

Electronic Supplementary Information:

Quantitative Analysis of CO₂ Emissions Reduction Potential of Alternative Light Olefins Production Processes

Marian Flores-Granobles and Mark Saeys

S1 Process model assumptions

The following general assumptions were considered for the process models developed in Aspen Plus® (Figures S1-S6):

1. Naphtha is supplied at 25 °C and atmospheric pressure. The composition of naphtha is shown in Table S1.
2. Pure CO₂ is supplied at 40 °C and 130 kPa. The electricity requirements and CO₂ emissions due to CO₂ capture from the CO₂ source and CO₂ purification were not considered in the models.
3. Pure H₂ is produced by water electrolysis using Proton-Exchange-Membrane Electrolyzers (PEME) or Solid Oxide Electrolyzers (SOE). When PEME are used, water is pumped to the required pressure before the electrolysis. When SOE are used, steam is electrolyzed and the produced H₂ is compressed to the required pressure. More details are available in the section "H₂ and syngas production by electrolysis".
4. Pumps, compressors, and turbines operate with 70% of isentropic efficiency.
5. Cooling water was used to cool streams to 35 °C. The energy required for this cooling cycle was not considered in the energy balance.
6. Chilled water was used to cool streams to between 15 °C and 35 °C. This refrigeration cycle uses 0.06 kWh per MJ heat removed.¹
7. Cooling to between -30 °C and 15 °C, to between -90 °C and -30 °C and to between -147 °C and -90 °C required cryogenic refrigeration cycles using C₃H₈, C₂H₄ and CH₄ as cooling media, respectively. The corresponding electricity requirements are calculated using separate Aspen Plus® models.
8. Electric boilers convert electrical energy to heat with 100% efficiency.
9. Membrane separation was used in the CO₂ to olefins (C²O) scenario to recover CO₂ and H₂ from the product stream for recycle to the C²O reactor. A cellulose acetate membrane is modeled in Matlab® using the permeability of the gas components in Table S2. The Matlab® model provides the pressure drop and split ratio per component to the permeate and retentate streams based on the system equations below²:

Variables:

P_1 : Feed gas pressure [bar]

P_2 : Permeate pressure [bar]

δ : membrane thickness [cm]

$\#_{comp}$: number of components [-]

q_i : Permeability of component "i" [Barrer] based on Table S2

x_i : Molar fraction of component "i" in retentate stream [-]

y_i : Molar fraction of component "i" in permeate stream [-]

J_i : Local flux of component "i" to the permeate stream [mol/s]

F_i : Flowrate of component "i" in the feed stream [mol/s]

L_i : Flowrate of component "i" in the retentate stream [mol/s]

V_i : Flowrate of component "i" in the permeate stream [mol/s]

Equations:

$$J_i = 3.35 \times 10^{-9} \cdot q_i \cdot \delta \cdot (P_1 \cdot x_i - P_2 \cdot y_i)$$

$$y_i = \frac{J_i}{\sum_1^{\#_{comp}} J_i}$$

$$F = L + V$$

$$F \cdot x_i^{feed} = L \cdot \bar{x}_i + V \cdot \bar{y}_i$$

10. Amine absorption is used to remove residual CO₂ from the light olefins (C_{2,3}=) stream. A separate Aspen Plus® model for CO₂ capture from a natural gas power plant using 30 wt% monoethanolamine (MEA) developed by Aspentech was used as a base case³. The process parameters were adjusted to limit the CO₂ concentration below 0.5 wt% in the C_{2,3}=-rich stream and to avoid flooding or drying within the stages of the columns. A gas-liquid separation unit was included before the scrubber to separate part of the captured CO₂ by the pressure difference, this reduces the scrubber loading. The resulting energy requirements for this process model are within the boundaries of operation.⁴
11. A polymer-grade ethylene/propylene mixture (> 99.5 wt%)⁵ is produced in each scenario.
12. The minimum energy requirements for each process were evaluated using Aspen Energy Analyzer. Figure S7 shows the Composite curves for the C²O-SOE scenario.

Table S1. Naphtha composition.⁶

Component	Mass fraction
n-butane	0.0216
isobutane	0.0012
n-pentane	0.2734
2-methyl-butane	0.2138
Cyclopentane	0.0358
n-hexane	0.2248
cyclohexane	0.0384
Benzene	0.0158
n-heptane	0.0711
Cycloheptane	0.0434
Cycloheptene	0.0157
n-octane	0.0224
Cyclooctane	0.0092
Cyclooctene	0.0055
n-nonane	0.0079

Table S2. Permeability of gas components for the modeled cellulose acetate membrane.⁷

Component	Permeability (Barrer)
CO ₂	6.3
H ₂	5.46
CO	0.26
CH ₄	0.21
C ₂ H ₄	0.45
C ₃ H ₆	0.46
C ₄ H ₁₀	0.18

