Supplementary Information (SI) for Green Chemistry.

Supplementary Information

Feasibility and sustainability of emerging CCU pathways for formic acid production

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VLE validation results for important pairs involved in separation procedure based on the experimental data

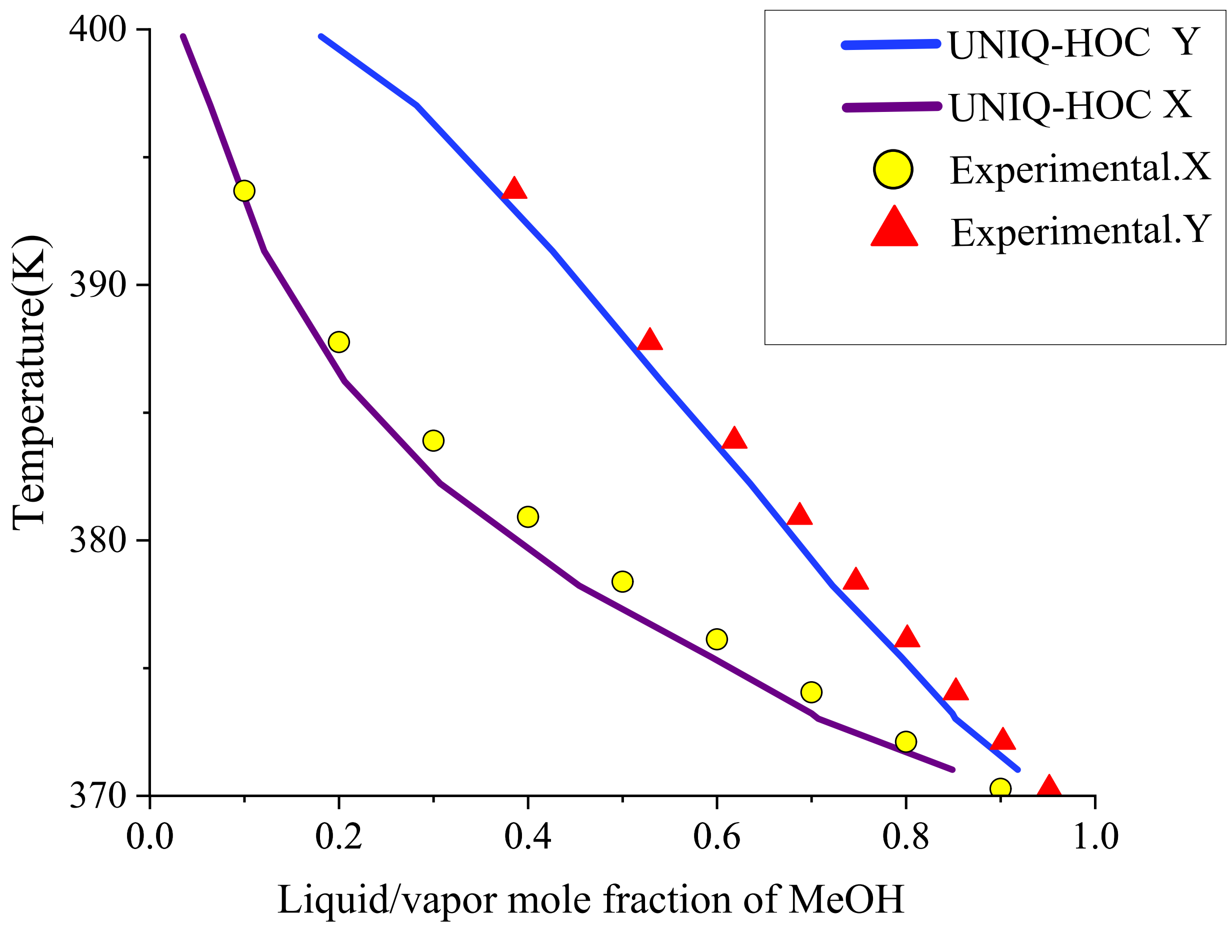

Fig.S1. T_{XY} diagram for H_2O/MeOH (3 bar) for the conventional process. Experimental data is form Hirata et al [1].

