

Technoeconomic Evaluation of a Process Capturing CO₂ Directly from Air

Romesh Pramodya Wijesiri ^{1,2}, Gregory Paul Knowles ², Hasina Yeasmin ¹, Andrew Forbes Alexander Hoadley ^{1,*} and Alan Loyd Chaffee ^{2,*}

¹ Department of Chemical Engineering, 16 Alliance Lane, Clayton Campus, Monash University, VIC 3800, Australia

² School of Chemistry, 17 Rainforest Walk, Clayton Campus, Monash University, VIC 3800, Australia

* Correspondence: Andrew.Hoadley@monash.edu (A.F.A.H.); Alan.Chaffee@monash.edu (A.L.C.); Tel.: +61-3-9905-3421 (A.F.A.H.); Tel.: +61-3-9905-4626 (A.L.C.)

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S1. Calculations for the unit operations in the process

S1.1. Adsorption stage

S1.1.1. Pressure drop across bed

Pressure drop across a sorbent bed was calculated by the Ergun equation [1] (Equation (S1)).

$$\frac{\Delta P_{bed,ads}}{H_{bed}} = 150 \frac{\mu_{air} V_{air}(1-\epsilon)^2}{d_{pellet}^2 \epsilon^3} + 1.75 \frac{\rho_{air} V_{air}^2 (1-\epsilon)}{d_{pellet} \epsilon^3}, \quad (S1)$$

Where $\Delta P_{bed,ads}$ is the pressure drop (Pa), H_{bed} is the bed height (m), μ_{air} is the dynamic viscosity of air (Pa s), V_{air} is the superficial velocity of air (m s⁻¹), ϵ is the void fraction of the bed, and d_{pellet} is the diameter of the sorbent pellets (m).

S1.1.2. Fan power calculation

The power requirement of the fan (W), W_{fan} , was calculated as the total of the energy required to push the air through the contactor (W), ($W_{fan,contactor}$), and the energy required to push the air through the heater/steam condenser (W), ($W_{fan,heater}$), according to Equations (S2) and (S3).

$$W_{fan} = W_{fan,contactor} + W_{fan,heater} \quad (S2)$$

$$W_{fan,contactor} = \frac{1}{\eta_{fan}} \Delta P \times V \times N_{beds} \times (N_{contactors} - 1) \quad (S3)$$

Where η_{fan} is the efficiency of the fan (80%), \dot{V} is the volumetric flow rate of air (m³ s⁻¹) through one bed, N_{beds} is the number of beds in a contactor and $N_{contactors}$ is the number of contactors in parallel in the system.

The energy needed to push the air through the fan cooler, $W_{fan,heater}$, was estimated as 0.005 W per 1 W of cooling [2]. The calculation of the cooling duty of the fan is done in Section S1.2.6.

S1.2. Desorption stage

S1.2.1. Pressure drops through heat exchangers

For the desorption stage, each heat exchanger was assumed to have a pressure drop of 5 kPa.

S1.2.2. Boiler

The boiler duty was calculated according to Equation (S4), where Q_{boiler} is the boiler duty (W), \dot{m}_{H_2O} is the mass flow rate of steam (kg s⁻¹), $C_{p,water}$ is the specific heat capacity of water (J kg⁻¹ °C⁻¹), and H_{latent,H_2O} is the latent heat of vaporization (J kg⁻¹). T_{boil} and T_{amb} are the bubble point temperature (°C) of water at the respective pressure and the inlet temperature (°C) of water, respectively.

$$Q_{boiler} = \dot{m}_{H_2O} (C_{p,water} (T_{boil} - T_{amb}) + H_{latent,H_2O}) \quad (S4)$$

T_{boil} was calculated according to the Antoine correlation for water (Equation (S5)), where P_{boiler} is the pressure in the boiler (mm Hg).

$$T_{boil} = \frac{1730.63}{8.07131 - \log_{10}(P_{boiler})} - 233.426 \quad (S5)$$

