimation
Total field cost (TFC: total direct cost + total indirect cost)	Sum of above
Other cost (power supply, water treatment and conditioning components, steam integration, temporary installations, transport, etc.)	20% of TFC
Engineering, procurement, and construction	9% of TFC
Risks and contingencies	30% of sum of above
Total Plant Cost (TPC)	Sum of above
Spare parts	0.5% of TPC
Start-up	2% of TPC
Operator costs (other studies, enquiry, land purchase, site access, permitting, etc.)	7% of TPC
Insurance	0.5% of TPC
Local taxes	0.5% of TPC
Interim interest	17.5% of TPC
Total Capital Requirement (TCR)	Sum of above from TPC
O&M cost	
Operational labor cost	10 jobs/100 $ktCO_2/yr$; 60 k€/(jobs.yr)
Annual maintenance cost	3% of TPC

Other assumptions	
Cost basis	$€_{2022}$, Rotterdam, The Netherlands
Load factor	90%
Discount rate	8%
Economic lifetime	20 years
Electricity cost	70 €/MWh
Heat cost	10 €/GJ
Sorbent cost	15.6 €/kg
Monolith support cost	2 €/kg
Sorbent/monolith lifetime	2 years
N_2 cost	1 €/kg
H_2O cost	0.5 €/tH_2O

Table 8. Assumptions for the capital costs estimation and the overall economic evaluation.

Clausius-Clapeyron

By plotting $ln(p_k) = f(\frac{1}{T})$ for a constant loading, the heat of adsorption ΔH_k (J/mol) of CO_2 and H_2O can be estimated using Clausius-Clapeyron equation. The slope of the linear curve gives the heat of adsorption at one loading:

$$\frac{dln(p_k)}{d\frac{1}{T}} = -\frac{\Delta H_k}{R} \qquad (12)$$

This was done for five different loadings in the case of CO_2 and H_2O, using the dry isotherms. As can be seen in **Figure S 4**, the heat of adsorption is dependent on the loading, as observed in previous literature (Low et al., 2023; Sonnleitner et al., 2018; Sutanto et al., 2017). For CO_2, the decision was to consider a typical median value (using the fitting curve equation) at a loading of 1.1 $molCO_2/kg$ for the CO_2 heat of adsorption implemented in the model, which gives $\Delta H_{CO_2} = 73$ $kJ/molCO_2$. For H_2O, the dependence is low, so it was decided to consider à reasonable value of $\Delta H_{H_2O} = 44$ $kJ/molH_2O$. These values are in the range of the values from literature (Low et al., 2023; Sonnleitner et al., 2018; Sutanto et al., 2017).

