

SUPPORTING INFORMATION

Supplementary information S1

S1.1: The detailed simulation of ammonia production from syngas HYSYS Documentation

The decision variables in this case are:

Reformer temperature: 900°C

Combuster temperature: 1400°C

Low temperature conversion reactor temperature (LTC): 160°C

Fluid package: Peng-Robinson

Component: Methane, Ethane, Propane, i-Butane, n-Butane, Nitrogen, CO, CO₂, H₂O,

Oxygen, Hydrogen, Ammonia, Methanol

Reactions:

Set-1

Rxn-1 Type: Equilibrium	Stoich Coeff
-------------------------	--------------

Methane	-1
---------	----

H ₂ O	-1
------------------	----

CO	1
----	---

Hydrogen	3
----------	---

Rxn-2 Type: Equilibrium	Stoich Coeff
-------------------------	--------------

Ethane	-1
--------	----

H ₂ O	-2
------------------	----

CO	2
----	---

Hydrogen	5
----------	---

Rxn-3 Type: Equilibrium	Stoich Coeff
-------------------------	--------------

Propane	-1
H ₂ O	-3
CO	3
Hydrogen	7
Rxn-4 Type: Equilibrium	Stoich Coeff
i-Butane	-1
H ₂ O	-4
CO	4
Hydrogen	9
Rxn-5 Type: Equilibrium	Stoich Coeff
n-Butane	-1
H ₂ O	-4
CO	4
Hydrogen	9

Set-2

Rxn-1 Type: Equilibrium	Stoich Coeff
CO	-2
Oxygen	-1
CO ₂	2
Rxn-2 Type: Equilibrium	Stoich Coeff
Hydrogen	-2
Oxygen	-1
H ₂ O	2
Rxn-3 Type: Equilibrium	Stoich Coeff
Methane	-1

Oxygen	-2
H ₂ O	2
CO ₂	1
Rxn-4 Type: Equilibrium	Stoich Coeff
Ethane	-2
Oxygen	-7
H ₂ O	6
CO ₂	4
Rxn-5 Type: Equilibrium	Stoich Coeff
Propane	-1
Oxygen	-5
H ₂ O	4
CO ₂	3

Set-3

Rxn-1 Type: Equilibrium	Stoich Coeff
CO	-1
Hydrogen	-3
Methane	1
H ₂ O	1
Rxn-2 Type: Equilibrium	Stoich Coeff
CO ₂	-1
Hydrogen	-4
Methane	1
H ₂ O	2

Set-4

Rxn-1 Type: Kinetic	Stoich Coeff
Ammonia	2
Hydrogen	-3
Nitrogen	-1

Set-5

Rxn-1 Type: Equilibrium	Stoich Coeff
CO	-1
H ₂ O	-1
CO ₂	1
Hydrogen	1

Streams:

HPSteam

Composition	Mole fraction
H ₂ O	1.0000

Temperature = 350 °C

Pressure = 120 bar

NaturalGas

Composition	Mole fraction
Methane	0.9575
Ethane	0.0267
Propane	0.0065
i-Butane	0.0010
n-Butane	0.0011
Nitrogen	0.0072

Temperature = 25 °C

Pressure = 35 bar

Air

Composition	Mole fraction
-------------	---------------

Nitrogen	0.7900
----------	--------

Oxygen	0.2100
--------	--------

Temperature = 800 °C

Pressure = 35 bar

Reformer_Feed (decision variable)

Temperature = 900 °C

Combuster_Feed (decision variable)

Temperature = 1400 °C

W_in_1

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 25 °C

Pressure = 1 bar

W_out_1

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 80 °C

Pressure = 1 bar

WSGR1_Feed

Temperature = 450 °C

W_in_2

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 25 °C

Pressure = 1 bar

W_out_2

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 80 °C

Pressure = 1 bar

WSGR2_Feed (decision variable)

