Supporting Information

Electrochemical CO_2 reduction with ionic liquids: Reviewing and evaluating

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1 Model and parameters

Table S1 Key parameters and assumption for calculating TPC of each target product from CO_2R

Parameters	**value**
Reference year	2022
System lifetime, yr*	20
Interest rate, %*	10
Operating time, day*	355
CO_2 feedstock flow rate, ton/day*	50
Single pass conversion of CO_2*	0.5
Total conversion of CO_2*	100
Electricity price, €/kWh [1]	0.05
ILs price, €/ton [2]	5800
H_2O price, €/ton [2]	0.44
CO_2 price, €/ton	40
Catalyst cost for CO product, €/kg [3]	8500
Catalyst cost for CH_3OH product, €/kg	5000
Catalyst cost for CH_4 product, €/g [4]	334.96
Catalyst cost for Syngas (1:1) product, €/g [5]	30.36
Au price (a component in the catalyst for Syngas (2:1) product), €/g [6]	56.64
Ni foam price (a component in the catalyst for Syngas (2:1) product), €/m^2 [7]	15.3

*Assumed data

Table S2 Main parameters adopted from the current central H2A production for calculating the electrolyzer cost [8]

Parameters	**value**
Stack price, $/kW (2010)	220
Current density, A/cm^2	0.175
Voltage, V	1.75
Stack price, €/m^2 (2022)	978.22

Installation cost factor	1.20

Table S3 Main parameters of PSA [9]

Parameters	**Value**
Ref. Gas flow, m^3/h	1000
Ref. Capital cost, € (2018)	1738000
Ref. Energy usage to operate separator/recycle, kWh/m^3	0.23
Scale factor	0.65

Table S4 Main parameters of distillation [10]

Parameters	**Value**
Ref. Liquid flow, kg/h	77469
Ref. Capital cost, € (2020)	13200000
Ref. Operating cost, €/day (2022)	63141
Scale factor	0.7

Table S5 The value of CEPCI in recent years

Year	**Cost Index**
2010	550.8
2011	585.7
2012	584.6
2013	567.3
2014	576.1
2015	556.8
2016	541.7
2017	567.5
2018	603.1
2019	607.5
2020	596.2
2021	773.1
2022	815.7

2 Calculations process

The calculation methods of TPC, TCC, and TOC for each product are similar, and thus in this part, CH_3OH as the target product was selected as an example to expound the detailed calculation process.

2.1 Calculation of material and energy streams

(1) The current needed for CO_2R:

$$I_{CO_2} = \frac{CO_2\,production\,rate}{M_{MeOH}} \times Z \times F = \frac{50\frac{ton}{day}}{44\frac{g}{mol}} \times \frac{day}{86400\,s} \times 10^6\frac{g}{ton} \times 6e^- \times 9648$$

(2) Total current:

$$I_{total} = \frac{I_{CO_2}}{FE} = \frac{7614030.9\,A}{0.886} = 8593714.4\,A$$

(3) The electrolyzer area (TEA) demand:

$$TEA = \frac{I_{total}}{j} = \frac{8593714.4\,A}{0.067\,A/cm^2} \times \frac{m^2}{10^4\,cm^2} = 12826.439\,m^2$$

(4) The power demand:

$$P = V \times I_{total} = 3.28\,V \times 8593714.4\,A \times \frac{kW}{1000\,W} = 28187.383\,kW$$

(5) MeOH production flow rate

$$MeOH\,production\,rate = \frac{CO_2\,inlet\,flow\,rate \times Total\,conversion\,rate}{M_{CO_2}} \times M_{MeOH} = \frac{50\frac{ton}{day} \times 100\%}{44\,g/mol} \times 32\frac{g}{mol} = 36.36\,ton/day$$

(6) In calculation, it was assumed that the methanol-rich electrolyte was recycled until reaching a steady-state concentration of 30 wt%. For other target products, there is no need to recycle the electrolyte, while the initial cost for providing electrolyte was calculated based on the assumption that the flow rate of the electrolyte is equal to the case of MeOH. The continuous electrolyte flow rate is:

$Electrolyte\ flow\ rate$

$$= MeOH\ production\ flow\ rate \div 0.3 = 36.36\frac{ton}{day} \div 0.3 = 121.21\ ton/day$$