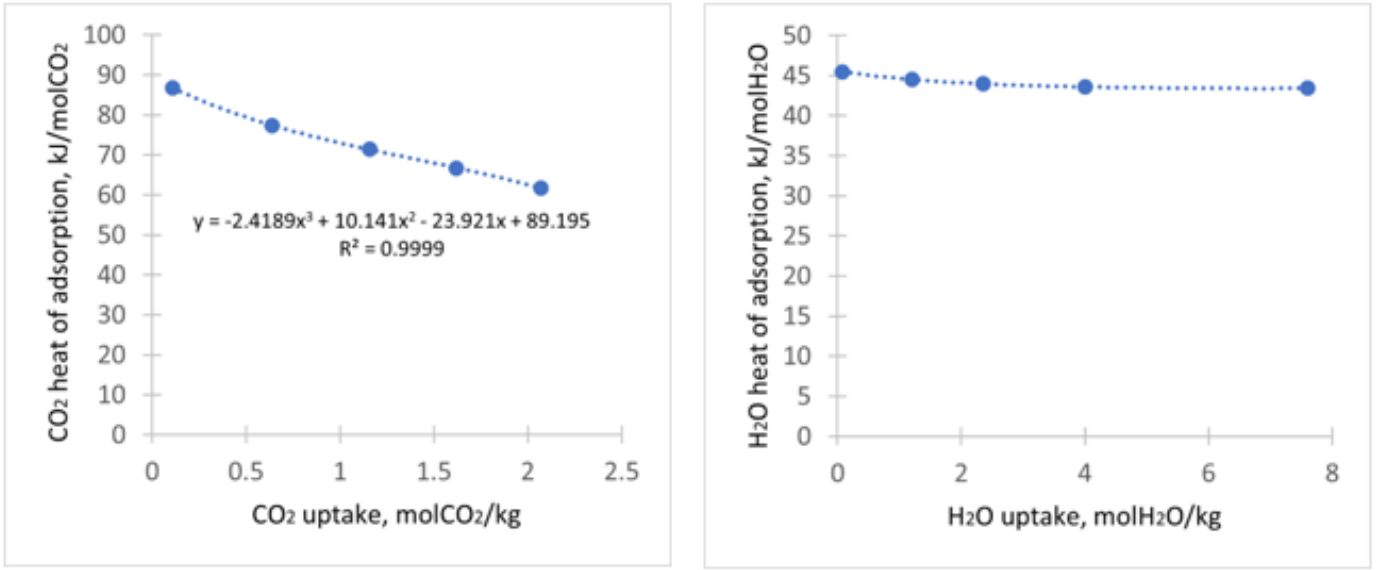


Figure S 4. CO_2 heat of adsorption (left) and H_2O heat of adsorption (right) as a function of the loading.

Sensitivity on monolith

Section 7: Qualitative effects on performance of the sensitivity analysis

Parameter	Effect	Remarks
↘ Bed height	↗ Productivity ↘ ΔP (& energy)	Minimum bed height is limited by economy of scale and technology limitations
↗ v_g	↗ Productivity ↗ ΔP (& energy)	Productivity is limited high velocity when gas residence time become lower to the CO_2 mass transfer characteristic time
↘ Coating thickness	↗ Productivity ↗/↘ Energy	Optimum can be found for minimum energy
↘ Air temperature	↗ Productivity	
↗ RH	↗ Productivity ↗ H_2O production ↘ Energy	
↗ Adsorption criterion	↗/↘ Productivity ↗/↘ Energy	Optimum Optimum
↘ Regeneration vacuum pressure	↗ Productivity ↗ VP work	Vacuum level can be limited by vacuum pump technology
↘ Desorption criterion	↗/↘ Productivity ↘ Energy	Optimum
↗ Regeneration temperature	↗ Productivity ↗/↘ Energy	Optimum
↗ Regeneration steam flowrate	↗ Productivity ↗ Energy	
↗ MTC	↗ Productivity ↘ Energy	

Table S 5. Qualitative analysis of design and operational parameters on performance for monolith design.

Monolith assumptions

2. The axial dispersion coefficient is calculated using the Taylor relation for laminar flow in tube (Taylor, 1953):

$$E_{z,k} = D_{m,k} + \frac{v_g^2\, a^2}{\varepsilon_i\, 192\, D_{m,k}} \qquad (21)$$

Where a is the channel space (m).

3. The heat transfer coefficient is obtained using the Hawthorn correlation for squared channel (Crittenden et al., 2011):

$$HTC = \frac{2.977\, k_g}{a}\left(1 + 0.095\frac{a\, Re\, Pr}{L}\right)^{0.45} \qquad (22)$$

Where L is the length of the channel (m).

4. MTC_k is the lumped mass transfer coefficient (1/s) for monolith (Rezaei and Webley, 2009):

$$MTC_k = \frac{\varepsilon_i K_k}{\rho_B \frac{dq_k^*}{dP_k} RTZ} \qquad (23)$$

Where K_k (1/s) is given as a lumped term comprising the external film resistance term and the pore diffusion term:

$$\frac{1}{K_k} = \frac{a}{4\, \varepsilon_i\, k_{f,k}} + \frac{c^2 - a^2}{32\, \varepsilon_i\, K_{p,k}} \qquad (24)$$

The film transfer coefficient $k_{f,k}$ (m/s) is obtained from the Sherwood Sh_k number (Rezaei and Webley, 2009):