Temperature = 160 °C

W_in_3

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 25 °C

Pressure = 1 bar

W_out_3

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 80 °C

Pressure = 1 bar

Separator1_Feed

Temperature = 100 °C

Methanizer_Feed

Temperature = 340 °C

W_in_5

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 25 °C

Pressure = 1 bar

W_out_5

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 80 °C

Pressure = 1 bar

Separator2_Feed

Temperature = 40 °C

Make-up N2

Composition	Mole fraction
-------------	---------------

Nitrogen	1.0000
----------	--------

Temperature = 0 °C

Pressure = 140 bar

Molar Flow = 1000 kgmole/hr

ComP1_out

Pressure = 80 bar

W_in_6

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 25 °C

Pressure = 1 bar

W_out_6

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 80 °C

Pressure = 1 bar

Cool2_out

Temperature = 90 °C

Comp2_out

Pressure = 150 bar

W_in_7

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 25 °C

Pressure = 1 bar

W_out_7

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 80 °C

Pressure = 1 bar

Separator3_Feed

Temperature = 40 °C

Feed_1

Temperature = 450 °C

W_in_9

Composition Mole fraction

H₂O 1.0000

Temperature = 25 °C

Pressure = 1 bar

W_out_9

Composition Mole fraction

H₂O 1.0000

Temperature = 80 °C

Pressure = 1 bar

Product_separator

Vapor-Phase Fraction = 1.0000

Recycle

Temperature = 20 °C

LN2_in

Composition Mole fraction

Nitrogen 1.0000

Temperature = -196.2 °C

Pressure = 1 bar

LN2_out

Composition Mole fraction

Nitrogen 1.0000

Temperature = -80 °C

Pressure = 1 bar

Liquid_to_separator

Temperature = -45 °C

Vessel:

Reformer

Reaction Set: Set-1

Combuster

Reaction Set: Set-2

HTC

Reaction Set: Set-5

LTC

Reaction Set: Set-5

Methanizer

Reaction Set: Set-3

For all plug flow reactors

Reaction Set: Set-4

Heater:

E-100

Inlet Stream: Feed

Outlet: Reformer_Feed

Delta P: 0 bar

E-113

Inlet Stream: Heated_re_pro

Outlet: Combuster_Feed

Delta P: 0 bar

E-104

Inlet Stream: SynGas_Flow

Outlet: Methanizer_Feed

Delta P: 0 bar

Heat Exchanger:

For all heat exchangers:

Heat Exchanger Model: Simple Weighted

Tube Side Delta P = 0 bar

Shell Side Delta P = 0 bar

Mixer:

MIX-100

Feed

Composition	Mole fraction
Methane	0.2522
Ethane	0.0070
Propane	0.0017
i-Butane	0.0003
n-Butane	0.0003
Nitrogen	0.0019
H ₂ O	0.7366

MIX-101

Heated_re_pro

Composition	Mole fraction
Methane	0.0326
Nitrogen	0.1192
CO	0.1294
H ₂ O	0.3066

Oxygen	0.0314
--------	--------

Hydrogen	0.3809
----------	--------

MIX-102

Feed_ammonia

Composition	Mole fraction
-------------	---------------

Methane	0.0147
---------	--------

Nitrogen	0.6462
----------	--------

H ₂ O	0.0005
------------------	--------

Hydrogen	0.3319
----------	--------

Ammonia	0.0066
---------	--------

MIX-103

32

Composition	Mole fraction
-------------	---------------

Methane	0.0012
---------	--------

Nitrogen	0.2585
----------	--------

H ₂ O	0.0027
------------------	--------

Hydrogen	0.7376
----------	--------

MIX-104

Feed_2

Composition	Mole fraction
-------------	---------------

Methane	0.0189
---------	--------

Nitrogen	0.6882
----------	--------

H ₂ O	0.0006
------------------	--------

Ammonia	0.2922
---------	--------

MIX-105

Feed_3

Composition	Mole fraction
Methane	0.0190
Nitrogen	0.6887
H ₂ O	0.0006
Ammonia	0.2917

TEE:

TEE-100

Split Fraction

Quench1 = 0.9950

Quench2 = 0.0025

Quench3 = 0.0025

TEE-101

Split Fraction

Purge = 0.0100

Product to liquid = 0.9900

Recycle:

RCY-3

Inlet Stream: N2_to_Recycle

Outlet Stream: N2_recycle

S1.2: The detailed simulation of methanol production with a recycle HYSYS Documentation