(7) Hydrogen flow rate is:

$$H_2\ flow\ rate = \frac{I_{total} \times FE_{H_2}}{Z \times F} \times M_{H_2} = \frac{8593714.4\ A \times 0.114}{2\ e^- \times 96485\ C/mol} \times 2\frac{g}{mol} \times \frac{ton}{10^6\ g} \times 86400\frac{s}{day}\frac{ton}{day} = 9747.59\ m^3/day$$

(8) Oxygen flow rate is:

$$O_2\ flow\ rate = \frac{I_{total}}{Z \times F} \times M_{O_2} = \frac{8593714.4\ A}{4\ e^- \times 96485\ C/mol} \times 32\frac{g}{mol} \times \frac{ton}{10^6\ g} \times 86400\frac{s}{day} = 61.56\frac{ton}{day}$$

(9) Consumption of water for reactions:

The reactions involving water as a reactant during CO_2R:

$4H_2O + 2CO_2 \rightarrow 2CH_3OH + 3O_2$

$2H_2O \rightarrow 2H_2 + O_2$

$$H_2O\ consumption = \frac{MeOH\ production\ flow\ rate}{M_{MeOH}} \times 2M_{H_2O} + \frac{H_2\ flow\ rate}{M_{H_2}} \times M_{H_2O} = \frac{36.36\ ton/day}{32\ g/mol} \times 2 \times 18\frac{g}{mol} + \frac{0.877\ ton/day}{2\ g/mol} \times 18\frac{g}{mol} = 48.80\ ton/day$$

(10) Total gas flow rate for PSA separation

$$CO_2\ single\ outlet\ flow\ rate = CO_2\ feedstock\ flow\ rate \times Single\ conversion\ rate\ of\ CO_2 = 50\frac{ton}{day} \times 0.5 = 25\frac{ton}{day} = 12690.355\ m^3/day$$

$$Total\ gas\ flow\ rate = CO_2\ single\ outlet\ flow\ rate + H_2\ flow\ rate = 12690.355\frac{m^3}{day} + 9747.59\frac{m^3}{day} = 22437.944\ \frac{m^3}{day}$$

2.2 Economic evaluation

2.2.1 Capital cost

(1) Stack cost and BoP cost

The electrolyzer cost for the stack component from the H2A analysis for a central grid electrolysis is 220 \$/kW in 2010 with the current density and voltage of 0.175 A/cm^2 and 1.75 V, respectively. Considering the inflation and exchange rate, the stack price was estimated to be 319.42 €/m^2 in 2022. Therefore, the stack price with installation is 1173.86 €/m^2 in 2022 according to the below calculation. Besides, the balance of plant (BoP) capital cost was set to be 35 % of the total cost, while that for the stack cost was 65 %.

$$Ref.\,Stack\ price(with\ installation)$$
$$= \frac{Stack\ price\ (2010) \times Inflation\ rate \times Current\ density\ \times Voltage \times Ins}{Exchange\ rate\ of\ EUR\ into\ USD}$$
$$= \frac{220\frac{\$}{kW} \times 1.48 \times 0.175\ A/cm^2 \times 1.75\ V \times 1.2 \times 10000}{1.02 \times 1000} = 1173.86\ €/m^2$$

$$Stack\ cost$$
$$= Ref.\,Stack\ price\ \ (with\ installation) \times TEA = 1173.86\frac{€}{m^2} \times 12826.439\ m$$

$$BoP\ cost = Stack\ cost \times \frac{0.35}{0.65} = 15.06\ M€ \times \frac{0.35}{0.65} = 8.11\ M€$$

(2) PSA capital cost

$$Ref.\,Capital\ cost\ (2022)$$
$$= \frac{Ref.\,Capital\ cost\ (2018)}{CEPIC\ (2018)} \times CEPIC\ (2022) = \frac{1738000\ €}{603.1} \times 815.7 = 23506$$

$$PSA\ capital\ cost$$
$$= Ref.\,Capital\ cost\ (2022) \times \frac{Gas\ flow\ rate}{Ref.\,Gas\ flow\ rate}^{Scale\ factor} = 2350665.89$$
$$= 2.25\ M€$$

(3) Distillation capital cost

$$Ref.\,Capital\ cost\ (2022)$$
$$= \frac{Ref.\,Capital\ cost\ (2020)}{CEPIC\ (2018)} \times CEPIC\ (2022) = \frac{13200000\ €}{596.2} \times 815.7 = 1805$$

$$Distillation\ capital\ cost$$
$$= Ref.\,Capital\ cost\ (2022) \times \frac{Liquids\ flow\ rate}{Ref.\,Liquid\ flow\ rate}^{Scale\ factor} = 18059779\ €$$
$$\times \frac{36.36\ ton/day \times 1000kg/ton}{77469\ kg/h \times \frac{24\ h}{day}}^{0.7} = 0.25\ M€$$

