

Storing Solar Energy with Chemistry: The Role of Thermochemical Storage in Concentrating Solar Power

SUPPLEMENTARY INFORMATION

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S.1 Optimization model

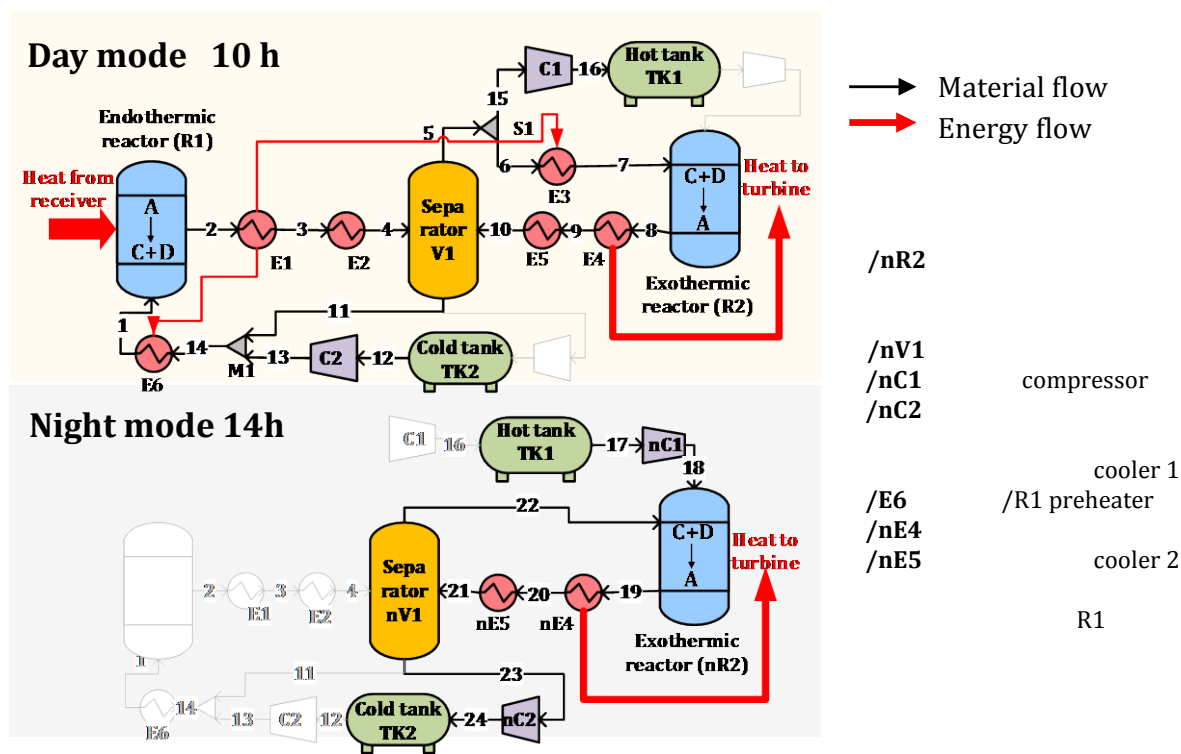


Fig. S1 Process flow diagram of TCES systems with separation

Sets

$i \in \mathbf{I}$	Units
$j \in \mathbf{J}$	Streams
$k \in \mathbf{K}$	Components

Subsets

\mathbf{I}^R	Reaction units {R1, R2, nR2}
\mathbf{I}^{SEP}	Separation units {V1, nV1}
\mathbf{I}^{TK}	Storage units {TK1, TK2}
\mathbf{I}^E	Heat exchangers {E1, E2, E3, E4, E5, E6, nE4, nE5}
\mathbf{I}^C	Compressor {C1, C2, nC1, nC2}
\mathbf{J}_i^{IN}	Inlet streams of unit i
\mathbf{J}_i^{OUT}	Outlet streams of unit i