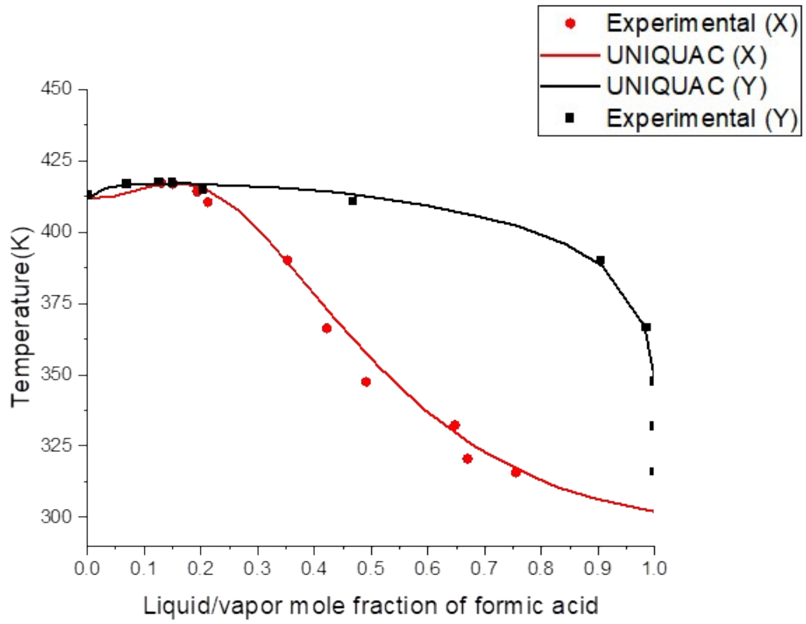

Fig.S2. T_{XY} diagram Formic acid/n-butylemidazol mixture for the thermo-catalytic process at 50 mmHg[2].

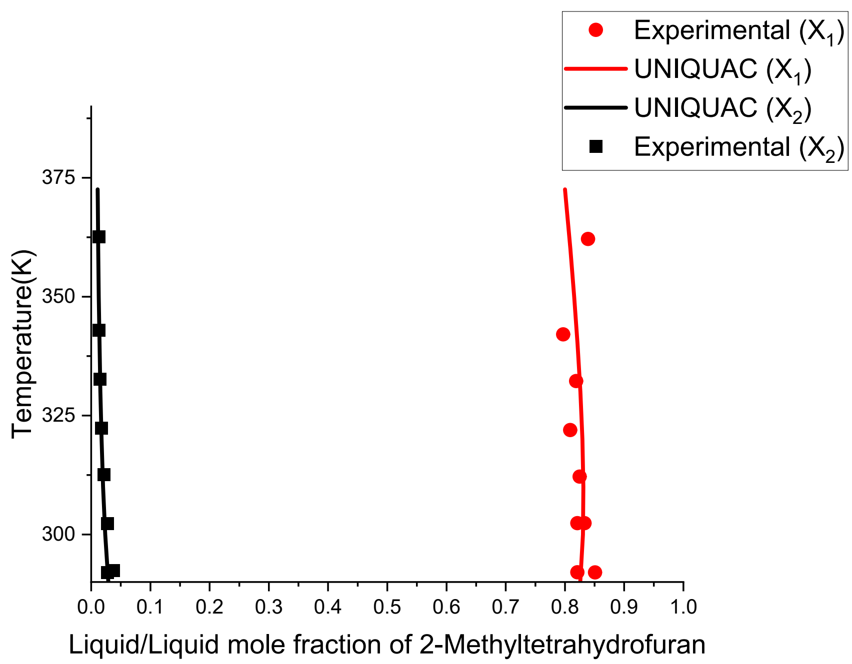

Fig.S3. T_{XX} diagram of water and 2-methyltetrahydrofuran LLE data for the electrochemical reduction process. The Experimental data was obtained from M. Glass et al (2017) [3].

Table S1. Process operating conditions and parameters used for designing the electrochemical reduction downstream process.

Unit operation	**Parameter**	**Value**	**Reference**
PSA	Operating conditions	400 °C, 30 bar	X. Zhu et al. [4]
	H_2 purification	99.99 mol%	
	Catalyst	Fe_2O_3-Cr_2O_3	
Extractor	Operating conditions	40 °C, 1 bar	A.T. Laitinen et al. [5]
Azeotropic Distillation	Operating conditions	1.1 bar, equilibrium	A.T. Laitinen et al.[5]
Striper	Operating conditions	4 bar, equilibrium	A.T. Laitinen et al.[5]

Table S2. CO_2 electrolyzer technoeconomic assumption parameters and process operating conditions.

Parameter	**Value**	**Reference**
Electrolyzer faradaic efficiency (HCOOH)	90%	X. Jiang et al. [6]
Current density	500 A/cm^2	X. Jiang et al. [6]
catalyst	BiO_n cluster	X. Jiang et al.[6]
HCOOH concentration	~5.04 g/l	K. Fernández-Caso et al. [7]
Total CO_2 conversion	44%	X. Jiang et al.[6]
Reference electrolyzer total cost per area	$\$48,493/m^2$	J.M. Spurgeon et al.[8]
scaling factor	0.65	

Table S3. PSA gas separation and recycle technoeconomic assumption parameters[9].