The decision variables in this case are:

Pressure of the equilibrium reactor (ERV-100): 70 bar

Temperature of the equilibrium reactor (ERV-100): 190 °C

Temperature of a separator (E-101): 60 °C

Recycle ratio (RCY-1): 1

Fluid package: Peng Robinson

Component: CO₂, Hydrogen, Methanol, CO, H₂O

Reactions:

Set-1

Rxn-1 Type: Equilibrium	Stoich Coeff
-------------------------	--------------

CO ₂	-1
-----------------	----

H ₂ O	1
------------------	---

Methanol	1
----------	---

Hydrogen	-3
----------	----

Rxn-2 Type: Equilibrium	Stoich Coeff
-------------------------	--------------

CO	-1
----	----

Methanol	1
----------	---

Hydrogen	-2
----------	----

Rxn-3 Type: Equilibrium	Stoich Coeff
-------------------------	--------------

CO ₂	-1
-----------------	----

CO	1
----	---

Methanol	1
----------	---

Hydrogen	-1
----------	----

Streams:

Hydrogen

Composition	Mole fraction
-------------	---------------

Hydrogen	1.0000
----------	--------

Temperature = 25 °C

Pressure = 20 bar

Carbon dioxide

Composition	Mole fraction
-------------	---------------

CO ₂	1.0000
-----------------	--------

Temperature = 40 °C

Pressure = 20 bar

Compressed feed (decision variable)

Pressure = 70 bar

Heated feed (decision variable)

Temperature = 190 °C

Water_in

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 20 °C

Pressure = 1 bar

Water_out

Composition	Mole fraction
-------------	---------------

H ₂ O	1.0000
------------------	--------

Temperature = 35 °C

Pressure = 1 bar

Cooled product (decision variable)

Temperature = 60 °C

Vessel:

ERV-100

Reaction Set: Set-1

Towers:

T-100

Number of stages: 10

Inlet Stream

Stream: Cooled product (decision variable)

Inlet Stage: 5_Main Tower

Condenser: Full Reflux

Condenser Pressure = 4 bar

Condenser Delta P = 0 bar

Reboiler Pressure = 4 bar

Reboiler Delta P = 0 bar

Specifications:

Condenser Temperature = 25 °C

Reflux Ratio = 0.6

T-101

Number of stages: 10

Inlet Stream

Stream: Methanol-water

Inlet Stage: 5_Main Tower

Condenser: Total

Condenser Pressure = 4 bar

Condenser Delta P = 0 bar

Reboiler Pressure = 4 bar

Reboiler Delta P = 0 bar

Specifications:

Comp Fraction

Stage: Condenser

Flow Basis: Mole Fraction

Phase: Liquid

Spec Value: 0.9950

Component: Methanol

Reflux Ratio = 30

Heater:

Inlet Stream: Mixed feed

Outlet: Heated feed

Delta P: 0 bar

Heat Exchanger:

For all heat exchangers:

Heat Exchanger Model: Simple Weighted

Tube Side Delta P = 0 bar

Shell Side Delta P = 0 bar

Mixer:

MIX-100

Feed

Composition

Mole fraction

Hydrogen	0.7500
----------	--------

CO ₂	0.2500
-----------------	--------

MIX-101

Mixed feed

Composition	Mole fraction
-------------	---------------

Hydrogen	0.8053
----------	--------

CO ₂	0.1886
-----------------	--------

CO	0.0013
----	--------

Methanol	0.0039
----------	--------

H ₂ O	0.0009
------------------	--------

MIX-102

Combined product

Composition	Mole fraction
-------------	---------------

Hydrogen	0.0025
----------	--------

CO ₂	0.0154
-----------------	--------

CO	0.0000
----	--------

Methanol	0.4911
----------	--------

H ₂ O	0.4910
------------------	--------

TEE:

TEE-100

Split Fraction

To recycle = 1.00

To mixer = 0.00

Recycle:

RCY-1

Inlet Stream: To recycle

Outlet Stream: Recycle

Valves:

VLV-101

Inlet stream: To mixer

Outlet stream:

Pressure = 4 bar

VLV-102

Inlet stream: Liquid_product

Outlet stream

Pressure = 4 bar

S1.3: The detailed simulation of CO₂ absorption by methanol via Rectisol

process HYSYS Documentation

The decision variables in this case are:

Lean methanol temperature: -25.0°C

The 3rd stage separator pressure: 1.2 bar

Stripper reflux ratio: 5

Stripper inlet temperature: 40°C

Distillation reflux ratio: 1

Fluid package: PRSV

Component: Methanol, CO₂, Ammonia, CO, Argon, Nitrogen, Hydrogen, H₂O, Methane

Streams:

Syngas Feed

Composition

Mole fraction

CO ₂	0.4590
Ammonia	0.0001
CO	0.0052
Argon	0.0085
Nitrogen	0.4923
Hydrogen	0.0272
H ₂ O	0.0027
Methane	0.0050

Temperature = 18.30 °C

Pressure = 17.24 bar

Lean Methanol

Composition	Mole fraction
Methanol	0.9998
H ₂ O	0.0002

Temperature (decision variable) = -25 °C

Pressure = 17.20 bar

5F (decision variable)

Temperature = 40 °C

Methanol

Composition	Mole fraction
Methanol	1.0000

Temperature = 21 °C

Pressure = 1.1 bar

NH-3

Composition	Mole fraction
-------------	---------------

Ammonia 1.0000

Temperature = -70 °C

Pressure = 1 bar

NH-4

Composition Mole fraction

Ammonia 1.0000

Temperature = -50 °C

Pressure = 1 bar

Towers:

Absorber

Number of stages: 15

Inlet Stream

Stream: 5B-3

Inlet Stage: 15_Main Tower

Top Stage Inlet: Lean Methanol

Stripper

Number of stages: 10

Inlet Stream

Stream: 5F (decision variable)

Inlet Stage: 10_Main Tower

Stream: 3C-3 (decision variable)

Inlet Stage: 11_Main Tower

Condenser: Full Reflux

Condenser Pressure = 1.1 bar

Condenser Delta P = 0 bar

Reboiler Pressure = 1.1 bar

Reboiler Delta P = 0 bar

Specifications:

Comp Ratio = 0.9500

Reflux Ratio (decision variable) = 5

Distillation

Number of stages: 12

Inlet Stream

Stream: 2

Inlet Stage: 5_Main Tower

Condenser: Full Reflux

Condenser Pressure = 3.4 bar

Condenser Delta P = 0 bar

Reboiler Pressure = 3.4 bar

Reboiler Delta P = 0 bar

Specifications:

Comp Ratio = 0.99

Reflux Ratio (decision variable) = 1

Heat Exchanger:

For all heat exchangers:

Heat Exchanger Model: Simple Weighted

Tube Side Delta P = 0 bar

Shell Side Delta P = 0 bar

Mixer:

MIX-100

6A-7

Composition	Mole fraction
Methanol	0.9999
H ₂ O	0.0001

MIX-101

1B

Composition	Mole fraction
Methanol	0.1862
CO ₂	0.2004
Ammonia	0.0002
CO	0.0036
Argon	0.0041
Nitrogen	0.3376
Hydrogen	0.2593
H ₂ O	0.0029
Methane	0.0059

MIX-102

4H

Composition	Mole fraction
Methanol	0.0146
CO ₂	0.9476
Ammonia	0.0008
CO	0.0001
Argon	0.0006
Nitrogen	0.0344

Hydrogen 0.0001

Methane 0.0019

TEE:

TEE-100

Split Fraction

$$6C = 0.9400$$

$$6D-1 = 0.0600$$

Valve:

VLV-100

Inlet stream: 3A

Outlet stream: 2

$$\text{Pressure} = 3.4 \text{ bar}$$

VLV-101

Inlet stream: 5C

Outlet stream: 5C-2

$$\text{Pressure} = 4.5 \text{ bar}$$

VLV-102

Inlet stream: 5A

Outlet stream: 5A-2

$$\text{Pressure} = 10.5 \text{ bar}$$

VLV-103

Inlet stream: 5D

Outlet stream: 5D-2

$$\text{Pressure} = 4.5 \text{ bar (decision variable)}$$

Recycle:

RCY-1

Inlet Stream: 5B

Outlet Stream: 5B-2

RCY-2

Inlet Stream: 6D-1

Outlet Stream: 6D

RCY-3

Inlet Stream: 6E-1

Outlet Stream: Lean Methanol

RCY-4

Inlet Stream: 3C-2

Outlet Stream: 3C

Supplementary information S2

S2.1: Final data set of Case study I: ammonia production from syngas

Sample no.	Decision variables			Response
	x_1	x_2	x_3	Ammonia production cost
1	1153.47	1522.43	190.06	576.03
2	989.12	1467.24	281.62	556.63
3	1164.99	1672.82	254.31	648.00
4	1124.42	1567.96	202.96	588.94
5	922.86	1406.41	205.68	505.25
6	1048.96	1547.33	264.13	585.45
7	1037.08	1505.91	167.29	549.42
8	1016.11	1603.51	162.43	583.59
9	976.84	1690.36	196.72	621.42
10	1095.54	1455.63	223.08	548.77
11	933.05	1417.36	173.66	506.26
12	906.22	1653.35	228.98	607.52
13	1189.02	1576.66	287.31	632.67
14	1180.29	1593.89	219.19	611.34
15	971.01	1620.26	245.53	599.96
16	1135.40	1495.11	270.62	583.39
17	1007.10	1665.00	271.47	633.36
18	1061.01	1536.71	248.98	577.83
19	1100.68	1481.80	184.73	552.14
20	955.61	1628.75	211.49	595.98
21	1074.02	1433.58	234.38	538.25
22	1121.60	1567.94	201.16	588.01
23	1114.70	1564.07	198.40	585.01
24	900.00	1432.03	160.00	507.48
25	1007.20	1666.77	275.99	636.49
26	1006.64	1659.79	272.91	631.79
27	900.00	1400.00	183.04	497.62
28	996.40	1468.40	283.43	559.07
29	990.83	1459.03	278.79	552.75
30	900.00	1400.00	177.05	497.13

S2.2: Final data set of Case study II: methanol production via carbon dioxide

hydrogenation

Sample no.	Decision variables				Response
	X ₁	X ₂	X ₃	X ₄	Methanol production cost
1	68.43	201.98	62.00	0.6046	1618.57
2	57.86	201.15	65.57	0.1721	2898.02
3	55.86	193.07	70.48	0.1395	2760.07
4	53.27	191.74	79.37	0.4834	2001.59
5	58.77	194.46	74.66	0.9282	1090.09
6	57.50	207.15	67.57	0.5387	2084.13
7	52.28	208.60	67.01	0.0128	4381.01
8	59.75	202.47	77.17	0.0714	3133.65
9	52.09	202.88	78.28	0.8561	1330.76
10	63.86	194.20	72.25	0.4568	1836.09
11	50.06	200.68	64.56	0.3766	2663.30
12	63.43	206.21	61.16	0.8169	1329.13
13	66.12	196.73	76.52	0.3568	2020.91
14	54.46	209.62	71.58	0.5097	2337.28
15	60.69	190.78	78.95	0.7016	1416.77
16	62.18	196.04	60.13	0.8650	1182.66
17	50.88	198.26	70.79	0.3058	2758.26
18	61.03	190.33	66.12	0.6630	1475.46
19	64.87	203.78	76.19	0.9625	1042.72
20	53.93	199.99	63.15	0.1054	3290.48
21	66.77	198.71	69.36	0.6396	1534.88
22	69.29	207.04	68.78	0.4074	2073.98
23	67.25	208.23	75.44	0.2771	2465.39
24	61.46	205.53	73.76	0.2200	2757.53
25	56.66	195.45	63.60	0.7194	1493.37
26	65.43	197.38	68.18	0.9656	1023.04
27	55.05	204.88	62.47	0.1968	3153.04
28	68.94	192.66	73.35	0.7778	1239.14
29	52.70	209.22	66.89	0.0189	4355.37
30	52.44	208.65	66.68	0.0089	4386.29
31	70.00	190.00	80.00	1.0000	942.45
32	56.17	193.42	70.66	0.1289	2784.44
33	55.83	193.07	70.06	0.1238	2801.24
34	68.41	204.98	60.00	1.0000	1002.55
35	55.54	204.82	62.81	0.2136	3063.39
36	55.20	204.34	62.14	0.1939	3127.63
37	70.00	190.00	70.96	1.0000	950.13
38	52.02	203.43	78.36	0.8399	1375.90
39	51.43	203.04	78.12	0.8220	1423.52