Parameters

CRF	Capital recovery factor
Cp_k	Specific heat capacity of component k (J/mol-K)
DNI^{ref}	Reference direct normal irradiance (W/m ²)
h^{avg}	Average heat transfer coefficient (W/m ² -K)
MAT	Minimum approach temperature (K)
$prate$	CSP plant rated capacity (MW)
$patm$	Atmosphere pressure (bar)
R	Gas constant (J/mol-K)
ΔH^0	Reaction enthalpy at reference temperature T^0 (kJ/mol)
ΔS^0	Reaction entropy at reference temperature T^0 (J/mol-K)
$d\Delta H$	Rate of reaction enthalpy changing with temperature (kJ/mol-K)
$d\Delta S$	Rate of reaction entropy changing with temperature (J/mol-K ²)
T^0	Reference temperature (K)
t^{day}	Daytime operation time (h)
t^{sto}	Storage time (h)
$z_k(P)$	Compressibility of component k at pressure P
η^c	Collector efficiency
η^{comp}	Compressor motor efficiency
η^{para}	plant parasitic efficiency
λ^c	Collector price (\$/m ² mirror aperture)
λ^r	Receiver price (\$/kW _{thermal})
λ^p	Power block price (\$/kW)
λ^{tk}	Storage tank price (\$/m ³)
λ^{hx}	Heat exchanger price (\$/m ²)
λ^{rxn}	Reactor price (\$/m ³)
λ^{comp}	Compressor price (\$/kW)
λ^{sep}	Separation price (\$/($\frac{mol}{s}$))
λ_k^{sm}	Storage media price of component k (\$/ton)
λ^{cu}	Cooling utility price (\$/kW-year)
λ^{om}	Operation and maintenance (\$/kW-year)
v_k	Stoichiometric coefficient of component k in the reversible reaction
τ^{rxn}	Reactor holding time (s)
ω_k	Molecular weight of component k (g/mol)

Variables

$AREA$	Solar field area (m ²)
$CAPEX$	Plant total capital cost (\$)
C^{cu}	Cooling utility cost (\$/year)
C^{tk}	Storage tank cost (\$)
C^{hx}	Heat exchanger cost (\$)
C^{rxn}	Reactor cost (\$)
C^{comp}	Compressor cost (\$)
C^{sep}	Separation cost (\$)
C^{sto}	TCES system cost (\$)
C^{sm}	TCES storage media cost (\$)
F_j^T	Total molar flow rate in stream j (mol/s)
$F_{j,k}$	Molar flow rate of component k in stream j (mol/s)
$LCOE$	Levelized cost of electricity (¢/kWh)
$OPEX$	Operation cost per year (\$/year)
P	Process pressure (bar)
P_i^H	Maximum storage pressure (bar), $i \in \mathbf{I}^{TK}$
P_i^L	Minimum storage pressure (bar), $i \in \mathbf{I}^{TK}$
PWR_i	Rated power of the compressor for storage tank i (MW), $i \in \mathbf{I}^{TK}$
Q_i	Heat duty of unit i (kW)
T_j	Temperature of stream j (K)
T_i	Temperature of unit i (K) defined as its outlet stream T ($T_i = T_j$, $j = \mathbf{J}_i^{OUT}$)
$\Delta T_{i/i'}$	Arithmetic mean temperature difference (K) of matched heat exchangers i and i'
ΔH_i	Reaction enthalpy at the temperature of reactor i (kJ/mol), $i \in \mathbf{I}^R$
ΔS_i	Reaction entropy at the temperature of reactor i (J/mol-K), $i \in \mathbf{I}^R$
V_i	Volume of storage tank (m ³), $i \in \mathbf{I}^{TK}$
W^{comp}	Total electricity consumed by compressor per day (MWh/day)
W^{ele}	Net electricity generation per day (MWh/day)
\bar{Z}_j	Average compressibility of stream j
$\zeta_{j,k}$	Split fraction of component k in the outlet stream j of the separation unit
η^r	Receiver efficiency
η^{sto}	Storage system round-trip efficiency
$\eta^{p,day}, \eta^{p,night}, \eta^p$	Day, night and overall turbine efficiency
η^{s-e}	Overall solar to electricity efficiency
ξ_j^{spl}	Split fraction for outlet stream of splitter S1

Cost data

Item		Price	Item		Price	Item		Price
Collector	λ^c	250 \$/m ²	Reactor	λ^{rxn}	6500\$/m ³	Ammonia	λ^{sm}	320 \$/ton
Receiver	λ^r	200 \$/kW _t	Separation	λ^{sep}	200\$/($\frac{mol}{s}$)	Methane		180 \$/ton
Turbine	λ^p	1200\$/kW	Heat exchanger	λ^{hx}	50\$/m ³	Carbone dioxide		40 \$/ton
Cooling	λ^{cu}	20\$/kWyr	Pressure vessel	λ^{tk}	5700\$/m ³	Compressor	λ^{comp}	1000 \$/kW
O&M	λ^{om}	65 \$/kWyr	Underground storage		50\$/m ³			

Model Equations

Total molar flow rate
$$F_j^T = \sum_{k \in \mathbf{K}} F_{j,k} \quad j \in \mathbf{J} \quad (\text{S. 1})$$

Mass balance
$$F_{j,k} = F_{j',k} \cdot \xi_j^{spl} \quad i = S1, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}}, k \in \mathbf{K} \quad (\text{S. 2})$$