(4) Initial electrolyte and catalyst costs

The electrolyte consists of 80 wt% [Bmim][BF_4] and 20 wt% H_2O for the case of CH_3OH

production. It was assumed that 4 h was needed to complete recycling the electrolyte. For the other target products, the composition of electrolytes is shown in **Fig. S8**. The prices of all the studied ILs in our work were set to be 5800 €/ton.

$$
\begin{aligned}
&\mathit{Initial\ electrolyte\ cost} \\
&\quad = 0.8 \times \mathit{Electrolyte\ flow\ rate} \times \mathit{Time} \times \mathit{ILs\ price} + 0.2 \times \mathit{Electrolyte\ flow\ rate} \\
&\quad \times \mathit{Time} \times H_2O\ \mathit{price} \\
&\quad = 0.8 \times 121.21 \frac{ton}{day} \times 4\ h \times 5800 \frac{€}{ton} + 0.2 \times 121.21 \frac{ton}{day} \times 4\ h \times 0.44 \frac{€}{ton} \\
&\quad = 0.094\ M€
\end{aligned}
$$

The amount of catalyst is 1 mg/cm^2.

$$
\begin{aligned}
&\mathit{Initial\ catalyst\ cost} \\
&\quad = \mathit{Catalyst\ price} \times \mathit{Catalyst\ amount} \times \mathit{TEA} = 5000 \frac{€}{kg} \times 1 \frac{mg}{cm^2} \times \frac{10000 cm^2}{m^2} \\
&\quad \times \frac{kg}{10^6\ mg} \times 12826.439\ m^2 = 0.64\ M€
\end{aligned}
$$

2.2.2 Operating cost

(1) CO_2 cost

$$
\begin{aligned}
&CO_2\ \mathit{cost} \\
&\quad = CO_2\ \mathit{feedstock\ flow\ rate} \times \mathit{Total\ conversion\ rate} \times \mathit{Operating\ time} \times (\\
&\quad \frac{ton}{day} \times 100\% \times 355 \frac{day}{yr} \times 40 \frac{€}{ton} = 0.71\ M€/yr
\end{aligned}
$$

(2) Electrolyte make-up

The early make-up was set to be 5 % of the initial amount for IL.

$$
\begin{aligned}
&\mathit{Electrolyte\ make-up} \\
&\quad = \mathit{Initial\ electrolyte\ flow\ rate} \times 0.8 \times \mathit{Time} \times \frac{5\ \%}{yr} \times \mathit{IL\ price} + H_2O\ \mathit{cons} \\
&\quad \times \mathit{Operating\ time} \times H_2O\ \mathit{price} \\
&\quad = 121.21 \frac{ton}{day} \times 0.8 \times 4\ h \times \frac{5\ \%}{yr} \times 5800 \frac{€}{ton} + 48.80 \frac{ton}{day} \times 355 \frac{day}{yr} \times 0.44 \frac{€}{ton} \\
&\quad = 0.012\ M€/yr
\end{aligned}
$$

(3) Electricity cost

$$
\begin{aligned}
&\mathit{Electricity\ cost} \\
&\quad = \mathit{Power\ demand} \times \mathit{Electrocity\ price} \times \mathit{Operating\ time} = 0.05 \frac{€}{kWh} \times 281 \\
&\quad \times 355 \frac{day}{yr} \times 24 \frac{h}{day} = 12.01\ M€/yr
\end{aligned}
$$

(4) PSA operating cost

$$
\begin{aligned}
&\mathit{PSA\ operating\ cost} \\
&\quad = \mathit{Ref.\ Energy\ usage\ to\ operate\ separator/recycle} \times \mathit{Electricity\ price} \times \\
&\quad = 0.23\ kWh/m^3 \times 22437.944 \frac{m^3}{day} \times 0.05 \frac{€}{kWh} \times 355\ day/yr = 0.092\ M€/y
\end{aligned}
$$

(5) Distillation operating cost

$$\begin{aligned}&\mathit{Distillation\ operating\ cost}\\&= Ref.\,Distillation\ operating\ cost\ \times \frac{MeOH\ flow\ rate}{Ref.\,Liquid\ flow\ rate} \times Operating\ time\\&= 63141\frac{€}{day} \times \frac{121.21\frac{ton}{day} \times 1000\ kg/ton}{\frac{77469kg}{h} \times \frac{24h}{day}} \times 355\frac{day}{yr} = 0.44\ M€/yr\end{aligned}$$

2.2.3 Total production cost (TPC)

$$TPC = ACC + TOC$$

$$ACC = \frac{TCC}{(1-(1+i)^{-L})/i}$$

where *L* and *i* are the project lifetime and interest rate, respectively.