S1.2.3. Superheater

The superheater duty was calculated according to Equation (S6), where $Q_{superheater}$ is the superheater duty (W), \dot{m}_{H_2O} is the mass flow rate of steam (kg s^{-1}) and $C_{p,steam}$ is the specific heat capacity of steam ($\text{J kg}^{-1} \text{ } ^\circ\text{C}^{-1}$). T_{boil} and T_d are the boiling point temperature ($^\circ\text{C}$) of water and the desorption temperature ($^\circ\text{C}$), respectively.

$$Q_{boiler} = \dot{m}_{H_2O} C_{p,steam} (T_d - T_{boil}) \quad (S6)$$

S1.2.4. Contactor

The mass flow of CO_2 and H_2O out from the contactor were calculated according to Equations (S7), where $\dot{m}_{i,in}$ is the mass flow rate in to the contactor (kg s^{-1}), m_{sorbent} is the mass of the sorbent in the contactor (kg) and MW_i is the molar mass (kg mol^{-1}). $\frac{dq_i}{dt}$ is the mass transfer rate of CO_2 and H_2O to/from the sorbent ($\text{mol kg}^{-1}\text{s}^{-1}$) calculated according to Equation (1).

$$\dot{m}_{i,out} = \dot{m}_{i,in} - \frac{dq_i}{dt} \times m_{\text{sorbent}} \times MW_i \quad (S7)$$

where the component i refers to either CO_2 or H_2O .

The heat supplied to the bed from the heat transfer fluid was calculated according to Equation (S8).

$$Q_{bed} = U_{\text{contactor}} A_{\text{contactor}} (T_{\text{heat}} - T_{\text{bed}}) \quad (S8)$$

Where Q_{bed} is the heat supplied to the bed (W), $U_{\text{contactor}}$ is the overall heat transfer coefficient for the contactor ($\text{W m}^{-2} \text{ } ^\circ\text{C}^{-1}$), $A_{\text{contactor}}$ is the heat transfer area (m^2) in the contactor and T_{bed} is the temperature ($^\circ\text{C}$) of the bed. T_{heat} is the temperature ($^\circ\text{C}$) of the heat transfer fluid, which is assumed to be $10 \text{ } ^\circ\text{C}$ higher than the target desorption temperature. $U_{\text{contactor}}$ was assumed to be $150 \text{ W m}^{-2} \text{ } ^\circ\text{C}^{-1}$ [3].

S1.2.5. Desuperheater

$\dot{m}_{\text{water,desuperheat}}$, the flow rate of water (kg s^{-1}) which needs to be added to desuperheat the steam was calculated according to Equation (S9). The CO_2 and steam entering the desuperheater were assumed to be at the same temperature as the contactor temperature, T_{bed} , as calculated according to Equation (7).

$$\dot{m}_{\text{water,desuperheat}} = \frac{\dot{m}_{\text{steam}} C_{p,steam} (T_{bed} - T_{boil}) + \dot{m}_{\text{CO}_2} C_{p,\text{CO}_2} (T_{bed} - T_{boil})}{C_{p,water} (T_{boil} - T_{amb}) + H_{\text{lat,water}}} \quad (S9)$$

S1.2.6. Condenser

The concentration of H_2O exiting the condenser was calculated according to Equations (S10), where $y_{\text{H}_2\text{O}}$ is the concentration of H_2O ($\text{mol-H}_2\text{O mol}^{-1}$), $P_{\text{sat,H}_2\text{O}}$ is the saturation vapour pressure of water (Pa), and $P_{\text{condenser}}$ is the pressure inside the condenser (Pa). $P_{\text{sat,H}_2\text{O}}$ was calculated according to the Antoine correlation (Equation (S11)), where $T_{\text{condenser}}$ is the condensation temperature ($^\circ\text{C}$).

$$y_{H_2O,out} = \frac{P_{sat,H_2O}}{P_{condenser}} \quad (S10)$$

$$p_{sat,H_2O} = 10^{8.07131 - \frac{1730.63}{233.426 + T_{condenser}}} \quad (S11)$$