Mass balance
$$F_{j,k} = \sum_{j' \in \mathbf{J}_i^{\text{IN}}} F_{j',k} \quad i = M1, j \in \mathbf{J}_i^{\text{OUT}}, k \in \mathbf{K} \quad (\text{S. 3})$$

Mass balance
$$F_{j,k} = F_{j',k} \quad i \in \mathbf{I}^{\text{E}} \cup \mathbf{I}^{\text{C}}, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}}, k \in \mathbf{K} \quad (\text{S. 4})$$

Mass balance
$$F_{j,k} = F_{j',k} \cdot \quad i \in \mathbf{I}^{\text{SEP}}, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}}, k \in \mathbf{K} \quad (\text{S. 5})$$

Mass balance
$$F_{j',k} \cdot t^{day} = F_{j,k} \cdot t^{sto} \quad i \in \mathbf{I}^{\text{STO}}, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}}, k \in \mathbf{K} \quad (\text{S. 6})$$

Mass balance
$$(F_{j,A} - F_{j',A}) \cdot v_k = (F_{j,k} - F_{j',k}) \cdot v_A \quad (\text{S. 7})$$

$$i \in \mathbf{I}^{\text{R}}, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}}, k = \text{B, C, D}$$

Reaction enthalpy
$$\Delta H_i = \Delta H^0 + (T_i - T^0) \cdot d\Delta H, i \in \mathbf{I}^{\text{R}} \quad (\text{S. 8})$$

Reaction entropy
$$\Delta S_i = \Delta S^0 + (T_i - T^0) \cdot d\Delta H, i \in \mathbf{I}^{\text{R}} \quad (\text{S. 9})$$

Reaction equilibrium
$$-RT \sum_{k \in \mathbf{K}} v_k \left[\ln(F_{j,k}) - \ln(F_j^T) + \ln\left(\frac{P}{P_{atm}}\right) \right] = \Delta H_i - T_i \Delta S_i \quad (\text{S. 10})$$

$$i \in \mathbf{I}^{\text{R}}, j \in \mathbf{J}_i^{\text{OUT}}$$

Unit heat duty
$$Q_i = \sum_{j \in \mathbf{J}_i^{\text{OUT}}} T_j \cdot \left(\sum_{k \in \mathbf{K}} F_{j,k} \cdot Cp_k \right) - \sum_{j' \in \mathbf{J}_i^{\text{IN}}} T_{j'} \cdot \left(\sum_{k \in \mathbf{K}} F_{j',k} \cdot Cp_k \right) \quad (\text{S. 11})$$

$$i \in \mathbf{I} \setminus \mathbf{I}^{\text{R}}$$

Unit heat duty
$$Q_i = \Delta H_i \cdot (F_{j',A} - F_{j,A}) + (T_j - T_{j'}) \cdot \left(\sum_k F_{j',k} \cdot Cp_k \right) \quad (\text{S. 12})$$

$$i \in \mathbf{I}^{\text{R}}, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}}$$

Constant T units
$$T_{j,k} = T_{j',k} \quad i \in \mathbf{I}^{\text{STO}} \cup \{S1, M1\}, j \in \mathbf{J}_i^{\text{OUT}}, j' \in \mathbf{J}_i^{\text{IN}} \quad (\text{S. 13})$$

No heat duty units
$$Q_i = 0 \quad i \in \mathbf{I}^{\text{SEP}} \cup \{S1, M1\} \quad (\text{S. 14})$$

Heat integration
$$-Q_{E1} = Q_{E3} + Q_{E6} \quad (\text{S. 15})$$

Minimum approach temperature
$$T_2 \geq T_7 + MAT; T_3 \geq T_6 + MAT; \quad (\text{E1-E3}) \quad (\text{S. 16})$$

$$T_2 \geq T_1 + MAT; T_3 \geq T_{14} + MAT; \quad (\text{E1-E6})$$

Mean Temperature Difference
$$\Delta T_{E1/E3} = (T_2 - T_7 + T_3 - T_6)/2 \quad (\text{S. 17})$$

$$\Delta T_{E1/E6} = (T_2 - T_1 + T_3 - T_{14})/2$$

Storage tank size
$$V_i^{tank} \cdot \frac{P_i^H - P_i^L}{R \cdot T} = \bar{Z}_j \cdot F_{j'}^T \cdot t^{day} \quad i \in \mathbf{I}^{\text{STO}}, j' \in \mathbf{J}_i^{\text{IN}} \quad (\text{S. 18})$$

Max power for charging

$$PWR_i \geq \bar{Z}_j \cdot F_{j'}^T \cdot T \cdot \left[\left(\frac{P_i^H}{P} \right)^{1.3} - 1 \right] / \eta^{comp} \quad i \in \mathbf{I}^{STO}, j' \in \mathbf{J}_i^{IN} \quad (S. 19)$$