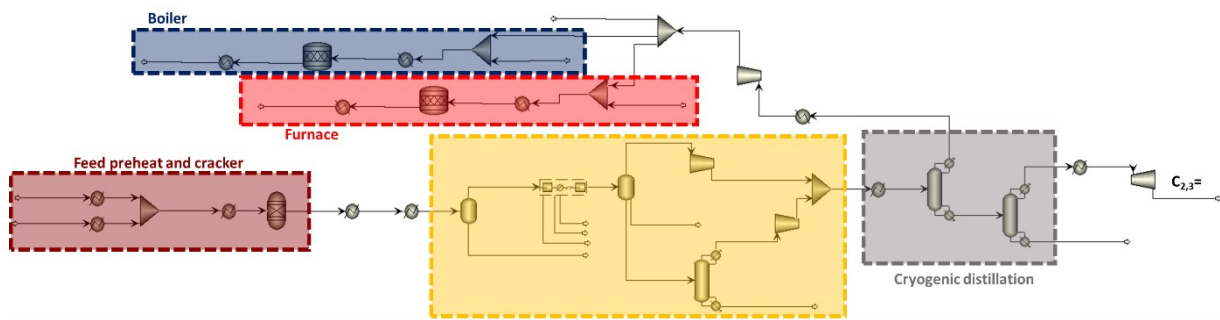


Figure S1. Process model for NSC and RDR

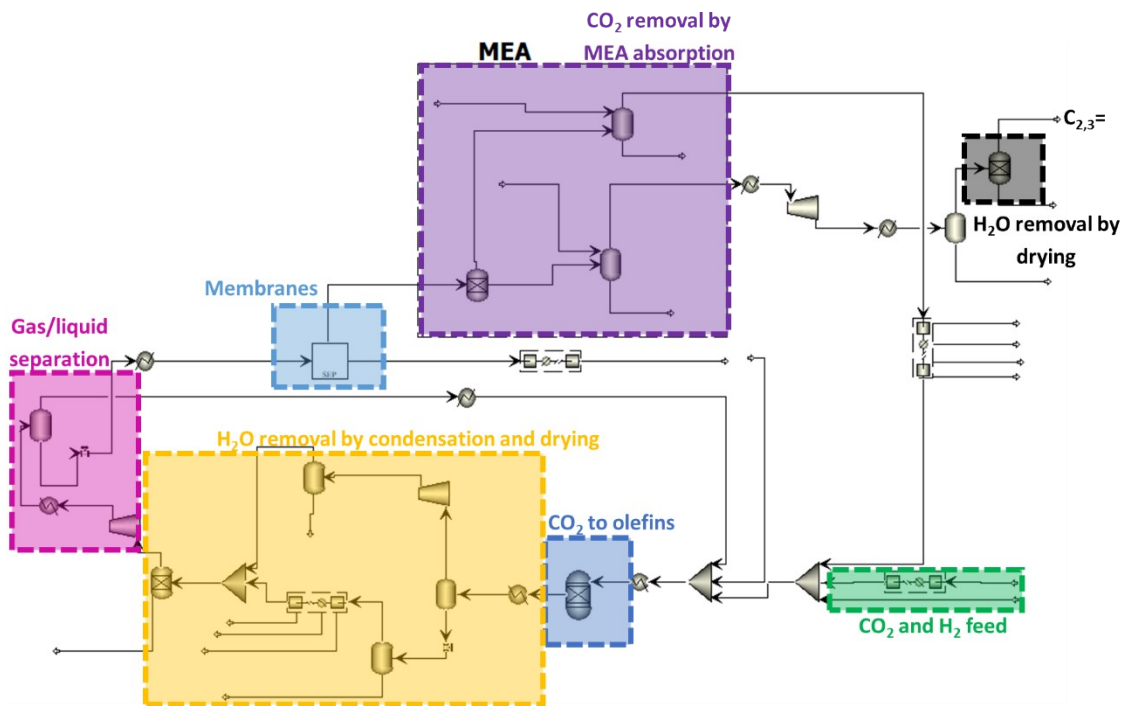


Figure S2. Process model for C^2O

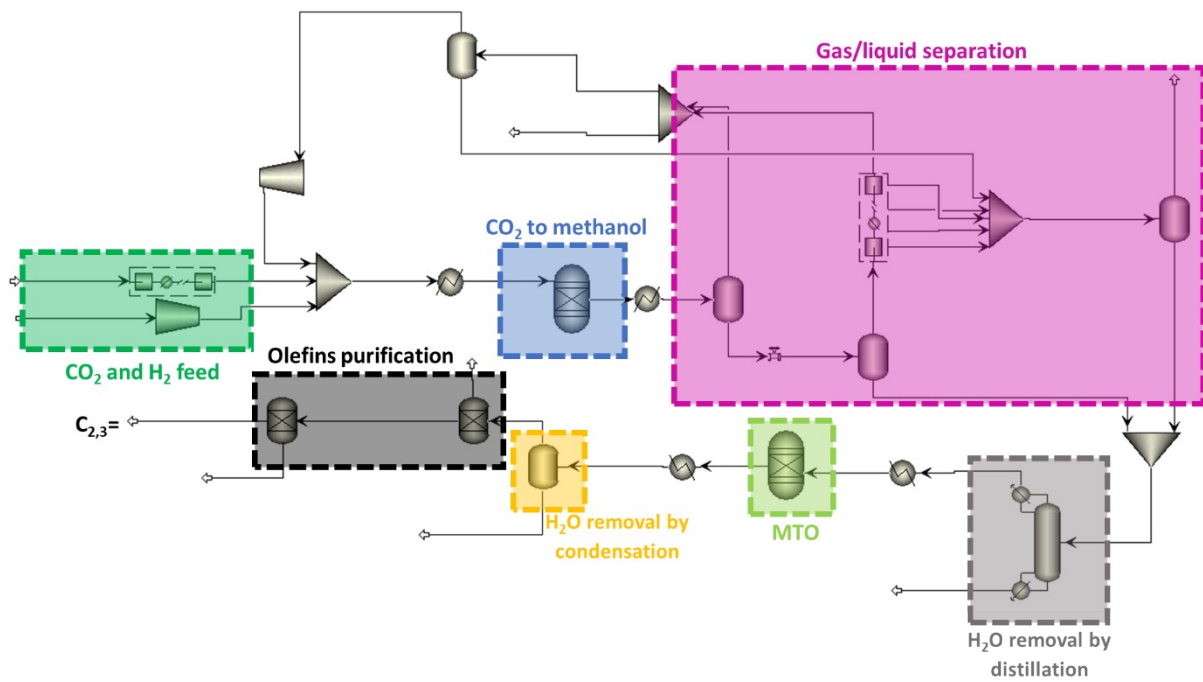


Figure S3. Process model for C²M+MTO

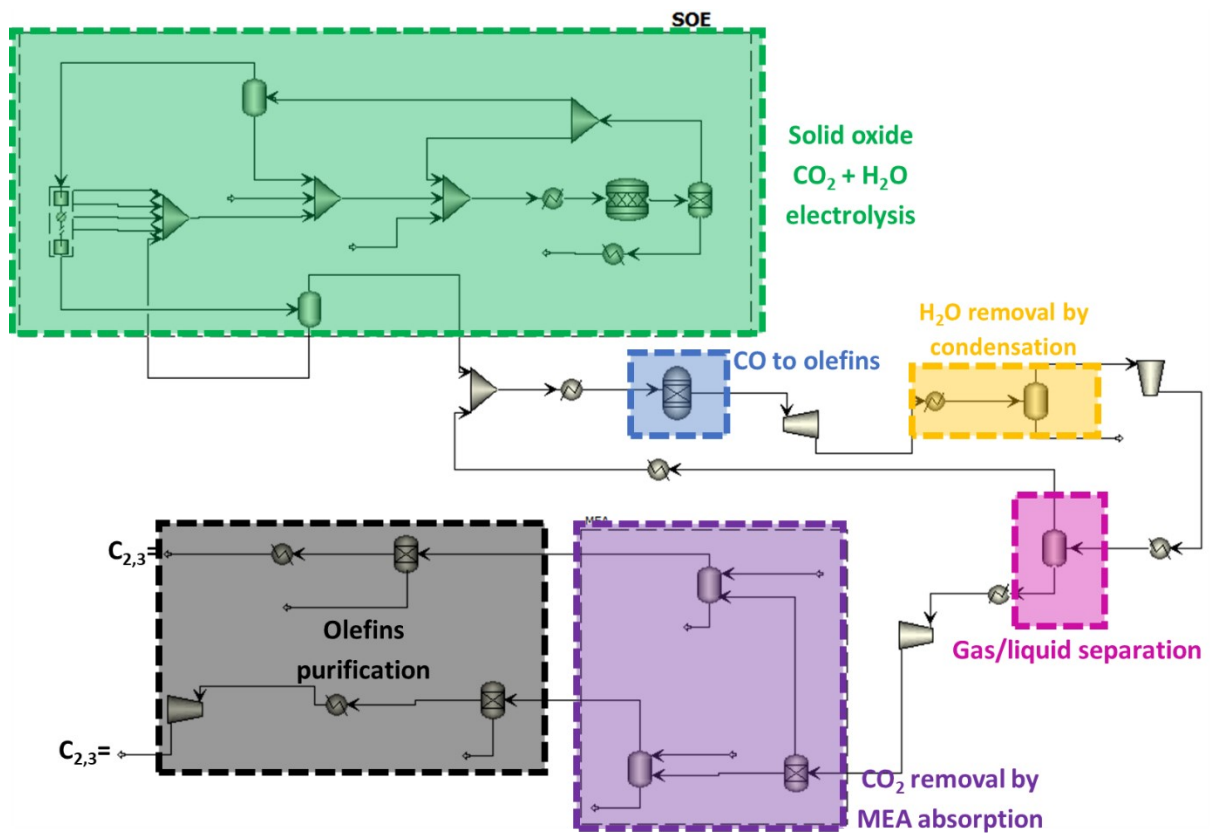


Figure S4. Process model for CO₂red+COhyd

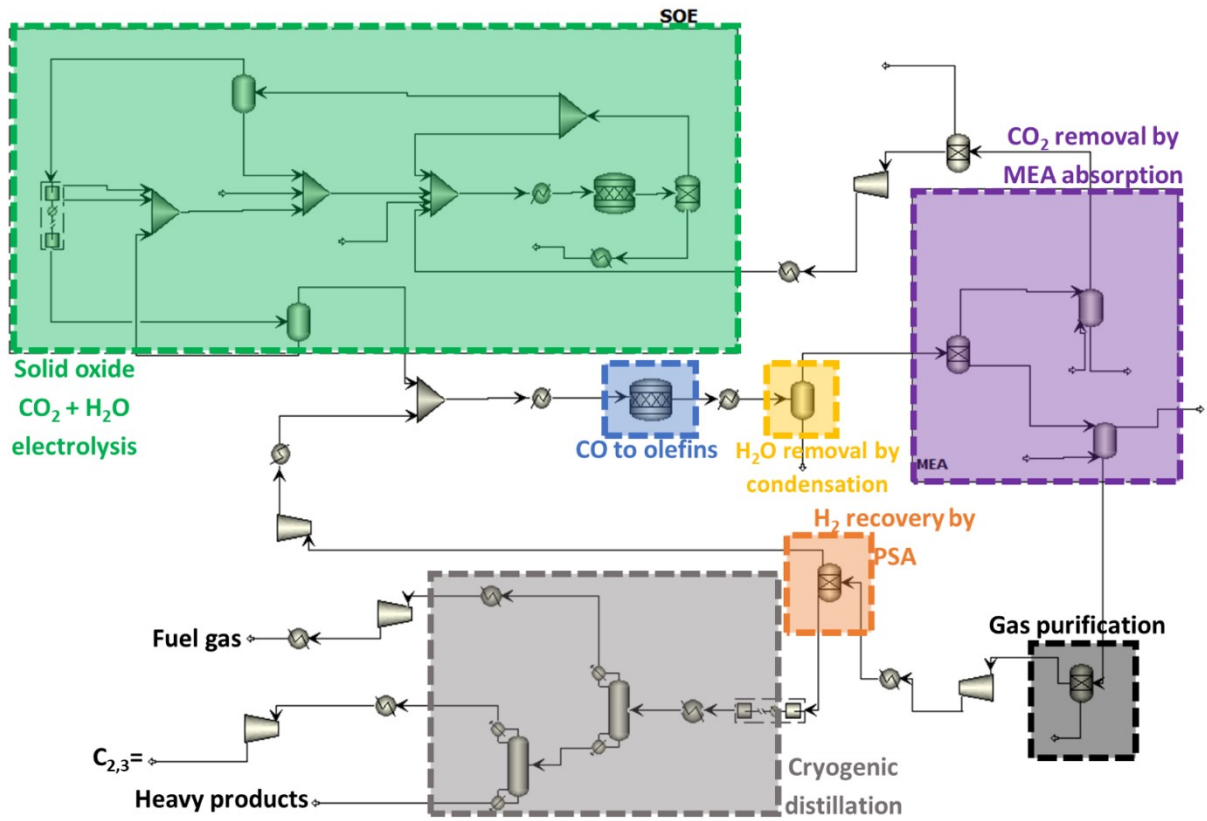


Figure S5. Process model for CO₂red+FTO

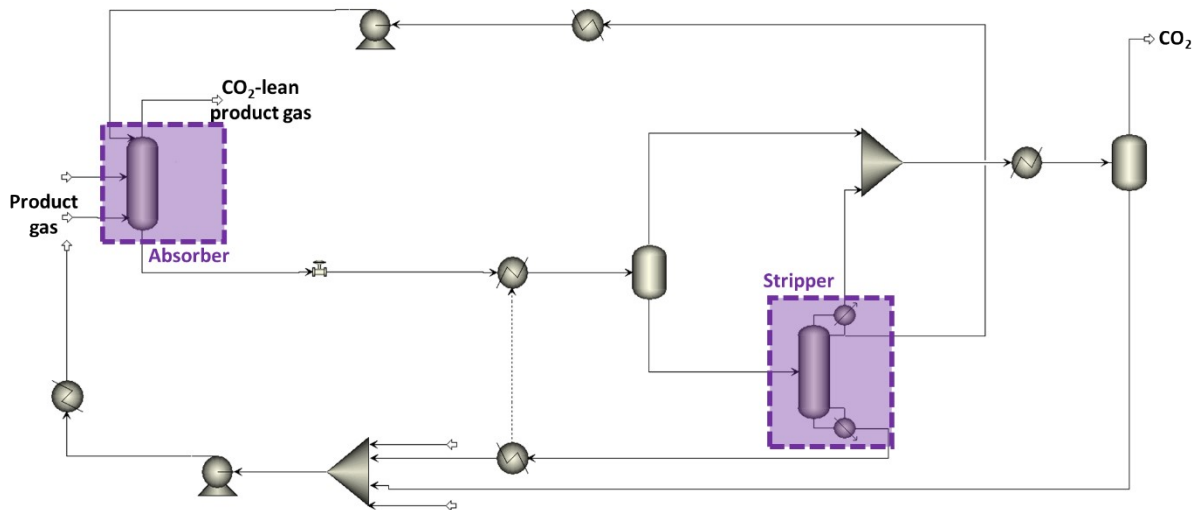


Figure S6. Process model for CO₂ removal using MEA absorption

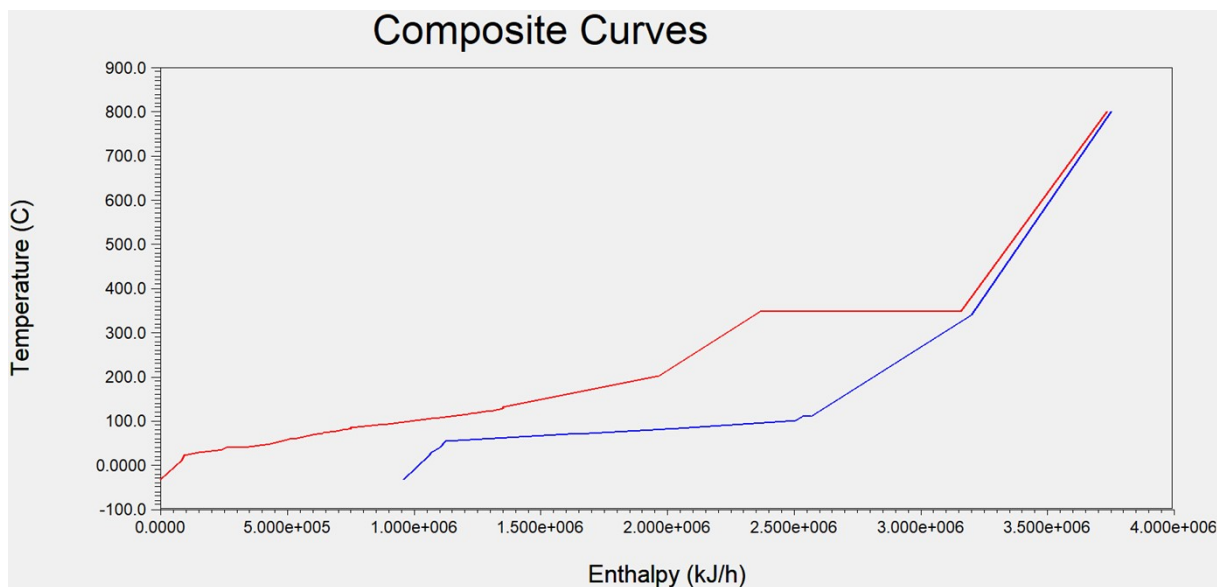


Figure S7. Composite curves from the Pinch analysis for the C²O-SOE scenario

S2 C²O Scenario: separation section evaluation

In the C²O scenario, CO₂, H₂ and the recycled gases are fed to the reactor where CO₂ and H₂ are converted to light olefins. Due to the complex product mixture, the separation