Parameter	Value
Reference total gas flow to separator	4.40 x 106 mol/h
Reference separator/recycle cost, 2018 USD	11,214,299
Scaling exponent	0.65
CO_2 recycling efficiency	97%
CO loss to recycle	0%
Case-dependent total gas flow to separator	*Y* mol/h
CO2/CO separator/recycle scaled capital expense	$[Y/(4.40 \times 10^{6})]^{0.65}$*(11,214,299)
Reference power to operate separator/recycle	10 kW
Reference annual energy usage to operate separator/recycle 8.76 x 10^4 kWh/yr	8.76 x 104 kWh/yr

Table S4. List of parameters with their cost reference

Materials and utilities	**Cost**
Gray H_2 (USD/kg)	1.88 [10]
Green H_2 (USD /kg)	3.53 [11]
Captured CO_2 (USD /kg)	0.035 [12]
TREA (USD /kg)	2.5 (from www.alibaba.com)
Catalyst (CH_3ONa, in USD /kg)	3.5 (from www.alibaba.com)
Catalyst (Ru/bpyTN-30-CTF, in USD /g)	175.35/g [13]
Catalyst ($BiO_{n,}$ in USD /g)	1.8/g[14]
KOH (USD /kg)	0.6 (from www.alibaba.com)
2-Methyltetrahydrofuran (USD /kg)	2.5 (from www.alibaba.com)
O_2 (Tonne /hr)	38.07(from www.alibaba.com)
CO (USD /Tonne)	70[15]
Methanol (USD /Tonne)	420(from www.alibaba.com)
CO_2 from DAC (USD /kg)	0.17[17]
Cooling water (USD 0.028/ Tonne)	0.354[16]
Electricity (USD /kWh)	0.06 [16]
LP Steam (USD /GJ)	14.05[16]
MP steam (USD /GJ)	14.83[16]
HP Steam (USD/GJ)	17.7[16]

Table S5. Total capital investment estimation used based on the ratio factor for fluid processing plant.

	Ratio factor for fluid processing plant (of delivered equipment cost)	**Reference**
Direct cost		MS Peters et al[18]
Delivery, % of purchased equipment	0.1	
Subtotal: Purchased equipment (delivered)	1	
Purchased equipment installation	0.47	
Instrumentation and Controls	0.36	
Piping	0.68	
Electrical systems	0.11	
Buildings (including services)	0.18	
Yard improvements	0.1	
Service facilities	0.7	
Indirect cost		
Engineering and supervision	0.33	
Construction expenses	0.41	
Legal expences	0.04	
Contractor's fee	0.22	
Contingency	0.44	
Working capital (WC)	0.89	