40	70.00	207.99	76.01	1.0000	989.75
41	52.46	208.82	66.99	0.0167	4359.64
42	52.27	208.64	66.90	0.0026	4435.38
43	65.91	191.32	80.00	1.0000	952.13
44	68.48	201.93	61.83	0.6047	1617.03
45	68.11	201.56	61.67	0.5767	1667.13
46	67.24	194.77	60.00	1.0000	972.79

S2.3: Final data set of Case study III: CO₂ absorption by methanol via Rectisol process

Sample no.	Decision variables					Response
	X ₁	X ₂	X ₃	X ₄	X ₅	Capture cost
1	-49.23	1.229	8.771	16.72	6.136	112.46
2	-23.43	1.606	6.422	22.06	9.323	117.24
3	-46.37	1.479	16.311	26.65	9.631	176.00
4	-52.99	1.939	5.777	26.26	4.232	98.36
5	-43.91	1.304	9.989	27.30	5.244	105.41
6	-35.83	1.734	11.425	33.87	6.573	122.39
7	-29.26	1.774	16.645	29.45	5.769	136.68
8	-20.29	1.630	5.076	29.81	3.585	72.66
9	-25.61	1.724	15.599	31.89	9.765	154.00
10	-33.49	1.407	14.172	14.41	2.664	102.97
11	-26.56	1.499	14.933	37.13	1.862	90.03
12	-47.20	1.701	12.639	11.87	8.579	163.88
13	-36.66	1.753	15.806	38.20	7.604	151.40
14	-51.74	1.209	8.156	21.18	2.375	85.44
15	-21.51	1.571	19.422	28.41	2.183	115.72
16	-45.17	1.360	11.636	23.43	4.487	112.75
17	-54.88	1.318	6.779	32.88	8.186	117.28
18	-27.94	1.908	17.157	25.36	5.035	139.27
19	-28.85	1.545	10.494	34.67	6.920	109.08
20	-40.91	1.453	13.837	13.33	1.080	102.48
21	-41.45	1.405	19.922	31.09	6.322	156.90
22	-22.28	1.876	8.943	19.14	2.952	90.87
23	-37.87	1.639	9.499	15.73	8.247	129.13
24	-39.25	1.285	10.769	35.07	7.702	116.51
25	-32.00	1.967	18.855	24.06	3.465	145.87
26	-50.41	1.909	14.659	17.47	5.441	169.91
27	-31.20	1.999	7.858	14.24	4.605	100.30
28	-48.86	1.858	17.927	39.68	6.867	189.42
29	-30.46	1.260	6.016	20.01	3.300	70.23
30	-42.96	1.542	13.111	18.21	3.837	121.05
31	-53.77	1.450	13.157	10.62	8.903	170.07
32	-38.17	1.811	7.449	20.61	7.288	114.72
33	-44.51	1.678	12.252	36.21	9.039	151.20
34	-34.99	1.339	18.361	11.32	1.331	110.01
35	-24.48	1.821	17.773	38.30	1.602	110.28
36	-48.02	1.858	18.145	39.71	6.791	188.79
37	-48.63	1.844	17.895	39.53	6.679	187.08
38	-31.21	1.200	5.000	33.95	1.000	46.35
39	-28.21	1.560	10.533	34.84	7.057	110.19
40	-28.71	1.549	10.237	34.10	7.031	109.09