section is key in determining the performance of the process. The possible separation section configuration includes a combination of the following units:

- i. **Gas-liquid separation units:** the boiling points of H₂, CO, C₂H₄, CO₂ and C₃H₆ at atmospheric pressure are -252.8 °C, -191.5 °C, -103.7 °C, -78.5 °C and -47.7 °C, respectively. The product mixture is separated in a gas-liquid separation unit (flash) operating at -14 °C or -30 °C and 1.8 MPa or 3.5 MPa. Most of the H₂ and CO remains in the gaseous stream while most of the CO₂ and C_{2,3}= remains in the liquid stream. The gaseous stream is recycled to the C²O reactor, and the liquid stream continues to the next unit of the separation section.
- ii. **H₂ removal by Pressure Swing Adsorption (PSA):** PSA is a popular method to produce high purity H₂. High H₂ recoveries are possible for gases with a H₂ content higher than 50%.⁸ A PSA unit was modelled using dry product gas at 2.06 MPa and with a H₂ recovery of 92%.
- iii. **H₂ and CO₂ removal by cellulose acetate (CA) membranes:** After gas-liquid separation, the CO₂ and H₂ molar fractions in the liquid stream are below 15% and 1%, respectively. CA membranes have shown high permeabilities for CO₂ and H₂, thus additional CO₂ and H₂ removal can be achieved using such a unit.
- iv. **CO₂ removal by absorption using a monoethanolamine (MEA) solution:** Amine scrubbing is a mature technology for CO₂ removal.⁹ In this process, CO₂ is captured from the product gas at 40 °C by absorption using a 30 wt% MEA solution. Regeneration of the CO₂-rich amine requires a high heating input, i.e., between 3.3 and 6.4 GJ per t CO₂ removed.² The required heat is provided by the exothermic C²O reactor.