2.3 Environmental impact evaluation

2.3.1 Life cycle inventory

Table S6 Life cycle inventory (LCI) per unit mass of the target product.

	Base case	Case 1	Case 2	Case 3
Raw material (kg)				
CO_2	1.375	1.375	1.375	1.375
H_2O	1.342	0.750	0.651	0.580
IL make-up	6.26×10^{-5}	6.26×10^{-5}	6.26×10^{-5}	6.26×10^{-5}
Products (kg)				
CH_3OH	1.000	1.000	1.000	1.000
H_2	0.024	0.021	0.010	0.002
O_2	1.693	1.667	1.579	1.515
Energy (kWh)				
Electricity for CO_2R	18.60	16.75	13.22	7.61
BoP energy	1.86	1.67	1.32	0.76
PSA energy	0.14	0.13	0.11	0.09
Distillation energy	8	8	8	8
Total energy	28.61	26.56	22.65	16.46

*The energy of the BoP was assumed to be 10 % of the energy required for CO_2R.

2.3.2 Global warming impact (base case as the example)

Table S7 GWI of the related materials and CH_3OH as the target product

	GWI (kg CO_2e/kg material or product)
Raw materials	
CO_2 [11]	-0.5
H_2O[a]	0.1794
IL	3.5
Products	
H_2 [b]	11.5
O_2[c]	0.046

[a]Calculated according to the specific energy consumption of H_2O (0.78 kWh/ton) [12] and the greenhouse gas emission intensity of electricity generation in Europe in 2020 (0.23 kg CO_2/kWh) [13]; [b]Calculated according to the specific energy consumption of H_2 (50 kWh/kg) and the greenhouse gas emission intensity of electricity generation in Europe in 2020 (0.23 kg CO_2/kWh) [13]; [c]Calculated according to the specific energy consumption of O_2 (200 kWh/ton) [14] and the greenhouse gas emission intensity of electricity generation in Europe in 2020 (0.23 kg CO_2/kWh) [13].

In order to compare and analyze the GWI of each target product, kWh was used (1 kWh=3600 kJ). Herein, the HHV of CH_3OH is 726.51 kJ/mol. [15]

$$CO_{2GWI} = \frac{-0.5\,\frac{kg\ CO_2e}{kg\ CO_2\ capture} \times 1.375\,\frac{kg\ CO_2}{kg\ CH_3OH} \times 32\frac{g}{mol}}{726.51\frac{kJ}{mol} \times \frac{10^3\ kg}{g}} \times \frac{3600\ kJ}{kWh} \ldots CO_2e/kWh$$

$$H_2O_{GWI} = \frac{0.23\,\frac{kg\ CO_2e}{kWh} \times 0.78\,\frac{kWh}{ton\ H_2O} \times 10^{-3}\frac{ton}{kg} \times 1.342\,\frac{kg\ H_2O}{kg\ CH_3OH} \ldots}{726.51\frac{kJ}{mol} \times \frac{10^3 kg}{g}} \ldots \frac{3600\ kJ}{kWh} = 3.818 \times 10^{-5}\ kg\ CO_2e/kWh$$

$$IL_{GWI} = \frac{3.5\frac{kg\ CO_2e}{kg\ IL}\times 6.26\times 10^{-5}\frac{kg\ IL}{kg\ CH_3OH}\times 32\frac{g}{mol}}{726.51\frac{kJ}{mol}\times\frac{10^3 kg}{g}}\times\frac{3600\ kJ}{kWh} = kg\ CO_2e/kWh$$

$$O_{2GWI} = \frac{0.046\frac{kg\ CO_2e}{kg\ O_2}\times 1.693\frac{kg\ O_2}{kg\ CH_3OH}\times 32\frac{g}{mol}}{726.51\frac{kJ}{mol}\times\frac{10^3 kg}{g}}\times\frac{3600\ kJ}{kWh} = 0.01$$