The mass flow rates (kg s^{-1}) of the water vapor exiting the condenser, $\dot{m}_{vap,out}$, and that of the condensate, $\dot{m}_{water,out}$, were calculated according to Equations (S12) and (S13), where $\dot{m}_{CO_2,in}$ and $\dot{m}_{H_2O,in}$ are the CO_2 and H_2O mass flow rates (kg s^{-1}) in to the condenser.

$$\dot{m}_{vap,out} = \dot{m}_{CO_2,in} \times \frac{y_{H_2O,out}}{1 - y_{H_2O,out}} \quad (S12)$$

$$\dot{m}_{water,out} = \dot{m}_{H_2O,in} - \dot{m}_{vap,out} \quad (S13)$$

The cooling duty of the condenser was calculated according to Equation (S14).

$$Q_{condenser} = \dot{m}_{water,out}(C_{p,water}(T_{condenser,out} - T_{condenser,in}) + H_{lat,H_2O}) + \dot{m}_{vap,out}C_{p,steam}(T_{condenser,out} - T_{condenser,in}) + \dot{m}_{CO_2,in}C_{p,CO_2}(T_{condenser,out} - T_{condenser,in}) \quad (S14)$$

S1.2.7. Vacuum Pump

The energy required for operating the vacuum pumps to pump the desorbed product out (W), $W_{vacuum\ pump}$, was calculated according to Equations (S15)–(S18) [3], where the η_{vacuum} is the efficiency of the vacuum pump (70%), \dot{n}_i is the molar flow rate (mol s^{-1}) in of CO_2 or H_2O , R is the gas constant ($\text{J mol}^{-1}\text{K}^{-1}$), and T_{in} is the temperature ($^{\circ}\text{C}$) of the gas entering the vacuum pump. P_{in} and P_{out} are the pressures (Pa) of the gas entering and exiting the vacuum pump.

$$W_{vacuum,pump} = \sum_{i=CO_2,H_2O} \frac{1}{\eta_{vacuum}} \dot{n}_i R (T_{in} + 273) \left(\frac{n_{p,i}}{n_{p,i} - 1} \right) \left(\left(\frac{P_{out}}{P_{in}} \right)^{\frac{n_{p,i} - 1}{n_{p,i}}} - 1 \right) \quad (S15)$$

$$m_{p,i} = \frac{\gamma_i - 1}{\gamma_i} \frac{1}{\eta_{vacuum}} \quad (S16)$$

$$\gamma_i = \frac{C_{p,i}}{C_{p,i} - R} \quad (S17)$$

$$\gamma_i = \frac{C_{p,i}}{C_{p,i} - R} \quad (S18)$$

where the component i refers to either CO_2 or H_2O .

S2. Calculation of the Energy consumption

The energy consumptions were calculated on a basis of GJ tonne^{-1} of CO_2 captured according to Equations (S19)–(S25).

S2.1. Electrical Energy

$$E_{fan} = \frac{\int_0^{t_{ads}} W_{fan} dt}{\text{mass of } CO_2 \text{ captured per cycle (kg)}} \times \frac{1}{1 \times 10^6} \text{GJ kg J}^{-1} \text{tonne}^{-1} \quad (S19)$$

Where t_{ads} is the total adsorption time (s).

$$E_{vacuum} = \frac{\int_0^{t_{des}} W_{vacuum} dt + E_{evacuation}}{\text{mass of } CO_2 \text{ captured per cycle (kg)}} \times \frac{1}{1 \times 10^6} \text{GJ kg J}^{-1} \text{tonne}^{-1} \quad (S20)$$

Where $E_{\text{evacuation}}$ is the energy needed (J) to evacuate the contactor to the desorption pressure according to Equation (S21) [3], where V is the void volume inside the contactor (m^3), m_{air} is the mass of air being evacuated (kg), and P_{initial} and P_{final} are the pressures (kPa abs) at the beginning and end of the evacuation respectively. n_{air} is calculated according to Equations (S16)–(S18).