Eight C²O separation scenarios are evaluated considering 4 different combinations of the separation units (Table S3). Water is removed by condensation and drying before entering the separation section in all scenarios.

Table S3. Separation section configuration evaluated for the C²O scenarios

Scenario	Separation section configuration
C²O-HP	H ₂ -PSA → Flash (10 °C and 3.54 MPa) → Membrane → MEA absorption
C²O-NP	H ₂ -PSA → Flash (-14 °C and 1.76 MPa) → Membrane → MEA absorption
C²O-2MEM	Membrane → Flash (25 °C and 3.54 MPa) → Membrane
C²O-SMA	Flash (-29.7 °C and 1.795 MPa) → Membrane → MEA absorption
C²O-SMT	Flash (-14 °C and 1.795 MPa) → Membrane → MEA absorption
C²O-SHP	Flash (-29.7 °C and 3.55 MPa) → Membrane → MEA absorption
C²O-STP	Flash (-14 °C and 3.55 MPa) → Membrane → MEA absorption
C²O-NM	Flash (-29.7 °C and 1.795 MPa) → MEA absorption

Key Performance Indicators (KPI) are defined to evaluate the performance of the C²O scenarios: Membrane load (Nm³ gas treated/t C_{2,3}=), CO₂ removed by MEA absorption (t CO₂ removed/t C_{2,3}=), Electricity required (MWh/t C_{2,3}=), Minimum cooling required (GJ/t C_{2,3}=), C_{2,3}= recovery in the separation section (%), C_{2,3}= molar fraction at the inlet of the C²O reactor (%) and CO₂ and H₂ conversions in the C²O reactor (%). An overview of the KPI for the 8 C²O scenarios is shown in Table S4.

Table S4. Key performance indicators for the 8 evaluated C²O scenarios

Case	C ² O-HP	C ² O-NP	C ² O-2MEM	C ² O-SMA	C ² O-SMT	C ² O-SHP	C ² O-STP	C ² O-NM
Electrolyzer type	PEM	PEM	PEM	PEM	PEM	PEM	PEM	PEM
Membrane load (Nm ³ gas/t C _{2,3} =)	1005	948	3787	765	717	879	811	-
CO ₂ removed by MEA absorption (t CO ₂ /t C _{2,3} =)	0.061	0.058	-	0.028	0.019	0.046	0.034	0.17
Electricity (MWh/t C _{2,3} =)	20.0	20.0	20.2	19.8	19.8	19.8	19.8	19.8
Cooling (GJ/t C _{2,3} =)	22.1	22.5	22.6	22.0	22.1	22.1	21.9	21.9
C _{2,3} = recovery in separation section (%)	68.5	68.9	54.3	55.5	40.7	66.2	56.4	63.6
C _{2,3} = molar feed to reactor (%)	2.89	2.84	5.09	4.91	8.43	3.21	4.74	3.60
CO ₂ conversion (%)	76.2	76.1	75.4	75.5	74.4	76.1	75.5	75.8
H ₂ conversion (%)	76.1	76.1	75.4	75.5	74.4	76.0	75.5	75.9

C²O-2MEM removes H₂ and CO₂ from the dry product stream using a highly selective membrane. The production of polymer-grade C_{2,3}= without MEA adsorption was possible using a 2-stage membrane separation (Figure 8). However, this scenario requires the most electricity per t C_{2,3}= due to the additional electricity for recompression of the recycle gas. This scenario requires a 5 times larger membrane load with respect to other C²O cases. The removal of the residual CO₂ and H₂O using adsorption beds is required in this case. This configuration is sensitive to the performance of the C²O reactor and membranes.

H₂ can be recovered using a H₂-PSA unit because of the high H₂ content in the dry product gas. Scenarios C²O-HP and C²O-NP include a H₂-PSA in the separation section (Figure S9). After H₂ recovery, CO and additional H₂ are separated in a gas-liquid separation unit. The liquid stream is vaporized and

the $C_{2,3}$ are recovered using membrane separation. To produce polymer-grade $C_{2,3}$, additional CO_2 is removed using MEA absorption and residual CO_2 and H_2O are removed using adsorption beds. When using the H_2 -PSA unit, the C^2O process achieves higher $C_{2,3}$ recoveries, lower $C_{2,3}$ molar fractions at the inlet of the C^2O reactor and slightly higher CO_2 conversions. Yet, these scenarios require the most electricity per t $C_{2,3}$ and the largest CO_2 absorption removal, after C^2O -2MEM.