$$H_{2GWI} = \frac{0.23\frac{kg\ CO_2e}{kWh}\times 50\frac{kWh}{kg\ H_2}\times 0.024\frac{kg\ H_2}{kg\ CH_3OH}\times 32\frac{g}{mol}}{726.51\frac{kJ}{mol}\times\frac{10^3 kg}{g}}\times\frac{3600}{kW} CO_2e/kWh$$

$$Energy_{GWI} = \frac{0.23\frac{kg\ CO_2e}{kWh}\times 28.61\frac{kWh}{kg\ CH_3OH}\times 32\frac{g}{mol}}{726.51\frac{kJ}{mol}\times\frac{10^3 kg}{g}}\times\frac{3600\ kJ}{kWh} = 1.043$$

$$Net_{GWI} = CO_{2GWI} + H_2O_{GWI} + IL_{GWI} + Energy_{GWI} - O_{2GWI} - H_{2burden} = \frac{CO_2e}{kWh} + 3.818\times 10^{-5}\ kg\frac{CO_2e}{kWh} + 3.474\times 10^{-5}\ kg\frac{CO_2e}{kWh} + 1.043\ e/kWh - 0.012\ kg\ CO_2e/kWh - 0.044\ kg\ CO_2e/kWh = 0.88\ kg\ CO_2e/kWh$$

3 Additional parameters for other products

Fig. S8 Components of the systems with the best performance for CO_2R to C1 products

Product	Electrocatalyst	Electrolyte (cathode)
CO [16]	CdN_4S_1/CN	0.5 M [Bmim][PF_6] in MeCN

CH_3OH [17]	Sn_1/V_0-CuO	25 mol% [Bmim][BF_4]/75 mol% H_2O
CH_4 [18]	$MoTe_2$ nanosheets	[Bmim][BF_4]/H_2O (with a mass ratio of 96:4)
Syngas (H_2/CO 1:1) [19]	F-g-In_2Se_3/CP	30 wt% [Bmim][PF_6]/65 wt% MeCN/5 wt% H_2O
Syngas (H_2/CO 2:1) [20]	Au NWs/Ni foam	0.1 M Bu_4NBr in DMF

Fig. S9 Prices of the raw materials for the production of other products

Raw materials	Price
MeCN [21]	3400 €/ton
DMF [22]	3120 \$/ton
Au [6]	56.64 €/g
Ni foam [7]	15.3 €/m^2
In_2Se_3 [5]	30.36 €/g
$MoTe_2$ nanosheets [4]	3497 SEK/g
CdS [3]	8500 €/kg

Fig. S10 GWI of the raw materials for the production of other products

Raw materials	GWI
MeCN [23]	1.4 kg CO_2e/kg MeCN
DMF [24]	2.98 kg CO_2e/kg DMF

Fig. S11 HHV of other products

Product	HHV
CO[a]	283.00 kJ/mol
CH_4[a]	890.31 kJ/mol
H_2[a]	285.80 kJ/mol
Syngas (H_2/CO 1:1)[b]	18960.00 kJ/kg
Syngas (H_2/CO 2:1)[b]	26706.25 kJ/kg

[a]The data were taken from the references; [15] [b]The data were calculated based on the values of CO and H_2 according to their ratio.

4 Identification of the state-of-the-art research for CH_3OH as the target product

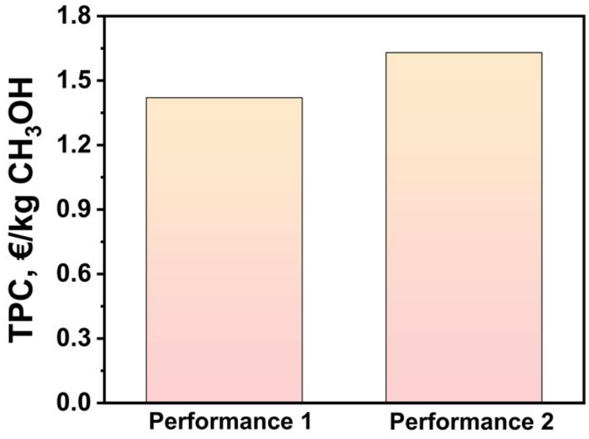

Fig. S1 TPC of CH_3OH form CO_2R with different performances. Performance 1: the FE of 88.6 % along with the current density of 67.0 mA cm^{-2}; Performance 2: the FE of 67.4 % along with the current density of 122.7 mA cm^{-2}.

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