Max power for discharging

$$PWR_i \geq \bar{Z}_j \cdot F_j^T \cdot T \cdot \left[\left(\frac{P}{P_i^L} \right)^{1.3} - 1 \right] / \eta^{comp} \quad i \in \mathbf{I}^{STO}, j \in \mathbf{J}_i^{OUT} \quad (S. 20)$$

Daily electricity for charging

$$W_i^{in} = \bar{Z}_j \cdot \left\{ \frac{\sum_k (F_{j',k} C p_k) \cdot V_i^{tank \cdot P}}{1.3} \cdot \left[\left(\frac{P_i^H}{P} \right)^{1.3} - 1 \right] - \bar{Z}_j \cdot \sum_k (F_{j',k} C p_k) \cdot V_i^{tank} \cdot (P_i^H - P) \right\} / (F_{j'}^T \cdot R \cdot \eta^{comp}) \quad i \in \mathbf{I}^{STO}, j' \in \mathbf{J}_i^{IN} \quad (S. 21)$$

Daily electricity for discharging

$$W_i^{out} = \bar{Z}_j \cdot \left\{ \frac{\sum_k (F_{j,k} C p_k) \cdot V_i^{tank \cdot P}}{0.7} \left[1 - \left(\frac{P_i^L}{P} \right)^{0.7} \right] - \bar{Z}_j \cdot \sum_k F_{j,k} C p_k \cdot V_i^{tank} \cdot (P - P_i^L) \right\} / (F_j^T \cdot R \cdot \eta^{comp}) \quad i \in \mathbf{I}^{STO}, j \in \mathbf{J}_i^{OUT} \quad (S. 22)$$

Total daily electricity consumption

$$W^{comp} = \sum_{i \in \mathbf{I}^{STO}} (W_i^{in} + W_i^{out}) \quad (S. 23)$$

Compression T

$$T_{j,k} = T_{j',k} \quad i = \mathbf{I}^C, j \in \mathbf{J}_i^{OUT}, j' \in \mathbf{J}_i^{IN} \quad (S. 24)$$

Power block efficiency day and night

$$\eta^{p,day} = 1 - \sqrt{330/T_{R2}}, \eta^{p,night} = 1 - \sqrt{330/T_{nR2}} \quad (S. 25)$$

Output in the day

$$-(Q_{R2} + Q_{E4}) \cdot \eta^{p,day} \cdot \eta^{para} = p^{rate} \quad (S. 26)$$

Output at night

$$-(Q_{nR2} + Q_{nE4}) \cdot \eta^{p,night} \cdot \eta^{para} = p^{rate} \quad (S. 27)$$

Solar field area

$$AREA = Q_{R1} / (DNI^{ref} \cdot \eta^c \cdot \eta^r) \quad (S. 28)$$

Receiver efficiency

$$\eta^r = -0.02T^{*3} - 0.08T^{*2} - 0.18T^* + 0.78 \quad (S. 29)$$

$$T^* = (T_{R1} - 1150) / 245$$

Average power block efficiency

$$\eta^p = \frac{Q_{r2} \cdot t^{day} \cdot \eta^{p,day} + Q_{nr2} \cdot t^{sto} \cdot \eta^{p,night}}{Q_{r2} \cdot t^{day} + Q_{nr2} \cdot t^{sto}} \quad (S. 30)$$

TES system efficiency

$$\eta^{sto} = \frac{Q_{E4} \cdot t^{day} + Q_{nE4} \cdot t^{sto} - \frac{W^{comp}}{\eta^{para} \cdot \eta^p}}{Q_{R1} \cdot t^{day}} \quad (S. 31)$$

Solar to electricity efficiency

$$\eta^{s-e} = \eta^c \cdot \eta^r \cdot \eta^{sto} \cdot \eta^p \cdot \eta^{para} \quad (S. 32)$$

Compressor cost

$$C^{comp} = \lambda^{comp} \cdot \sum_{i \in \mathbf{I}^{STO}} PWR_i \quad (S. 33)$$

Separation cost

$$C^{sep} \geq \lambda^{sep} \cdot \sum_{j' \in \mathbf{J}_i^{IN}} F_{j'}^T \quad i = V1, nV1 \quad (S. 34)$$

Heat exchanger cost

$$C^{hx} = \lambda^{hx} \sum_{i=E3,E6} \frac{Q_i}{\Delta T_i \cdot h^{avg}} \quad (S. 35)$$

Reactor cost (R2)

$$C^{rxn} \geq \lambda^{rxn} F_j^T RT_i \tau^{rxn} / P \quad i = R2, nR2; j \in \mathbf{J}_i^{OUT} \quad (S. 36)$$