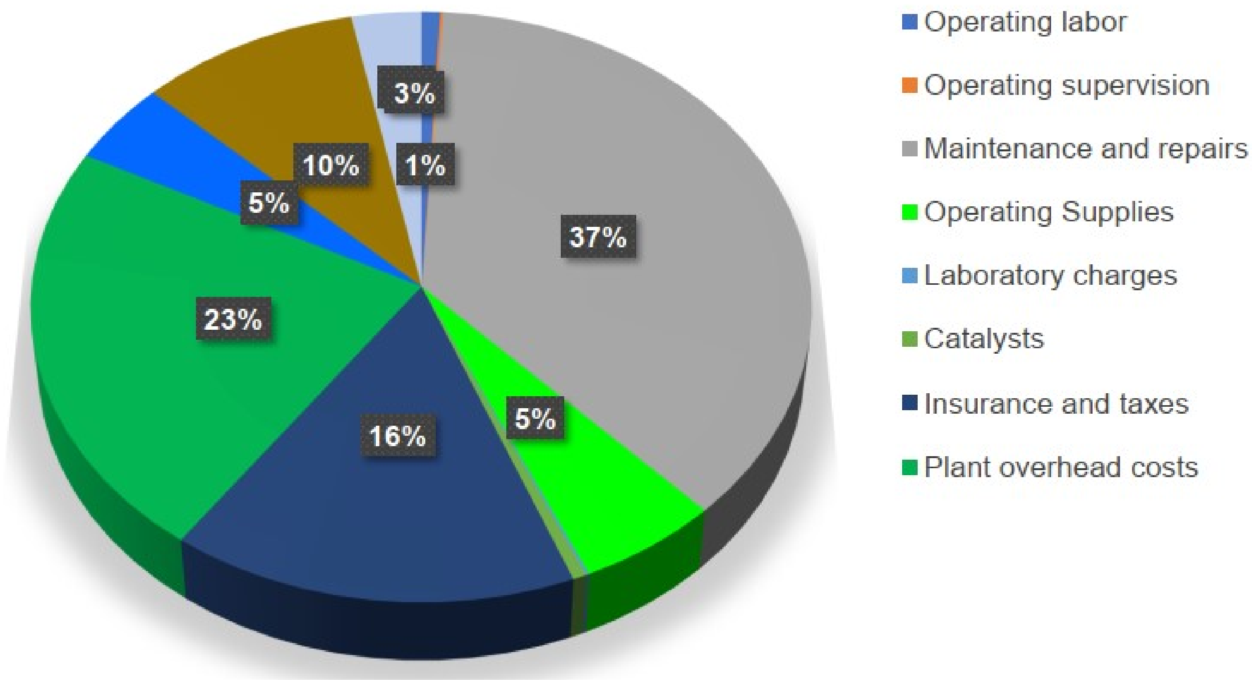

Fig.S4. cost break down of other operational expenses for the electrochemical reduction pathway (CCUER)