$$E_{\text{evacuation}} = -\frac{1}{\eta_{\text{vacuum}}} P_{\text{initial}} V_{\text{contactor}} \frac{n_{p,\text{air}}}{n_{p,\text{air}} - 1} \left(\left(\frac{P_{\text{final}}}{P_{\text{initial}}} \right)^{\frac{n_{p,\text{air}}}{n_{p,\text{air}} - 1}} - 1 \right) m_{\text{air}} \quad (\text{S21})$$

The total electrical energy requirement was calculated according to Equation (S22).

$$E_{\text{electrical}} = E_{\text{fan}} + E_{\text{vacuum}} \quad (\text{S22})$$

S2.2. Thermal Energy

$$E_{\text{steam}} = \frac{\int_0^{t_{\text{des}}} (Q_{\text{boiler}} + Q_{\text{superheater}}) dt}{\text{mass of } \text{CO}_2 \text{ captured per cycle (kg)}} \times \frac{1}{1 \times 10^6} \text{GJ kg J}^{-1} \text{tonne}^{-1} \quad (\text{S23})$$

Where t_{des} is the desorption time (s)

$$E_{\text{bed}} = \frac{\int_0^{t_{\text{des}}} Q_{\text{bed}} dt}{\text{mass of } \text{CO}_2 \text{ captured per cycle (kg)}} \times \frac{1}{1 \times 10^6} \text{GJ kg J}^{-1} \text{tonne}^{-1} \quad (\text{S24})$$

The total electrical energy requirement was calculated according to Equation (S25).

$$E_{\text{electrical}} = E_{\text{steam}} + E_{\text{bed}} \quad (\text{S25})$$

S3. Capital cost estimation

S3.1. Contactor

The air contactor was costed as a shell and tube heat exchanger due to its unique configuration, according to the cost correlation (Equations (S26)) found in Sinnott [3].

$$C_{\text{contactor}} = 28000 + 54A_{\text{contactor}}^{1.2} \quad 10 < A_{\text{contactor}} < 1000 \quad (\text{S26})$$

Where $C_{\text{contactor}}$ is the cost of a contactor (USD) and $A_{\text{contactor}}$ is the heat transfer area (m^2).

In scaling up the contactor, it was assumed that the $\frac{UA}{m_{\text{sorbent}}}$ ratio would be kept constant. So the UA of the scaled up contactor was calculated according to Equation (S27).

$$C_{\text{contactor}} = 28000 + 54A_{\text{contactor}}^{1.2} \quad 10 < A_{\text{contactor}} < 1000 \quad (\text{S27})$$

$A_{\text{contactor}}$ was calculated by assuming $U_{\text{contactor}}$ to be $150 \text{ W m}^{-2} \text{ } ^\circ\text{C}$ [3].

S3.2. Fan

Costed according to correlation (Equation (S28)) in Couper [2], where C_{fan} is the cost of the fan (USD) and \dot{V}_{air} is the air flow rate in SCFM.

$$C_{\text{fan}} = 2680e^{0.4692+0.1203 \ln(\dot{V}_{\text{air}})+0.0931(\ln(\dot{V}_{\text{air}}))^2} \quad \text{for } 2000 < \dot{V}_{\text{air}} < 500000 \quad (\text{S28})$$

S3.3. Boiler

Costed according to correlation (Equation (S29)) in Sinnott [3], where C_{boiler} is the cost of the boiler (USD) and A_{boiler} is the heat transfer area (m^2) of the boiler.

$$C_{\text{boiler}} = 29000 + 400A_{\text{boiler}}^{0.9} \quad 10 < A_{\text{boiler}} < 500 \quad (\text{S29})$$

A_{boiler} was calculated using Equation (S30), by assuming a log mean temperature difference (ΔT_{LM}) of 15 °C, and a U_{boiler} of 1000 W m⁻² °C⁻¹ [3].

$$A_{boiler} = \frac{Q_{boiler}}{U_{boiler} \Delta T_{LM}} \quad (S30)$$

S3.4. Superheater

Costed according to correlation (Equation (S31)) in Sinnott [3], where $C_{superheater}$ is the cost of the superheater (USD) and $A_{superheater}$ is the heat transfer area of the superheater (m²).