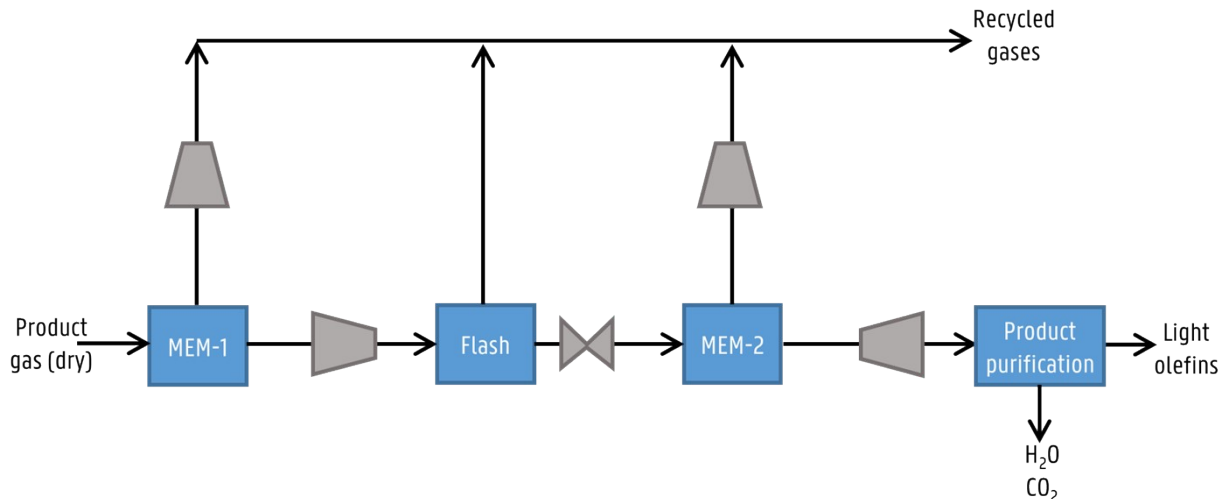


Figure S8. Separation section for C^2O -2MEM.

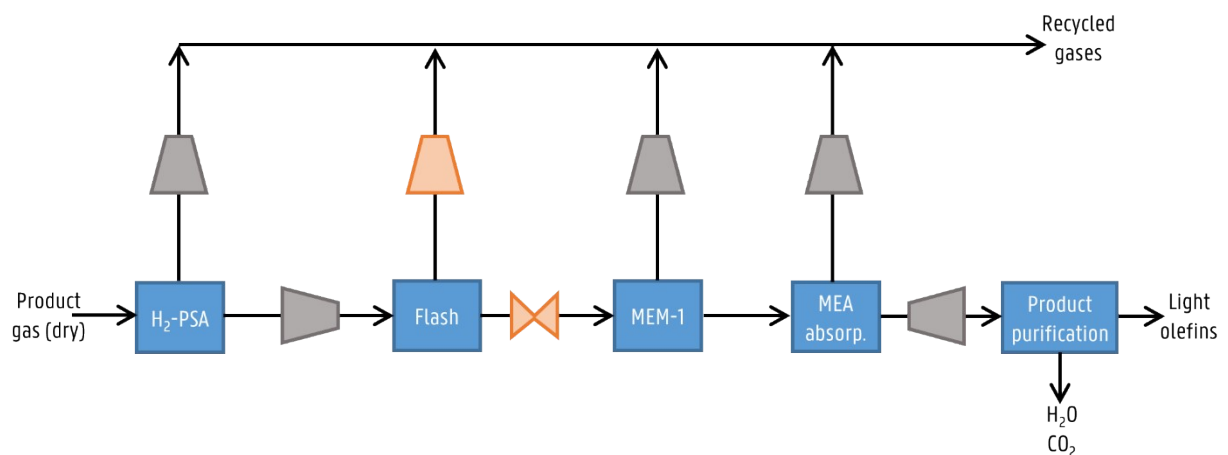


Figure S9. Separation section for C^2O -HP and C^2O -NP. C^2O -HP does not require the orange compressor and C^2O -NP does not require the orange valve.

The simplest separation is applied in C^2O -NM, this scenario does not use membranes (Figure S10). H_2 and CO are removed by a gas-liquid separation unit and the CO_2 is removed by MEA absorption. The H_2O from MEA absorption is removed by drying to achieve a polymer-grade product. The C^2O reactor provides enough heat for the regeneration of the amines, thus no external heat is required.

C^2O -SMA, C^2O -SMT, C^2O -SHP and C^2O -STP use the separation configuration of C^2O -HP and C^2O -NP but without the H_2 -PSA unit (Figure 11). The combination of 2 pressures (1.795 MPa and 3.55 MPa) and 2 temperatures ($-29.7\text{ }^\circ\text{C}$ and $-14\text{ }^\circ\text{C}$) is evaluated in the gas-liquid separators. Most of the H_2 and CO are removed in the flash, thus the membrane loads are the lowest. Polymer-grade $C_{2,3}$ is produced by removing CO_2 using MEA absorption and H_2O using a drying bed.

C^2O -SHP, C^2O -SMA, C^2O -STP, C^2O -SMT and C^2O -NM require the least electricity among the 8 evaluated scenarios (in bold in Table S4). C^2O -SHP is selected as the optimal C^2O scenario because it has the

highest $C_{2,3=}$ recovery in the product, the lowest $C_{2,3=}$ molar feed to the reactor and the highest CO_2 and H_2 conversions.

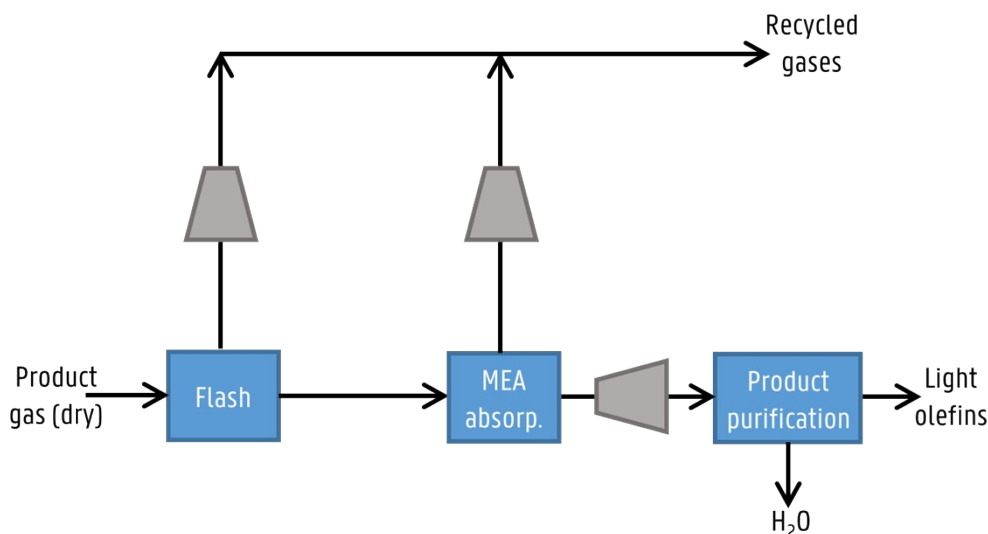


Figure S10. Separation section for C^2O -NM.