$$k_{f,k} = Sh_k \frac{D_{m,k}}{a} \qquad (25)$$

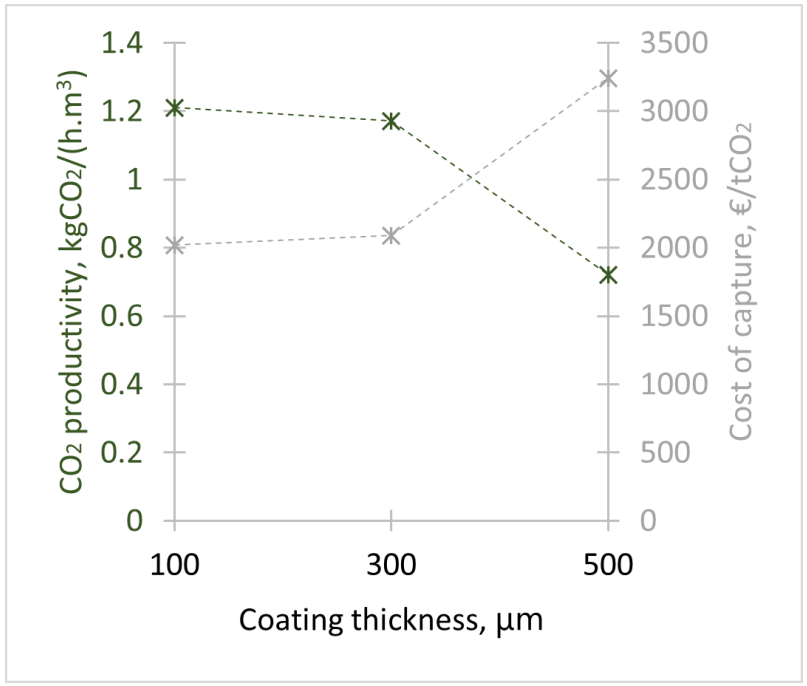
Where $Sh_k = 2.95\left(1 + 0.139\, Sc_k\, Re\, \frac{a}{H_b}\right)^{0.81}$, with $Sc_k = \frac{\mu}{D_{m,k}\rho_g}$ the Schmidt number and $Re = \frac{v_g 2r_p \rho_g}{\mu}$ the Reynolds number.

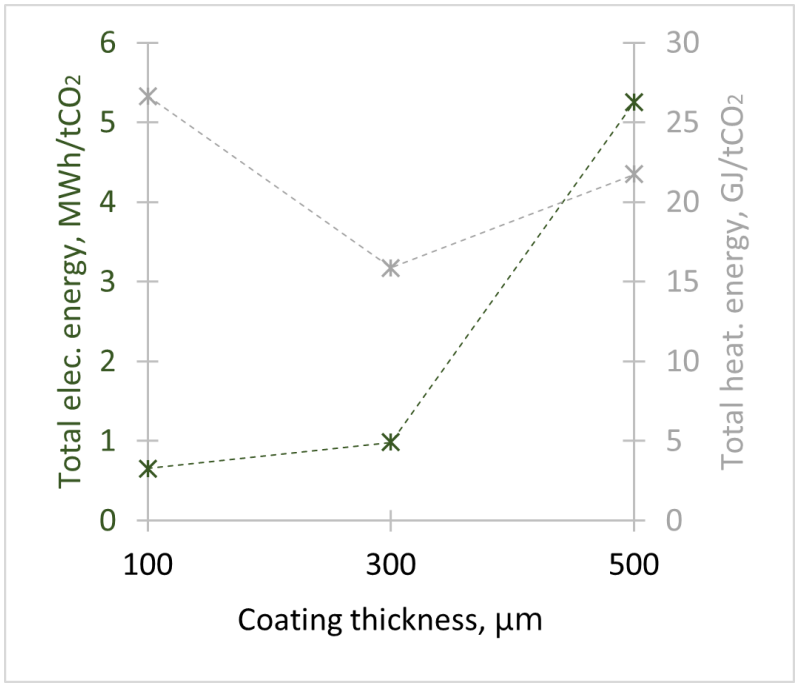
5. The pore diffusion coefficient $K_{p,k}$ is the same as for pellets, except for the tortuosity which is equal to $\tau = 1$.