Storage tank cost $C^{tk} = \sum_{i \in I^{sto}} \lambda_i^{tk} \cdot (P_i^H / 200)^{0.44} \cdot V_i^{tank}$, aboveground storage (S. 37)

$C^{tk} = \sum_{i \in I^{sto}} \lambda_i^{under} \cdot V_i^{tank}$, underground storage

Storage media cost $C^{sm} = t^{sto} \cdot \sum_k F_{17,k} \cdot \lambda_k^{sm} \cdot \omega_k$ (S. 38)

Storage system cost $C^{sto} = C^{sm} + C^{tk} + C^{rxn} + C^{hx} + C^{comp} + C^{sep}$ (S. 39)

Capital cost $CAPEX = C^{sto} + \lambda^{sf} \cdot AREA + \lambda^r \cdot Q_{R1} / \eta^r + \lambda^p \cdot prate / \eta^{para}$ (S. 40)

Cooling utility cost $C^{cu} = \lambda^{cu} [(Q_{E2} + Q_{E5}) \cdot t^{day} + Q_{NE5} \cdot t^{sto}] / 24$ (S. 41)

Annual Operation cost $OPEX = C^{cu} + prate \cdot \lambda^{om}$ (S. 42)

Daily net electricity output $W^{ele} = prate \cdot (t^{day} + t^{sto}) - W^{comp}$ (S. 43)

LCOE calculation $LCOE = (CAPEX \cdot CFR + OPEX) / (365 \cdot W^{ele})$ (S. 44)

S.2 Separation surrogate model

In the ammonia TCES system, a flash tank is used to separate unreacted ammonia from hydrogen and nitrogen (see Fig. 5(a)). To model the flash tank, we derive the relation of ammonia liquid phase recovery (ζ_{NH_3}) by fitting the simulation data from ASPEN Plus.

Through ASPEN Plus simulation, 66 sample points are generated by varying the feed stream ammonia molar fraction (x_{NH_3}) and flash tank temperature (T_{flash}) between 0.1-0.6 and 10-60°C respectively. Using the sample points, the function $\zeta_{NH_3} = f(x_{NH_3}, T_{flash})$ is fitted through polynomial regression in Matlab. Before fitting, variables are scaled and centered using standard normalization ($x^* = (x - \bar{x}) / \sigma$). Different polynomial order combinations are tried to decide a relatively simple and accurate fitting. Fig.S2 shows the sample points and the fitting surface using the fitting function described as Eqn. (S.47).

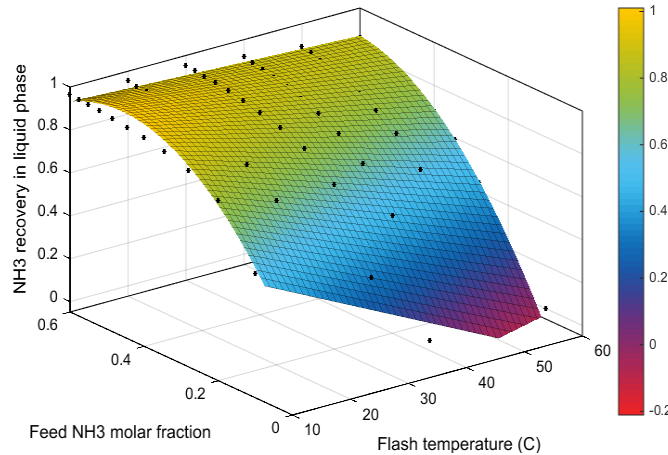


Fig. S2 Sample points and fitting surface of ζ_{NH_3}

$$\zeta_{NH_3} = 0.84 - 0.095 T^* + 0.15 x^* + 0.06 T^* x^* - 0.09 x^{*2} \quad (S. 45)$$

S.3 Detailed optimization results (Case 1)

Table S1. Stream temperature (T) and component molar flow rate (F)

Stream #	1	2	3	4	5	6	7	8	9	10	11	12
T(°C)	251	386	27	17	17	17	349	539	80	17	17	17
F_{NH_3} (mol/s)	34445	21956	21956	21956	1777	836	836	3034	3034	3034	23214	11231
F_{H_2} (mol/s)	392	19124	19124	19124	29319	13789	13789	10491	10491	10491	296	96
F_{N_2} (mol/s)	196	6440	6440	6440	9894	4653	4653	3554	3554	3554	100	96

Stream #	13	14	15	16	17	18	19	20	21	22	23	24
T(°C)	17	17	17	17	17	17	399	80	17	17	17	17
F_{NH_3} (mol/s)	11231	34445	941	941	672	672	8636	8636	8636	614	8022	8022
F_{H_2} (mol/s)	96	392	15530	15530	11093	11093	6844	6844	6844	6775	68	68
F_{N_2} (mol/s)	96	196	5241	5241	3743	3743	6844	6844	6844	6775	68	68