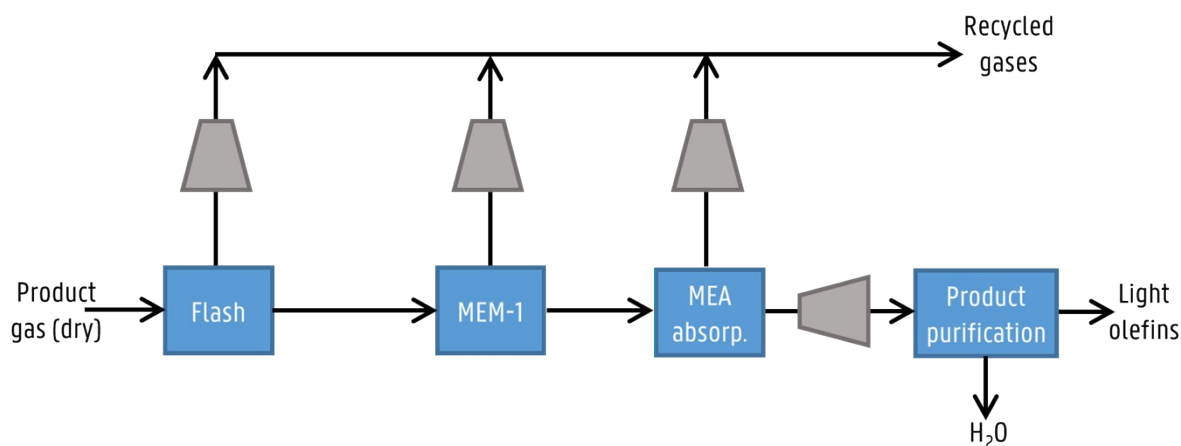


Figure S11. Separation section for C^2O -SMA, C^2O -SMT, C^2O -SHP and C^2O -STP.

S3 C^2O -SOE Scenario: sensitivity analyses

Five additional simulations evaluate the sensitivity of the C^2O -SOE scenario to the following parameters: (i) lower CO_2 conversion (C^2O -LC), (ii) lower $C_{2,3=}$ selectivity (C^2O -LS), (iii) lower propylene/ethylene ($C_3=/C_2=$) ratio (C^2O - C_3C_2R), (iv) lower CO_2 permeability for the membrane (C^2O -LPM) and (v) including paraffins as possible products in the C^2O reactor (C^2O -paraffins). An overview of the KPIs is shown in Table S5.

(i) C^2O -LC: lower CO_2 conversion in the C^2O reactor

The CO_2 conversion is reduced from 75.5% to 30.0%, the product distribution was defined by the RGIBBS reactor. The selectivity to $C_{2,3=}$ is 100% and, due to the stoichiometric $H_2:CO_2$ molar feed, the H_2 conversion equals the CO_2 conversion. The lower conversions cause a larger gas recycle, thus the membrane size and the CO_2 removed by MEA absorption are 4 and 7 times larger, respectively. Around 6% more electricity and 70% more cooling are required, due to: (i) the compression of a larger gas recycle and (ii) lower temperature required by cryogenic refrigeration to reach a similar $C_{2,3=}$ recovery in the flash. It is

challenging to recover $C_{2,3=}$, yet the $C_{2,3=}$ molar fraction at the inlet of the C^2O reactor is halved.

(ii) C^2O -LS: lower $C_{2,3=}$ selectivity in the C^2O reactor

The CO_2 conversion is fixed to 75.5% and the CO selectivity is increased from 0.01% to 80%. The H_2 conversion is lower than the CO_2 conversion due to the lower H_2 consumption for CO production (1:1 $H_2:CO_2$). The gaseous stream from the gas-liquid separation is rich in CO, thus a CO hydrogenation (RGIBBS) reactor is implemented to produce additional $C_{2,3=}$. This reactor operates at 420 °C and 4 MPa. This new section required an additional H_2O removal step and gas-liquid separation unit. The liquid streams from the flashes are mixed and vaporized for membrane separation. The CO_2 concentration is larger due to the lower yield to $C_{2,3=}$, thus the membrane size increases by 20% and 50% more CO_2 is removed by MEA absorption. The total electricity requirements increase by 0.4 MWh/t $C_{2,3=}$ due to additional cryogenic cooling and compression. Around 60% additional cooling are required per t $C_{2,3=}$. This case achieves a lower $C_{2,3=}$ recovery and a lower $C_{2,3=}$ molar fraction at the inlet of the C^2O reactor.

(iii) C^2O - C_3C_2R : lower $C_3=/C_2=$ ratio in the C^2O reactor

The CO_2 conversion and CO selectivity are fixed to 75.5% and 0.01% and the $C_3=/C_2=$ ratio is reduced from 95 to 3. The $C_{2,3=}$ recovery decreases from 56% to 52% in the gas-liquid separation unit due to the slightly larger $C_2=$ losses to the gaseous stream. More $C_{2,3=}$ are recycled to the C^2O reactor and the $C_{2,3=}$ molar feed composition increases to 6.9%. The membrane size and CO_2 removed by MEA absorption increase by around 10% and the electricity requirements increase by around 1%, due to additional cryogenic cooling.

(iv) C^2O -LPM: lower CO_2 permeability for the membrane

The CO_2 permeability of the membrane was halved and the performance of the C^2O reactor is defined by the RGIBBS block. The membrane size slightly increases by around 2% but less CO_2 is separated in the unit, thus the CO_2 removed by MEA absorption doubles. Yet, the electricity and cooling requirements did not increase significantly. In this scenario, the $C_{2,3=}$ losses in the permeate slightly increase and the $C_{2,3=}$ recovery, $C_{2,3=}$ feed molar ratio and the CO_2 conversion slightly decrease.