$$C_{superheater} = 28000 + 54A_{superheater}^{0.9} \quad 10 < A_{superheater} < 1000 \quad (S31)$$

$A_{superheater}$ was calculated using Equation (S32), by assuming a log mean temperature difference (ΔT_{LM}) of 15 °C, and a $U_{superheater}$ of 30 W m⁻² °C⁻¹ [3].

$$A_{superheater} = \frac{Q_{superheater}}{U_{superheater} \Delta T_{LM}} \quad (S32)$$

S3.5 Condenser

Costed according to correlation (Equations (S33)) in Couper [2], where $C_{condenser}$ is the cost of the condenser (USD) and $A_{condenser}$ is the heat transfer area (ft²) of the condenser.

$$C_{condenser} = 475000A_{condenser}^{0.4} \quad 50 < A_{condenser} < 200000 \quad (S33)$$

$A_{superheater}$ was calculated using Equation (S34), by assuming a $U_{condenser}$ of 500 W m⁻² °C⁻¹ [2].

$$A_{condenser} = \frac{Q_{condenser}}{U_{condenser} \Delta T_{LM}} \quad (S34)$$

S3.6. Vacuum pump

Costed according to correlation (Equation (S35)) in Couper [2], where C_{vacuum} is the cost of the vacuum pump (USD), \dot{m}_{gas} is the mass of gas being pumped (lbs h⁻¹) and P_1 is the suction side pressure (torr). The average mass flow rate through the vacuum pump during the desorption was taken as \dot{m}_{gas} .

$$C_{vac} = 9930 \left(\frac{\dot{m}_{gas}}{P_d} \right)^{1.03} \quad 0.3 < \frac{\dot{m}_{gas}}{P_1} < 15 \quad (S35)$$

S3.7. Adjustment for inflation

As the cost data was based on data from 2003 (Couper [2]) and 2010 (Sinnott [3]) All the equipment capital costs were adjusted for inflation using Equation (S36) and the CEPCI data in Table S1.

$$\frac{C_{year\ x}}{C_{year\ y}} = \frac{CEPCI_{year\ x}}{CEPCI_{year\ y}} \quad (S36)$$

Table S1. CEPCI data for the relevant years.

Year	2003	2010	2017
CEPCI	402	550	567

S3.8. Total plant cost

The total plant cost (USD), C_{plant} , was calculated according to Equation (S37), where the cost of insulation, piping, instrumentation, electrical work, civil and structures, and lagging are accounted for by multiplying the total equipment cost by a factor, f_i , from Table S2 [4].

$$C_{plant} = \sum f_i \times (C_{fan} + C_{bed} + C_{boiler} + C_{superheater} + C_{condenser} + C_{vacuum}) \quad (S37)$$

Table S2. Factors to account for the total plant cost.

	Installation	Piping	Instruments	Electrical	Civil	Structural	Lagging
f_i	0.08	0.2	0.1	0.19	0.1	0.02	0.04

S4. Calculation of the Cost of sorbent

The cost of raw material for the production of the sorbent was assumed based on the prices of generic mesoporous silica and branched PEI, to be 4 USD kg⁻¹ [5] and 2 USD kg⁻¹ [6], respectively. It was also assumed that the cost of raw material is only 1/3 of the total production cost. For a 65% wt PEI sorbent like the one considered in this study, $C_{sorbent, material}$, was calculated according to Equation (S38).

$$C_{sorbent} = (0.35 \times 4 + 0.65 \times 2) \times 3 = 8.1 \text{ USD kg}^{-1} \quad (S38)$$

S5. NPV analysis

To determine the cost of capture (USD tonne⁻¹), C_{CO_2} , a NPV analysis was carried out according to Equations (S39) and (S40) for a breakeven scenario, for 20 years with a discount factor of 10%, where CR is the annual capture rate (tonne yr⁻¹) of CO₂.