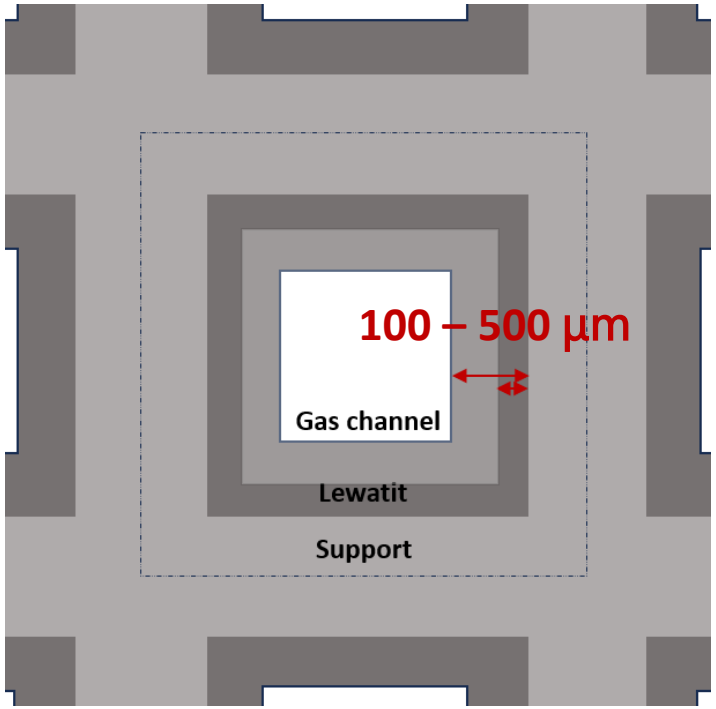
6. Pressure drop across the monolith considers Hagen-Poiseuille equation for squared channel (Patton et al., 2004):

$$\frac{\partial P}{\partial z} = -\frac{28.4\, \mu}{\varepsilon_i\, a^2} v_g \qquad (26)$$

Results : Sensitivity on coating thickness for monolith







Monolith

Key message:

Larger coating thickness:

- Smaller productivity
- Higher air velocity → higher pressure drop → higher elec. requirement
- Optimum to find on heat. Energy

→ Higher cost of capture

Main references

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Isotherme d'adsorption de la Lewatit

Isotherme de Toth, en condition sèche

$$q_{CO2}^{dry}(T, p_{CO2}) = q_S . \frac{b(T).p_{CO2}}{(1 + (b(T).p_{CO2})^{t(T)})^{\frac{1}{t(T)}}}$$

avec

$$b(T) = b_0 . e^{\frac{\Delta H_0}{R.T_0}.\left(\frac{T_0}{T}-1\right)}$$

$$t(T) = t_0 + \alpha . \left(1 - \frac{T_0}{T}\right)$$

Estimation des paramètres q_S, b_0, ΔH_0, t_0 et α par régression d'isothermes de la littérature

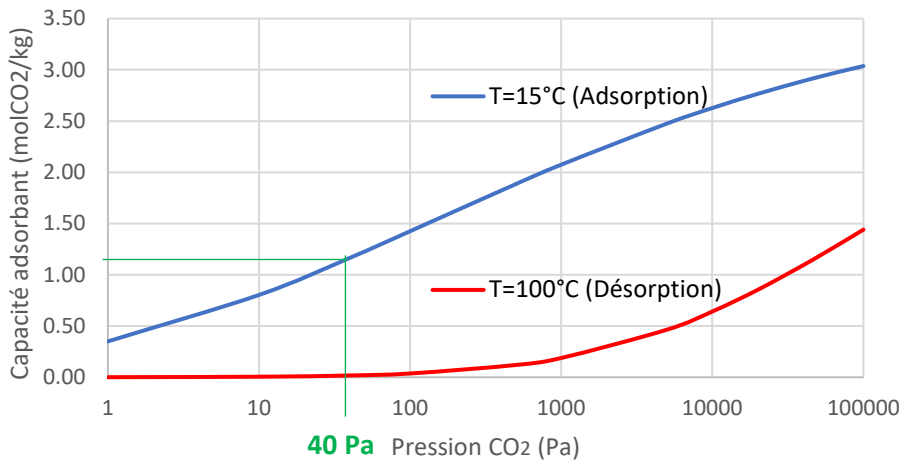
MATLAB

Pressure range (Pa)	Temperature range (°C)	Nb data	Ref
10-100	5-35	26	(H. M. Schellevis, Van Schagen, et Brilman 2021)
40-1000	5-35	21	(M. Schellevis 2023)
30-100000	25-100	59	(Young et al. 2021)
10-100000	15-120	324	(M.-Y. A. Low et al. 2023)
1500-50000	40-95	24	(Sonnleitner, Schöny, et Hofbauer 2018)
1000-80000	30-100	42	(R. Veneman et al. 2015)
5-100000	25-90	51	(Wu et al. 2022)

	Parameter	Value
Toth isotherm (Pure CO_2)	q_S (mol_{CO2}/kg)	3.7604
	b_0 (1/Pa)	0.001382
	ΔH_0 (kJ/mol_{CO2})	103.05
	T_0 (K)	353.15
	t_0 (-)	0.30894
	α (-)	0.34148
	R^2	0.9803