(v) C^2O -paraffins: paraffins are possible products in the C^2O reactor

The CO_2 conversion and CO selectivity are fixed to 75.5% and 0.01% and the $C_{2,3=}$ selectivities are adapted to produce a paraffins/olefins ratio of 5. Cryogenic distillation is required to remove the paraffins from the $C_{2,3=}$, this increases the $C_{2,3=}$ recovery and decreases the recycle of $C_{2,3=}$ to the C^2O reactor. The H_2 conversion is slightly higher than the CO_2 conversion due to the stoichiometry of the CO_2 to paraffins reactions. The electricity required increases by 3% because more H_2 is required per t $C_{2,3}$ but also because the process requires 1% more electricity and cryogenic refrigeration 54% more. This case does not produce enough excess steam for the SOE, thus electric boilers use additional 0.28 MWh/t $C_{2,3}$ electricity. Around 20% additional cooling is required and the cooling water volume triplicates. The membrane separation and CO_2 removal by MEA absorption increase by 3% and 4%, respectively.

Table S5. Key performance indicators for the C²O sensitivity analyses

Case	C ² O	C ² O-LC	C ² O-LS	C ² O-C ₃ C ₂ R	C ² O-LPM	C ² O-paraffins
Electrolyzer type	SOE	SOE	SOE	SOE	SOE	SOE
Membrane load (Nm ³ gas/t C _{2,3} =)	879	2799	1048	947	905	851 Nm ³ gas/t C _{2,3}
CO ₂ removed by MEA absorption (t CO ₂ /t C _{2,3} =)	0.046	0.31	0.070	0.046	0.096	0.048
Electricity (MWh/t C _{2,3} =)	16.0	17.0	16.4	16.2	16.0	16.5
Cooling (GJ/t C _{2,3} =)	8.1	14.1	12.7	8.75	8.1	9.9
C _{2,3} = recovery in separation section (%)	66.2	62.4	63.8	51.9	64.4	65.6
C _{2,3} = molar feed to reactor (%)	3.21	1.54	1.91	6.92	3.46	2.99
CO ₂ conversion (%)	76.1	30.0	75.5	75.5	75.9	75.5
H ₂ conversion (%)	76.0	30.0	35.1	75.5	75.9	76.9

S4 Economic feasibility of the scenarios

The break-even electricity prices were estimated to evaluate the economic feasibility of the different scenarios. The Break-Even Electricity price (BEE) was calculated using the equation below and the assumed utilities and material costs in Table S6. Light olefins, fuel gas, by-products and excess steam produce a profit, while the operational costs included water for electrolysis, water for cooling and naphtha. The cost of CO₂ as feedstock was not considered.

$$BEE \left(\frac{EUR}{MWh} \right) = \frac{\sum profit \left(\frac{EUR}{ton C_{2,3}=} \right) - \sum costs \left(\frac{EUR}{ton C_{2,3}=} \right)}{Electricity\ required \left(\frac{MWh}{ton C_{2,3}=} \right)}$$

Table S6. Assumed utilities and material costs for the processes

Materials and Utilities	Value	Reference
Utilities:		
Water for cooling	0.35 EUR/GJ	Based on ¹⁰ .
Water for electrolysis	0.07 EUR/kg H ₂	Based on ¹¹ .
High pressure steam	17.7 EUR/GJ	Based on ¹⁰ .
Medium pressure steam	13.7 EUR/GJ	Average value between ¹⁰ and ¹¹ .
Low pressure steam	9.6 EUR/GJ	Based on ¹² .
Materials:		
Naphtha	640 EUR/t naphtha	Based on ¹³ .
Light olefins	1000 EUR/t C _{2,3} =	Average in the range given by ¹⁴ .
Fuel gas	11.4 EUR/GJ	Based on ¹⁵ .
By-products	854 EUR/t by-products	Selected to obtain a break-even electricity price of 50 EUR/MWh for NSC

The price of by-products was adjusted so the NSC case would break-even at an electricity price of 50 EUR/MWh, this would allow for a comparative analysis between scenarios. The break-even electricity prices are summarized in Table S7 considering: (a) profit from steam as by-product and no CO₂ tax, (b) profit from steam as by-product and including CO₂ tax (100 EUR/t CO₂), and (c) no profit from steam as by-product and including CO₂ tax (100 EUR/t CO₂).

The RDR scenarios are the most feasible due to the lower naphtha input per ton C_{2,3=} than NSC and the lower electricity consumption than the CO₂-based routes. The e-NSC scenario requires a slightly more economic electricity than NSC (below 43 EUR/MWh) to be profitable. The C²O-SOE and CO₂red+COhyd cases are the next most feasible cases, mainly due to the lower electricity required per ton C_{2,3=} among the other CO₂-based scenarios, they are break-even for an electricity price of 61-86 EUR/MWh, depending on the additional income from steam export and the CO₂ tax price. C²M+MTO-SOE is slightly less feasible due to the lower excess steam available for export. CO₂red+FTO is the least feasible among the CO₂-based routes due to the high electricity required per ton C_{2,3=} and lack of steam available for export.

Table S7. Break-even electricity price

Scenario	Break-even electricity price	Break-even electricity price	Break-even electricity price
	steam as by-product and without CO ₂ tax [EUR/MWh]	steam as by-product and including CO ₂ tax [EUR/MWh]	including CO ₂ tax [EUR/MWh]
NSC	50	4	50
e-NSC	44	43	44
RDR	93	90	93
e-RDR	114	113	114
C ² O-PEME	58	73	50
C ² O-SOE	67	86	62
C ² M+MTO-PEME	53	68	50
C ² M+MTO-SOE	63	82	61
CO ₂ red+COhyd	67	85	61
CO ₂ red+FTO	41	46	41

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