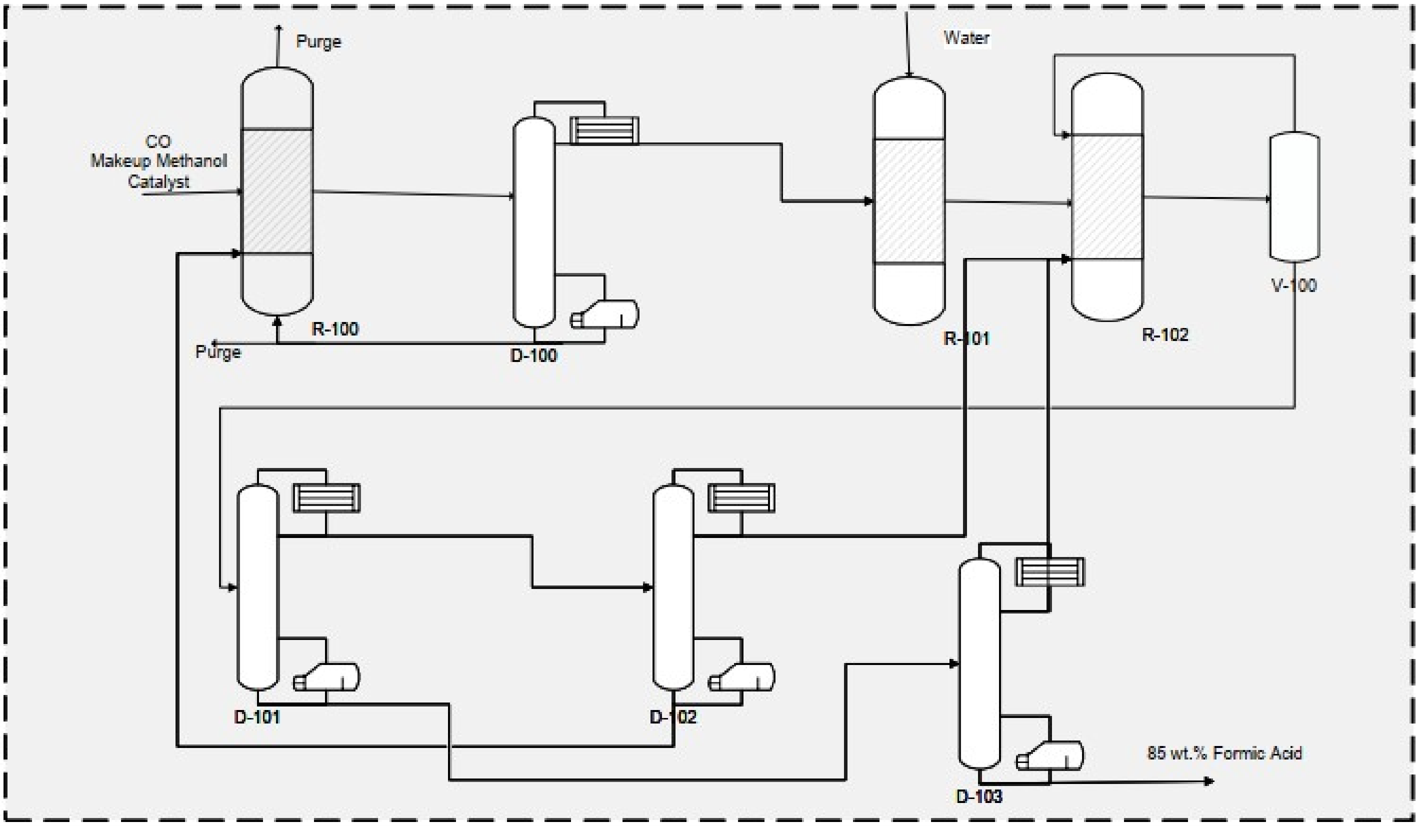

Fig.S5. System boundary of the conventional process pathway

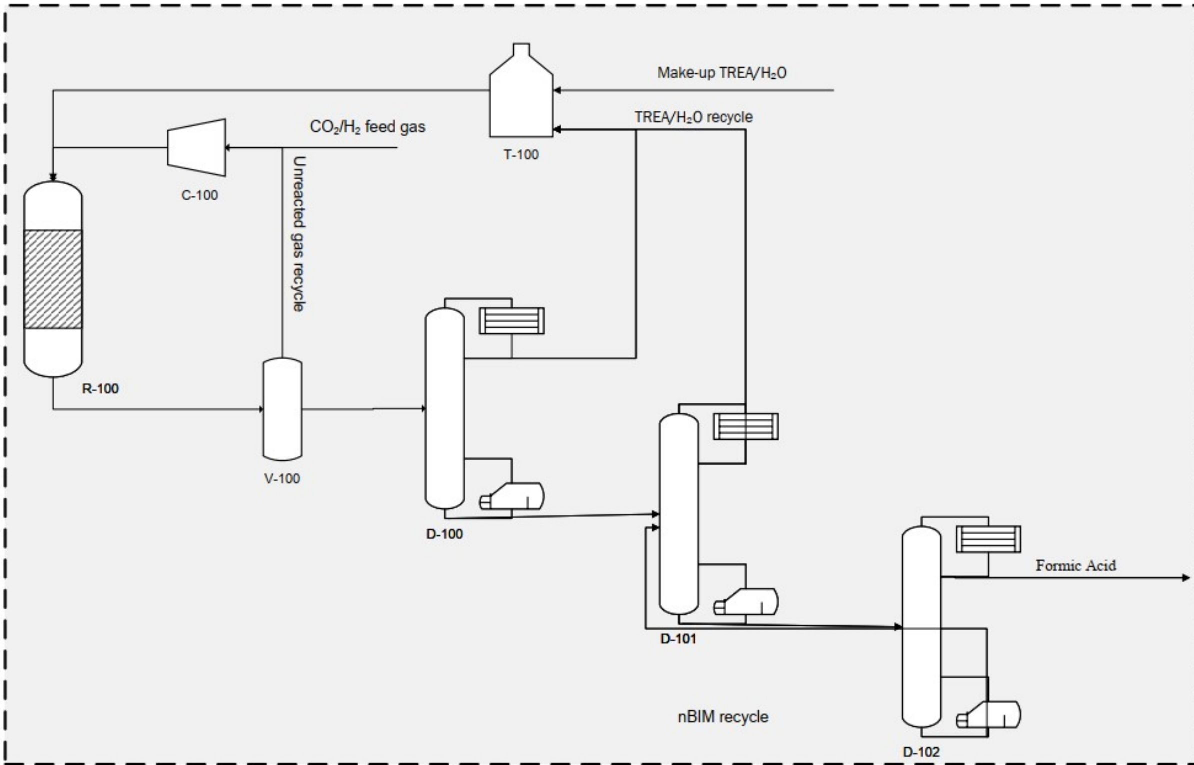

Fig.S6. System boundary of the thermo-catalytic process pathway

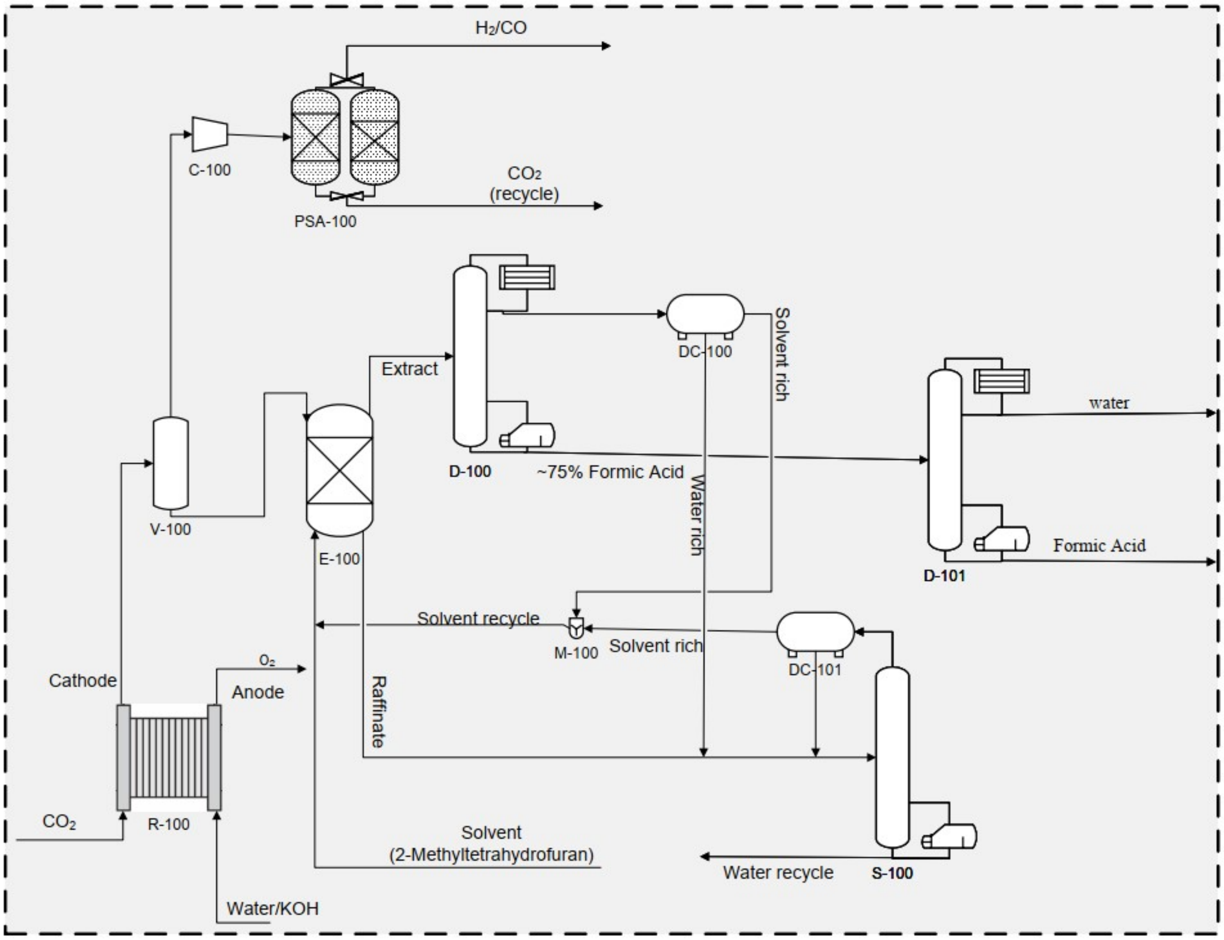

Fig.S7. System boundary of the electrochemical reduction process pathway

Table S6. LCA methodology and GWI values of the conventional pathway compared to previous works.

Reference	This work	N. Thonemann et al (2019) [20]	D. Kang et al (2021) [21]	Y. Ahn et al (2019) [23]
Scope	Cradle to gate	Cradle to gate	Cradle to gate	Cradle to gate
Software	SimaPro LCA	openLCA 1.7.4	SimaPro LCA	SimaPro LCA
Database	Ecoinvent 3.9	_	Ecoinvent 3	_
LCIA method	ReCiPe 2016	ILCD 1.0.8 (2016)	ReCiPe 2008	ReCiPe 1.13
Lifetime horizon of the LCA	100 years (GWP100)	100 years (GWP100)	100 years (GWP100)	100 years (GWP100)
Purity	85.0 wt% FA	85.0 wt% FA	98 wt%	98%
GWI	4.23 $kgCO_{2eq}$/kg FA	4.4 $kgCO_{2eq}$/kg FA	2.04 $kgCO_{2eq}$/kg FA	2.007 $kgCO_{2eq}$/kg FA

Table S7. LCA methodology and GWI values of the thermo-catalytic pathway compared to previous works.

Reference	This work	D. Kang et al (2021) [21]	Y. Ahn et al (2019) [23]
Scope	Cradle to gate	Cradle to gate	Cradle to gate
Software	SimaPro LCA	SimaPro LCA	SimaPro LCA
Database	Ecoinvent 3.9	Ecoinvent 3	_
LCIA method	ReCiPe 2016	ReCiPe 2008	ReCiPe 1.13
Lifetime horizon of the LCA	100 years (GWP100)	100 years (GWP100)	100 years (GWP100)
Purity	85.0 wt% FA	99 wt%	98%
GWI	0.75 to 2.59 kg CO_{2eq}/kg FA	0.27 kg CO_{2eq}/kg FA	0.098 $kgCO_{2eq}$/kg FA

Table S8. LCA methodology and GWI values of the electrochemical reduction pathway compared to previous works.