Table S2. Unit temperature, pressure, heat duty and work load (day mode)

Unit	R1	R2	TK1	TK2	V1	C1	C2	S1	M1
Temperature (°C)	386	539	17	17	1	17	1	17	17
Pressure (bar)	300	300	115-564	188-393	300	300-564	188-300	300	300
Heat duty (MW)	1005	0	0	0	0	0	0	0	0
Work (MWh/day)	0	0	0	0	0	192	32	0	0

Unit	E1	E2	E3	E4	E5	E6
Temperature (°C)	386-27	27-17	17-349	539-80	80-17	17-251
Pressure (bar)	300	300	300	300	300	300
Heat duty (MW)	-968	-25	216	-310	-43	753
Work (MWh/day)	0	0	0	0	0	0

Table S3. Unit temperature, pressure, heat duty and work load (night mode)

Unit	nR2	nV1	nC1	nC2	nE4	nE5
Temperature (°C)	399	17	17	17	399-80	80-17
Pressure (bar)	300	300	115-300	300-393	300	300
Heat duty (MW)	0	0	0	0	-370	-72
Work (MWh/day)	0	0	140	34	0	0

S.4 Geological map for underground storage

There are three types of underground storage facilities: depleted gas reservoirs, aquifers, and mined salt caverns. ¹ Fig. S3 and Fig. S4 shows the potential geologic underground storage areas around the world and in the United States.

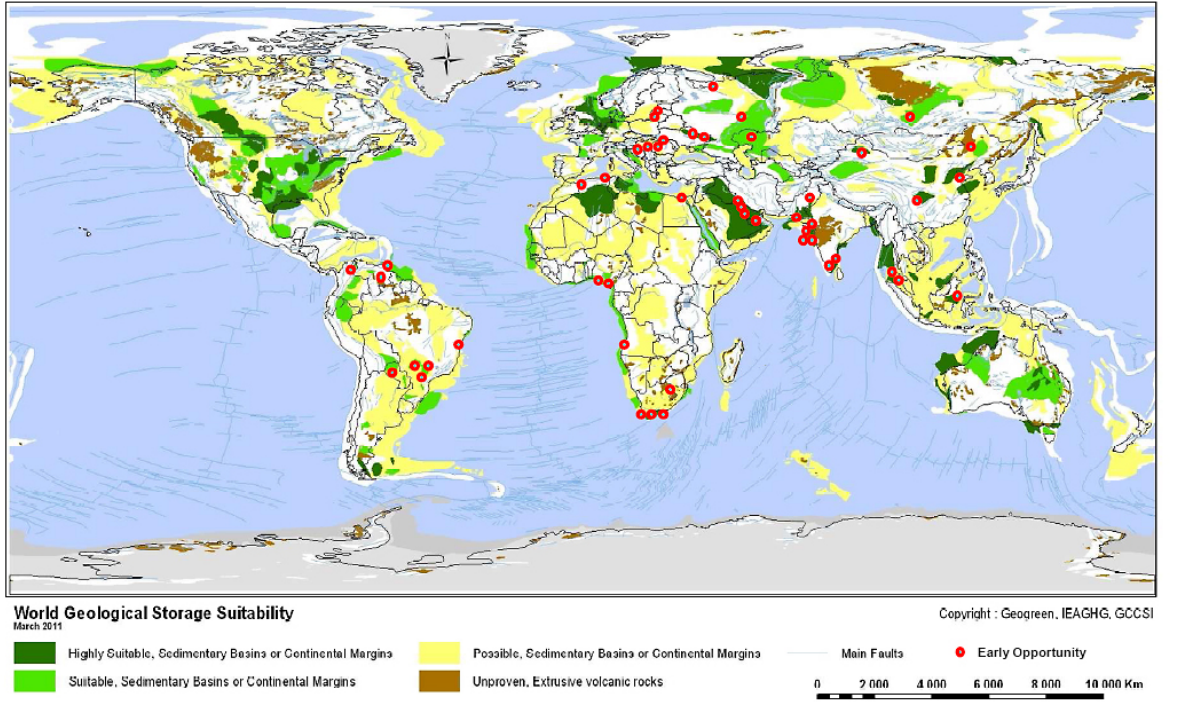


Fig. S3 Safe locations for underground carbon dioxide storage ²



Fig. S4 Potential geologic underground storage areas in the United States ³

References

- (1) Federal energy regulatory commission. *Current State of and Issues Concerning Underground Natural Gas Storage*; 2004.
- (2) Co-Firing EPA-Outlawed Coal Power Plants with Carbon Capture
<http://stoppingclimatechange.com/co-firing.htm> (accessed Dec 29, 2016).
- (3) Lord, A. S. *Overview of Geologic Storage of Natural Gas with an Emphasis on Assessing the Feasibility of Storing Hydrogen.*; 2009.