$$NPV = 0 = -C_{plant} - C_{annual\ opex} \sum_{x=1}^{20} \frac{1}{(1+0.1)^x} + C_{sorbent} \sum_{y=0,4,\dots,16,20} \frac{1}{(1+0.1)^y} + C_{CO_2} \times CR_{annual} \times \sum_{x=1}^{20} \frac{1}{(1+0.1)^x} \quad (S39)$$

$$C_{CO_2} = \frac{(C_{plant} + C_{annual\ opex} \sum_{x=1}^{20} \frac{1}{(1+0.1)^x} + C_{sorbent} \sum_{y=0,4,\dots,16,20} \frac{1}{(1+0.1)^y})}{CR \times \sum_{x=1}^{20} \frac{1}{(1+0.1)^x}} \quad (S40)$$

S6. Desorption Stage: Model Validation contd.

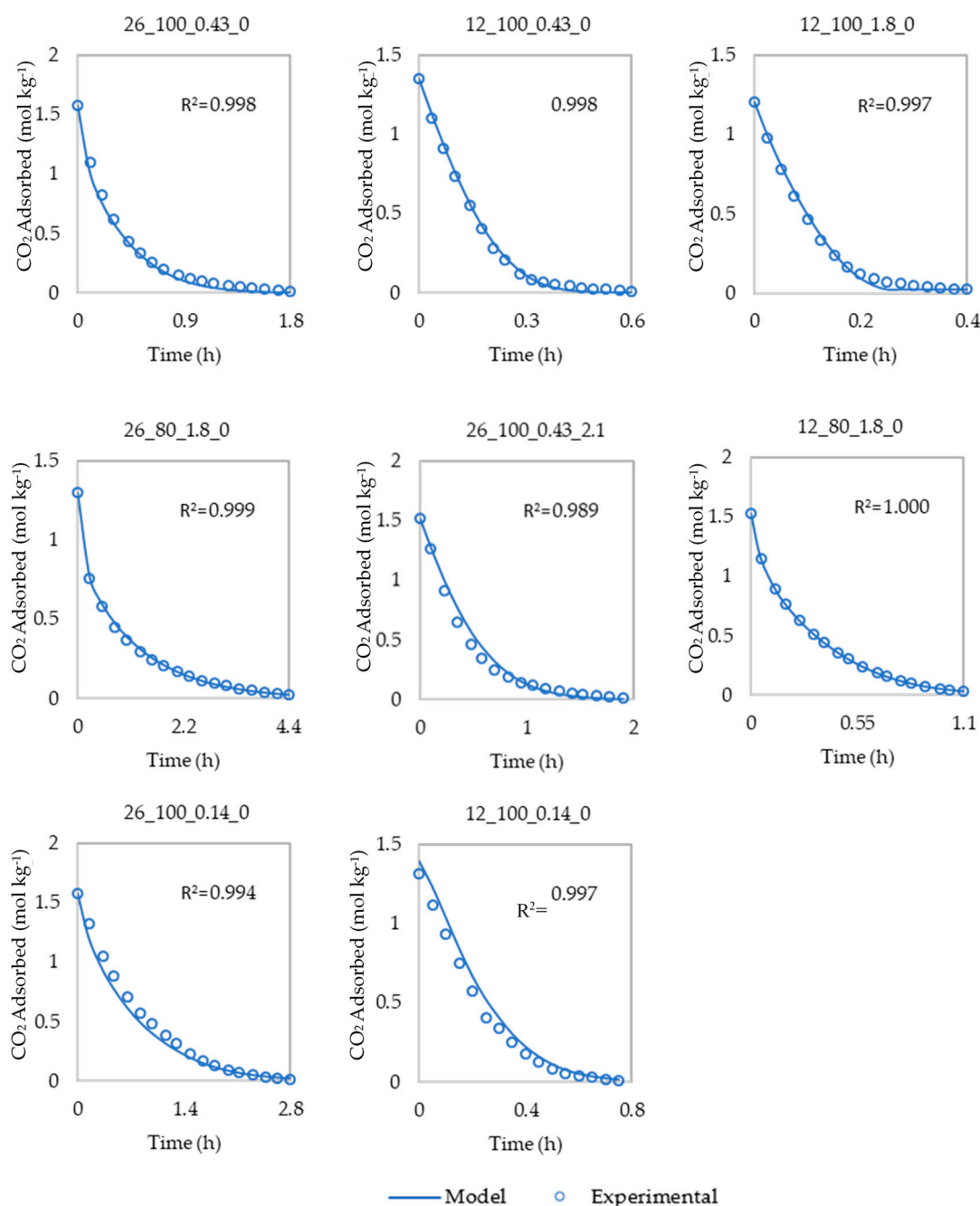


Figure 1. The experimental data and the model predictions for CO₂ mass transfer kinetics in the desorption stage. Legend for titles AA_BBB_CCC_DD (AA- desorption pressure (kPa), BBB- desorption temperature (°C), CCC- desorption steam flow rate (kg h⁻¹ kg-sorbent⁻¹), DD- amount of water adsorbed during adsorption stage (mol kg⁻¹)).