Reference	This work	A. Dominguez-Ramos et al (2015) [19]	N. Thonemann et al (2019) [20]	D. Kang et al (2021) [21]	A. Banu et al (2023) [22]
Scope	Cradle to gate	Cradle to gate	Cradle to gate	Cradle-to-gate	Cradle-to-gate
Software	SimaPro LCA	_	openLCA 1.7.4	SimaPro LCA	GaBi professional software
Database	Ecoinvent 3.9	Ecoinvent v2.2	Ecoinvent 3.4	Ecoinvent 3	_
LCIA method	ReCiPe 2016	ReCiPe, 2011	ILCD 1.0.8 (2016)	ReCiPe 2008	ReCiPe 1.08
Lifetime horizon of the LCA	100 years (GWP100)	100 years (GWP100)	100 years (GWP100)	100 years (GWP100)	10 years (GWP10)
Purity	85 wt.%	84 wt.% formate solution	85.0 wt.% FA	85 wt.%	_
GWI	2.91 to 8.23 kgCO_{2eq}/kg FA	32 to 519 kg $CO_{2eq.}$/ kg $HCOO^-$	4.2 to 7.2 kg CO2-eq/kg FA	2 kgCO_{2eq}/kg FA	3.27 kg$CO_{2\text{-}eq}$/kg FA

Table S9. Total electricity and heat use per unit of product of the electrochemical reduction pathway

Reference	Electricity (MWh/Tonne FA)	Heat (MJ/Tonne FA)
This work	2.98	13.356
D. Kang et al (2021) [21]	4.79	21.78
R. Aldoco et al (2019)[24]	4.59	62
A. Dominguez-Ramos et al (2015)[19]	8.259-16	35-5169

Table S10. Yield of formic acid for the electrochemical reduction pathway

Reference	CO_2 Conversion (%)	Faraday's efficiency (%)	Formic acid yield (%)
This work	44	90	39.6
Z. Xing et al (2021)[25]	35	83	29.05
Qian et al (2020)[26]	36	81	29.16

Table S11. Heat and mass balance of Conventional pathway

Stream Name	Units	Feed to R-100(CO)	MeOH with Recycle	R-100 to R-101	Water feed to R-101	D-101 to D-103	D-103
Description							
From						D-101	D-103
To		R-100	R-100	R-101	R-101	D-103	
Stream Class		CONVEN	CONVEN	CONVEN	CONVEN	CONVEN	CONVEN
Temperature	C	80	79.8526401	80	80	158.6559053	25
Pressure	bar	40	40	3	3	4.053	4
Enthalpy Flow	cal/sec	-156789008.6	-15053754234	-29683423.11	-382529113.6	-17969715.05	-9235223.017
Average MW		28.0104	28.40887903	41.87809528	18.01528	25.91353893	36.82065841
Mole Flows	kmol/hr	21650	2022439.133	1543.50942	20460	880.1403768	369.6589583
CO	kmol/hr	21650	1996341.702	205.7666268	0	2.91036E-35	0
MF	kmol/hr	0	25015.03052	827.2613747	0	1.59179E-21	0
METHANOL	kmol/hr	0	1082.4	0	0	4.45641E-13	0
WATER	kmol/hr	0	0	510.4814185	20460	631.9619648	121.4805463
FA	kmol/hr	0	0	3.00921E-09	0	248.178412	248.178412
Mole Fractions							
CO		1	0.987096061	0.133310898	0	3.3067E-38	0
MF		0	0.012368743	0.535961338	0	1.80857E-24	0
METHANOL		0	0.000535195	0	0	5.06329E-16	0
WATER		0	0	0.330727764	1	0.718024058	0.328628709
FA		0	0	1.94959E-12	0	0.281975942	0.671371291
Mass Fractions							
CO		1	0.973250493	0.089165745	0	3.57427E-38	0
MF		0	0.026145864	0.768560514	0	4.19121E-24	0

METHANOL	0	0.000603643	0	0	6.26078E-16	0
WATER	0	0	0.142273741	1	0.499175527	0.150788494
FA	0	0	2.14267E-12	0	0.500824473	0.849211506