S.5 Temperature dependent heat capacity

Fig. S5 shows how the heat capacities of NH_3 , H_2 and N_2 change with temperature at 300 bar. We note that ammonia has a significant higher heat capacity between 80-300 °C due to the continuous phase change. To account for the latent heat, we divide the temperature into 3 stages: cold (10-80 °C), warm (80-300 °C) and hot (>300 °C). For each component k , different average heat capacities are used for each temperature stage (denoted by Cp_k^{cold} , Cp_k^{warm} and Cp_k^{hot}).

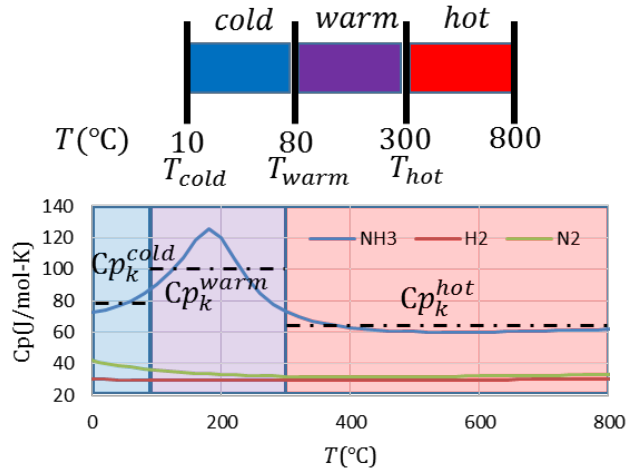


Fig. S5 Heat capacities of NH_3 , H_2 and N_2 at 300 bar

We then classify each process stream j into either cold stream (J^{cold}), warm stream (J^{warm}) or hot stream (J^{hot}) based on the stream temperature range (see Fig. S6).

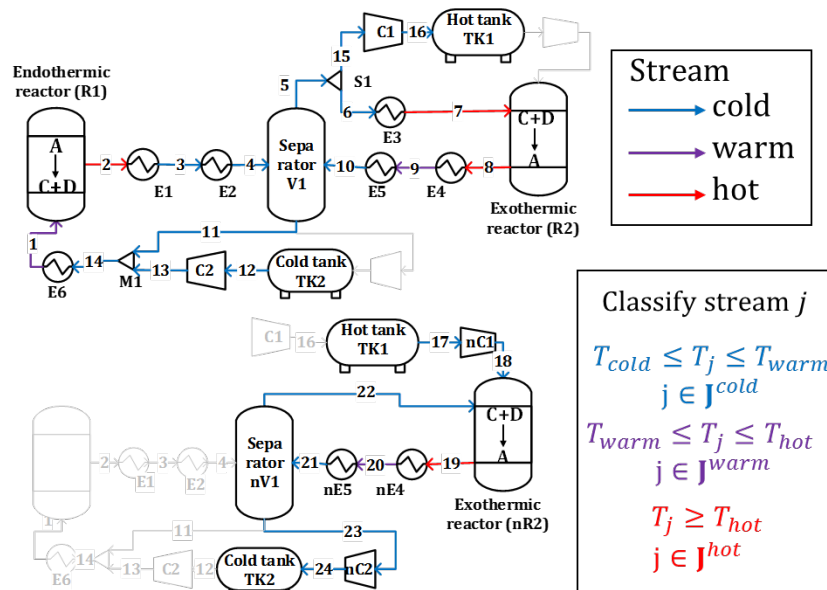


Fig. S6 Classify process streams into cold, warm and hot stream.

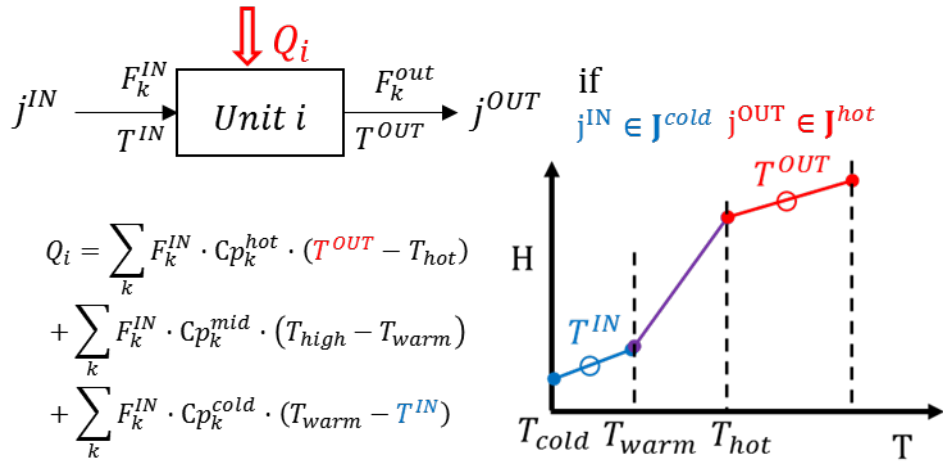


Fig. S7 An example of unit heat duty calculation.