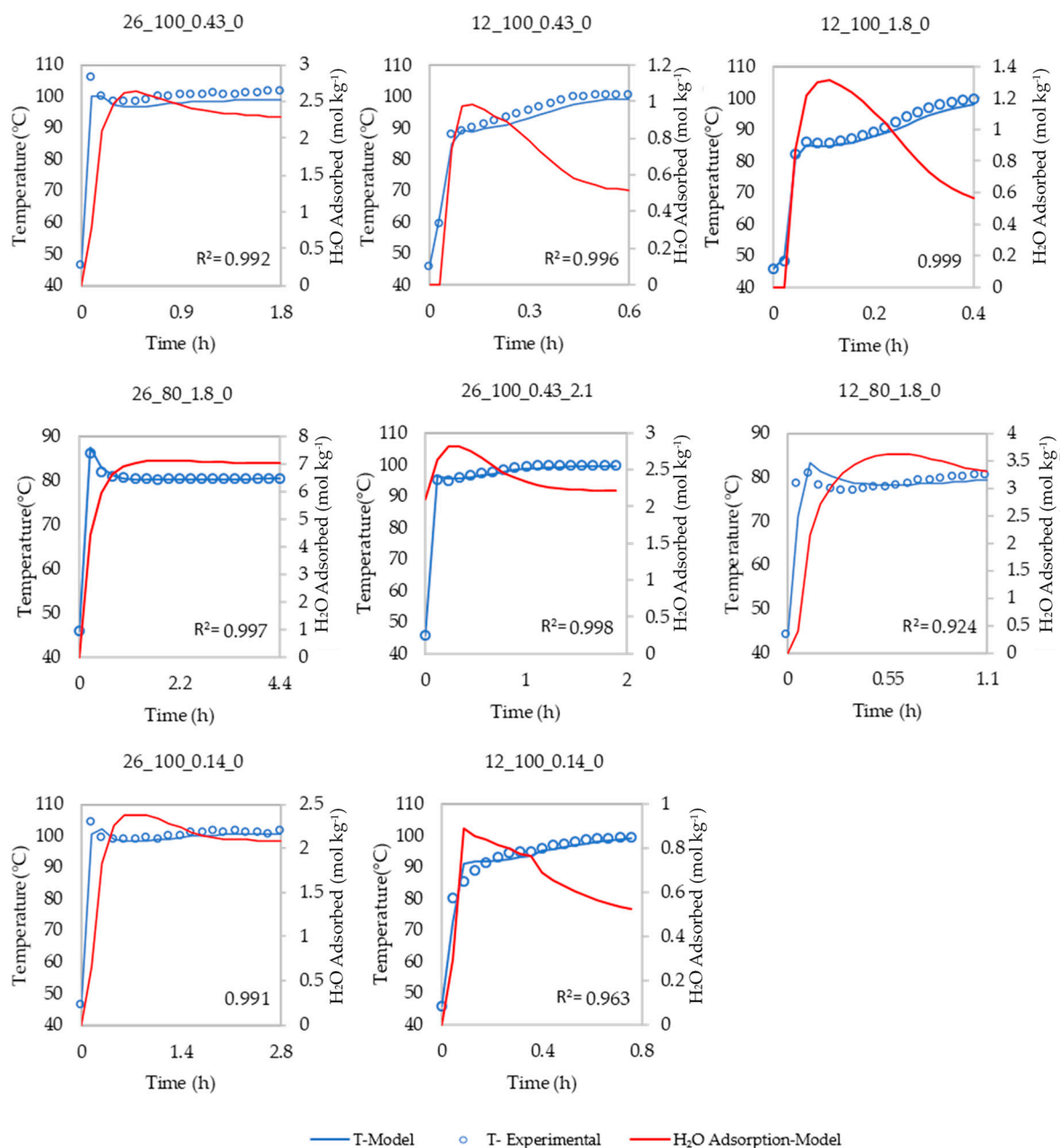


Figure 2. The experimental data and the model predictions for heat transfer and H₂O mass transfer kinetics in the desorption stage. Legend for titles AA_BBB_CCC_DD (AA- desorption pressure(kPa), BBB-desorption temperature (°C), CCC- desorption steam flow rate (kg h⁻¹ kg-sorbent⁻¹), DD- amount of water adsorbed during adsorption stage (mol kg⁻¹)).

S7. Sensitivity of the results to the MOO options

The results of the sensitivity analysis done on the MOO options is presented in Figure S3. ‘base’ refers to the default options used for the results discussed in the study. A to E refers to different cases where the MOO options were varied. The MOO options used for these cases are presented in Table S3 and the variations made are highlighted with bold text. The descriptions for each option is given in Table S4 [7].

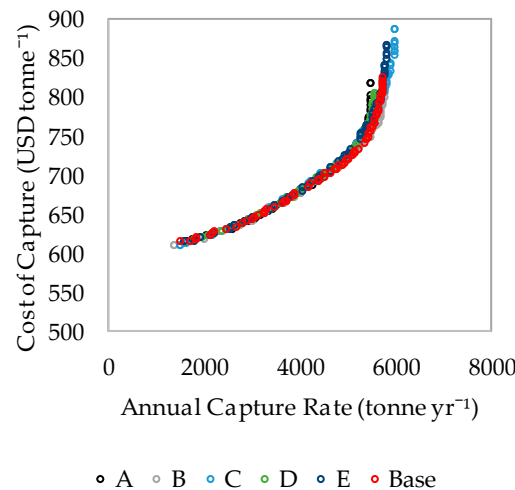


Figure S3. The Pareto plots for MOO carried out with different option. ‘base’ refers to the default options used for the results discussed in the study. The details on the variations made for A to E are given in Table S3.

Table S3. The details of the MOO options used for the sensitivity analysis.

	Base	A	B	C	D	E
Function Tolerance	1e-3	0.5e-3	1e-3	1e-3	1e-3	1e-3
Crossover fraction	0.8	0.8	0.9	0.8	0.8	0.8
Crossover function	1	1	1	0.9	1	1
Mutation	Adaptive feasible	Adaptive feasible	Adaptive feasible	Adaptive feasible	Uniform (0.01)	Uniform (0.05)

Table S4. The description of the MOO options varied for the sensitivity analysis [7].

Option	Description
Function tolerance	If the weighted average relative change in the spread of the Pareto solutions is less than Function tolerance, then the algorithm stops.
Crossover fraction	The fraction of the next generation that crossover produces. Mutation produces the remaining individuals in the next generation.
Crossover function	Creates children by a random weighted average of the parents. Intermediate crossover is controlled by a single parameter, Ratio $child1 = parent1 + random \times Ratio \times (parent2 - parent1)$
Mutation	Mutation functions make small random changes in the individuals in the population, which provide genetic diversity and enable the genetic algorithm to search a broader space -Adaptive Feasible- Randomly generates directions that are adaptive with respect to the last successful or unsuccessful generation. A step length is chosen along each direction so that linear constraints and bounds are satisfied -Uniform-First, the algorithm selects a fraction of the vector entries of an individual for mutation, where each entry has the same probability as the mutation rate of being mutated. In the second step, the algorithm replaces each selected entry by a random number selected uniformly from the range for that entry

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