41	-20.00	1.200	5.000	23.67	1.000	52.59
42	-54.08	1.337	6.945	33.02	8.083	117.89
43	-54.86	1.322	6.803	32.38	8.018	116.86
44	-35.74	1.200	5.000	33.89	1.000	47.83
45	-34.31	1.351	18.645	11.61	1.513	112.37
46	-34.72	1.346	18.324	10.94	1.279	110.24
47	-20.00	1.281	5.000	39.31	1.000	46.12
48	-40.07	1.474	13.826	13.09	1.129	102.81
49	-40.84	1.455	13.619	12.96	1.042	99.59
50	-26.47	1.280	5.000	40.00	1.000	43.95
51	-24.38	1.829	17.729	38.81	1.694	111.06
52	-24.87	1.827	17.530	38.38	1.617	109.80
53	-37.16	1.511	5.000	40.00	1.000	50.04

Supplementary information S3

S3.1 Weights and biases of an artificial neural networks for training the data set of random sampling.

S3.1.1 Case study I: ammonia production from syngas

Mean square errors:

Data set	MSE
Training	1.52E-05
Validation	1.19E-04
Testing	4.52E-05

Weight 1, $WI_{r,k}$

Neurons	Input nodes		
	1	2	3
1	-2.3682	0.6228	0.5254
2	-0.1803	-0.0498	-1.3409
3	0.1440	0.2659	-0.0401
4	-1.4446	1.7557	0.5200
5	-0.1177	0.1200	0.4391
6	-1.3274	0.0407	-1.3057
7	-0.2760	-1.7613	0.0743
8	2.0677	-1.8151	-0.6159

Weight 2, $W2_r$

Neurons	Output node
1	-0.0416
2	-0.3629
3	2.3181
4	0.0745
5	0.8902
6	-0.0186
7	-0.0540
8	-0.1241

Bias 1, BI_r

Neurons	Output node
1	2.3922
2	1.3427
3	-0.2083
4	-0.3426
5	1.4267
6	-0.1934
7	-1.8252
8	2.5504

Bias 2, B2	0.0129
------------	--------

S3.1.2 Case study II: methanol production via carbon dioxide hydrogenation

Mean square errors:

Data set	MSE
Training	3.20E-07
Validation	8.24E-06
Testing	4.61E-06

Weight 1, $WI_{r,k}$

Neurons	Input nodes			
	1	2	3	4
1	0.9948	0.6365	-0.1825	-0.1942
2	0.3067	0.2540	-0.2935	-2.6807
3	-0.9732	1.3545	1.0360	-1.0357
4	0.7542	-0.6835	0.0090	-0.7380
5	0.0794	-0.0215	-0.0181	0.7138
6	1.0944	-0.9190	0.0708	-0.6277
7	-0.2468	0.2019	-0.0045	-0.4551
8	-1.1972	0.5411	-0.0324	-1.2129

Weight 2, $W2_r$

Neurons	Output node
1	0.0225
2	0.0272
3	-0.0055
4	0.0792
5	-0.3989
6	0.1319
7	2.1486
8	0.2470

Bias 1, BI_r

Neurons	Output node
1	-0.8868
2	2.1283
3	1.4075
4	0.2115
5	-0.1466
6	1.9636
7	-1.0273
8	-2.7433

Bias 2, B2	1.3462
------------	--------

S3.1.3 Case study III: CO₂ absorption by methanol via Rectisol process

Mean square errors:

Data set	MSE
Training	2.85E-05
Validation	5.50E-05
Testing	4.22E-05

Weight 1, $Wl_{r,k}$

Neurons	Input nodes				
	1	2	3	4	5
1	-1.1612	0.4267	0.6170	0.0776	-0.0070
2	-2.0083	-0.0265	-0.0583	0.0438	-0.1383
3	0.0076	0.0483	-0.0250	0.0230	-0.1016
4	-0.1773	0.2575	0.0805	0.0104	-0.0002
5	2.2137	0.0507	0.0991	-0.0537	0.1723
6	0.1480	-0.3700	0.1323	-0.0025	-0.0004
7	-0.6150	0.1493	-0.2343	0.0470	0.0128
8	-1.7773	0.5226	2.2852	0.3430	-0.1023

Weight 2, $W2_r$

Neurons	Output node
1	0.3551
2	-0.8543
3	-3.6878
4	2.6417
5	-0.6012
6	1.1263
7	-1.2372
8	-0.0256

Bias 1, Bl_r

Neurons	Output node
1	-1.5419
2	1.4949
3	-0.0233
4	-0.2838
5	-1.4584
6	0.6507
7	-1.7367
8	-2.5763

Bias 2, B2	-0.9123
------------	---------

S3.2 Weights and biases of an artificial neural networks for training the data set of adaptive Latin hypercube sampling.