For a general unit i , the temperature stage of its inlet and outlet streams are known from the stream classification. Accordingly, the unit heat duty calculation equation is derived. Fig. S7 illustrates the heat duty calculation for a unit with cold inlet stream and hot outlet stream.

S.6 Compressibility factor

In our TCES systems, gas products are compressed and stored at high pressure (150-550 bar). To account for the real gas behavior, we introduce compressibility factors for the calculation of compression work and storage tank volumes. Fig. S8 shows the compressibility factors of all gas products at 100 °C (obtained from ASPEN PLUS property database using Schwartzentruber-Renon equation-of-state). Based on these data, we can calculate the stream average compressibility factor as a function of stream pressure and composition.

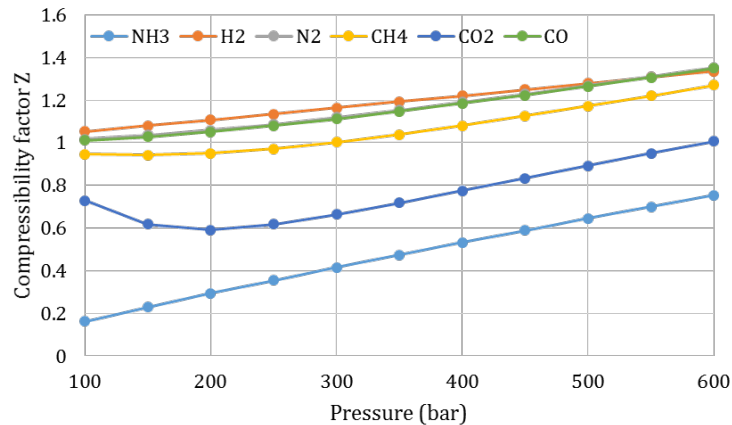
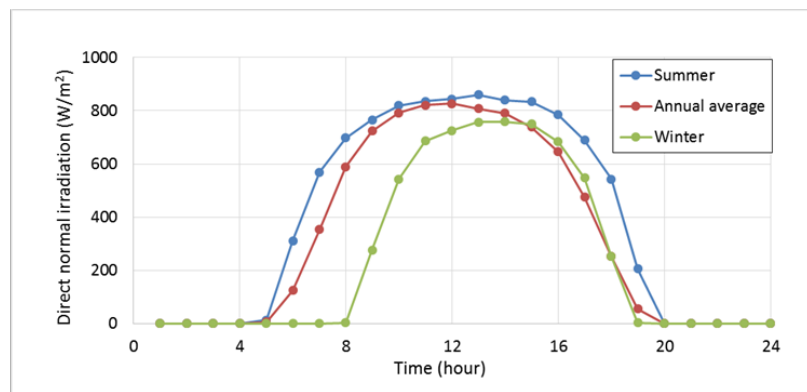


Fig. S8 Compressibility factor of different components at 100 °C

S.7 Seasonal variation of solar irradiation

Fig. S9 shows the solar irradiation profile over one day and the performance of our system on an annual average day, typical summer and winter day respectively. Our plant is sized to support a 24-hour 100 MW turbine output under average sunlight condition. On a summer day, when the day time is longer, the turbine can easily maintain the rated output and the compression power is slightly lower since only 12 hours of storage are now needed. Around 15% solar collection is curtailed through heliostat defocus, which is what is done in practice. On a winter day, when less solar irradiation is available, the turbine adjusts to a lower output level, 77 MW to maintain continuous operation. Although storage for more hours is needed, less compression work is required due to the reduced turbine output level. Thus, sizing based on an *average day* is reasonable: it results in moderate collection curtailments during summer days and lower turbine output levels during winter days. Sizing for a hot summer day would to unused capacity (and therefore large capital normalized costs) while sizing based on a winter day would lead to a very restrictive design.



	Day hours	DNI (W/m ²)	Collection curtailment	Turbine output (MW)	Compression (MW)	Grid output (MW)
Design point	10	800	0	100	17	83
Summer	12	748	15%	100	16	84
Winter	8	800	0	77	12	65

Fig. S9 Solar resource and CSP plant performance on an annual average, summer and winter day.