CO_2 en condition sèche (RH=0%)

*RH=Humidité Relative



Isotherme d'adsorption de la Lewatit

L'humidité de l'air favorise l'adsorption du CO_2, isotherme prenant en compte l'humidité de l'air (M. Schellevis 2023) :

$$q_{CO2}^{wet}\left(q_{CO2}^{dry}, RH\right) = \left(b.e^{c.q_{CO2}^{dry}(T,p_{CO2})}.RH + 1\right).q_{CO2}^{dry}(T,p_{CO2})$$

avec

RH l'humidité relative

$q_{CO2}^{dry}(T,p_{CO2})$ calculé par l'isotherme de Toth

Estimation des paramètres b et c par régression d'isothermes de la littérature

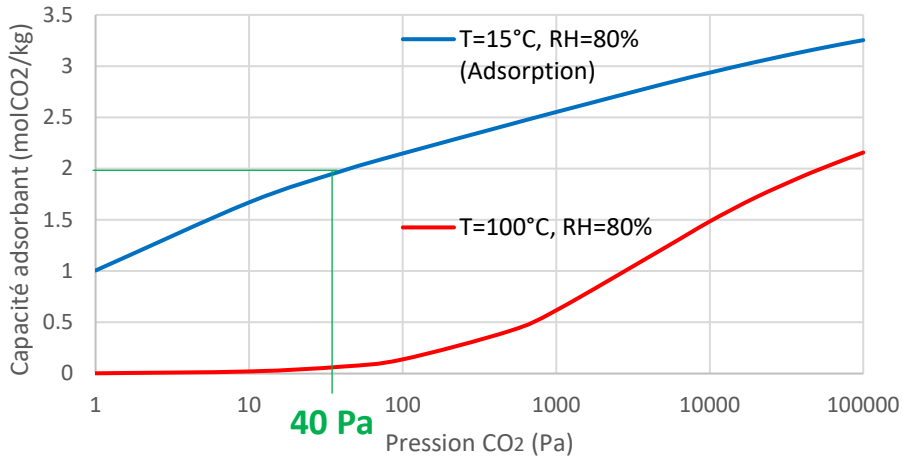
MATLAB

Pressure range (Pa)	Temperature range (°C)	RH range (-)	Nb data	Ref
40-1000	5-35	0-0.9	76	(M. Schellevis 2023)
10-100000	25-70	0.3-0.55	42	(Young et al. 2021)
40	15-35	0-0.6	12	(R. Veneman et al. 2015)
15-130	15-30	0.35-0.37	17	(Chimani et al. 2024)

	Parameter	Value
Co-adsorption isotherm CO_2	b (-)	3.58
	c (kg/mol$_{CO2}$)	-1.215
	R^2	0.873

CO_2 en condition humide (RH=80%)

*RH=Humidité Relative



Isotherme d'adsorption de la Lewatit

L'adsorption de l'eau n'est pas influencé par le CO_2, l'isotherme Guggenheim-Anderson-de-Boer (GAB) est employé :

$$q_{H2O}(T, RH) = q_G . \frac{C_G(T).K_G(T).RH}{(1 - K_G(T).RH).(1 + (C_G(T) - 1).K_G(T).RH)}$$

avec

$$C_G(T) = e^{\frac{(E_1 - E_{10+})}{R.T}}$$

$$K_G(T) = e^{\frac{(E_{2-9} - E_{10+})}{R.T}}$$

$$E_1 = C - e^{D.T}$$

$$E_{2-9} = F + G.T$$

$$E_{10+} = -44.38.T + 57220$$

Estimation des paramètres par régression d'isothermes de la littérature

MATLAB

Pressure range (Pa)	Temperature range (°C)	RH range (-)	Nb data	Ref
230-2000	20	0.1-0.9	6	(Chimani et al. 2024)
20-2200	15-35	0.01-0.9	138	(M.-Y. A. Low et al. 2023)
30-2500	8-33	0.01-0.9	33	(M. Schellevis 2023)
150-30000	25-100	0.03-0.9	55	(Young et al. 2021)

	Parameter	Value
GAB isotherm (H_2O)	q_G (mol_{H2O}/kg)	2.15
	C (J/mol_{H2O})	48459
	D (1/K)	0.0234
	F (J/mol_{H2O})	57197
	G ($J/mol_{H2O}/K$)	-44.93
	R^2	0.988

H_2O