S3.2.1 Case study I: ammonia production from syngas

Mean square errors:

Data set	MSE
Training	9.92E-07
Validation	2.85E-04
Testing	6.46E-04

Weight 1, $WI_{r,k}$

Neurons	Input nodes		
	1	2	3
1	0.7357	-1.3657	2.1587
2	-2.0546	-0.9905	-1.7479
3	2.0853	0.3863	1.8296
4	1.6385	2.1145	0.0074
5	1.9052	-1.9173	-1.3953
6	1.7018	-2.2192	-0.4386
7	1.0337	1.9477	-0.0395
8	-2.0165	1.2170	0.9853

Weight 2, $W2_r$

Neurons	Output node
1	-0.0577
2	-0.2594
3	-0.0703
4	0.3227
5	-0.1809
6	-0.0355
7	0.4293
8	0.2925

Bias 1, BI_r

Neurons	Output node
1	-2.9187
2	2.0097
3	-1.4948
4	-0.6331
5	-0.1269
6	1.0142
7	2.1234
8	-3.1478

Bias 2, B2	0.1989
------------	--------

S3.2.2 Case study II: methanol production via carbon dioxide hydrogenation

Mean square errors:

Data set	MSE
Training	3.06E-08
Validation	1.29E-04
Testing	1.02E-05

Weight 1, $WI_{r,k}$

Neurons	Input nodes			
	1	2	3	4
1	-0.3066	-1.5063	-0.9558	0.7309
2	1.0728	-0.4369	0.0888	-1.6636
3	0.0529	-0.0215	0.0344	1.5948
4	0.4170	-1.7917	-0.8440	-1.9836
5	1.4311	-1.3899	1.1017	0.9473
6	-0.2295	0.2196	0.0183	-0.9931
7	2.1562	-0.3368	-0.8320	1.2068
8	-0.9796	0.7576	-0.3045	-1.2917

Weight 2, $W2_r$

Neurons	Output node
1	-0.0626
2	-0.0206
3	-0.2198
4	0.0139
5	-0.0210
6	0.7670
7	-0.0847
8	0.6752

Bias 1, BI_r

Neurons	Output node
1	-2.9615
2	-0.4146
3	-0.9196
4	0.5101
5	1.2467
6	-0.7789
7	3.0020
8	-3.3325

Bias 2, B2	0.6054
------------	--------

S3.2.3 Case study III: CO₂ absorption by methanol via Rectisol process

Mean square errors:

Data set	MSE
Training	1.32E-04
Validation	4.27E-04
Testing	2.33E-03

Weight 1, $Wl_{r,k}$

Neurons	Input nodes				
	1	2	3	4	5
1	-2.0167	0.7220	-0.6211	-0.7957	-1.0699
2	-0.0506	0.3918	-0.0145	-1.3910	1.2934
3	-1.0740	0.0579	1.1195	-0.2202	-0.1242
4	0.4088	-0.3947	0.7916	0.9237	-1.1863
5	1.7848	-0.5047	-0.3678	1.4842	1.2589
6	0.2225	0.4808	0.9969	-0.0013	0.7223
7	0.3278	0.1813	0.4486	1.1427	-0.6860
8	-1.0223	0.9647	1.4222	0.1476	0.2411

Weight 2, $W2_r$

Neurons	Output node
1	-0.3738
2	0.2561
3	0.4370
4	-0.2361
5	0.1715
6	0.2842
7	0.0372
8	0.3227

Bias 1, Bl_r

Neurons	Output node
1	2.9622
2	-2.1864
3	0.0097
4	-0.7930
5	0.4856
6	0.7373
7	2.5068
8	-1.8019

Bias 2, B2	0.5801
------------	--------