Table S12. Heat and mass balance of Thermo-catalytic pathway

Stream Name	**Units**	**CO_2**	**H_2**	**TREA**	**H_2O**	**R-100 feed with recycle**	**From R-100 to V-100**
Description							
From							R-100
To		C100	C100	T-100	T-100	R-100	V-100
Stream Class			CONVEN	CONVEN	CONVEN	CONVEN	CONVEN
Temperature	C	25	25	25	25	120	120
Pressure	bar	1	30	120	120	120	120
Enthalpy Flow	cal/sec	-15990156.5	1025.9174	-3933304	-62473484	-79591740.11	-81137215.78
Average MW		44	2	101	18	24.81784387	28.26006907
Mole Flows	kmol/hr	612	612	324	3294	4842	4254.48
CO_2	kmol/hr	612	0	0	0	612	318.24
H_2	kmol/hr	0	612	0	0	612	318.24
H_2O	kmol/hr	0	0	0	3294	3294	3294
N_2	kmol/hr	0	0	0	0	0	0
O_2	kmol/hr	0	0	0	0	0	0

NTRIET	kmol/hr	0	0	0	0	0	0
TREA	kmol/hr	0	0	324	0	324	30.24
AMMON-01	kmol/hr	0	0	0	0	0	0
HCOOH	kmol/hr	0	0	0	0	0	0
TREA-FO	kmol/hr	0	0	0	0	0	293.76
N-ISO-01	kmol/hr	0	0	0	0	0	0
Mole Fractions							
CO_2		1	0	0	0	0.126394052	0.074801151
H_2		0	1	0	0	0.126394052	0.074801151
H_2O		0	0	0	1	0.680297398	0.774242681
N_2		0	0	0	0	0	0
O_2		0	0	0	0	0	0
NTRIET		0	0	0	0	0	0
TREA		0	0	1	0	0.066914498	0.007107802
AMMON-01		0	0	0	0	0	0
HCOOH		0	0	0	0	0	0
TREA-FO		0	0	0	0	0	0.069047216
N-ISO-01		0	0	0	0	0	0
Mass Fractions							
CO_2		1	0	0	0	0.224086279	0.116462937

H_2	0	1	0	0	0.01018574	0.00529377
H_2O	0	0	0	1	0.493409227	0.493146999
N_2	0	0	0	0	0	0
O_2	0	0	0	0	0	0
NTRIET	0	0	0	0	0	0
TREA	0	0	1	0	0.272318754	0.025402909
AMMON-01	0	0	0	0	0	0
HCOOH	0	0	0	0	0	0
TREA-FO	0	0	0	0	0	0.359693385
N-ISO-01	0	0	0	0	0	0

Stream Name	**Units**	**V-100 to D-100**	**D-100 to D-101**	**nBIM**	**D-101 to D-102**	**D-102 Distillate**
Description						
From		V-100	D-100	D-102	D-101	D-102
To		D-100	D-101	D-101	D-102	
Stream Class		CONVEN	CONVEN	CONVEN	CONVEN	CONVEN
Temperature	C	100	34.61082716	25	100	29.11059499
Pressure	bar	120	0.199983553	0.199983553	0.133322368	0.066661184
Enthalpy Flow	cal/sec	-74789011.65	-11598123.81	-1048297.032	-9558658.557	-8686064.137
Average MW		27.98776683	66.62370624	124.18576	76.24208293	37.50160635

Mole Flows	kmol/hr	3912.356832	661.8297544	276	628.3459354	356.4989791
CO_2	kmol/hr	265.203606	0	0	0	0.37497292
H_2	kmol/hr	0.012593845	0	0	0	2.16122E-11
H_2O	kmol/hr	3077.64008	92.32920241	0	92.26638277	108.402864
N_2	kmol/hr	0	0	0	0	0
O_2	kmol/hr	0	0	0	0	0
NTRIET	kmol/hr	0	0	0	0	0
TREA	kmol/hr	295.0464022	295.0464022	0	9.17039E-21	0
AMMON-01	kmol/hr	0	0	0	0	0
HCOOH	kmol/hr	274.4541498	274.4541498	0	260.0795527	247.7211422
TREA-FO	kmol/hr	0	0	0	0	0
N-ISO-01	kmol/hr	0	0	276	276	6.2393E-224
Mole Fractions						
CO_2		0.06778615	0	0	0	0.00105182
H_2		3.21899E-06	0	0	0	6.06236E-14
H_2O		0.786646058	0.139505971	0	0.146840104	0.304076226
N_2		0	0	0	0	0
O_2		0	0	0	0	0
NTRIET		0	0	0	0	0
TREA		0.075413981	0.445804076	0	1.45945E-23	0

AMMON-01	0	0	0	0	0
HCOOH	0.070150592	0.414689953	0	0.413911411	0.694871954
TREA-FO	0	0	0	0	0
N-ISO-01	0	0	1	0.439248485	1.7502E-226
Mass Fractions					
CO_2	0.106567652	0	0	0	0.001234083
H_2	2.30028E-07	0	0	0	3.23312E-15
H_2O	0.505922074	0.0376909	0	0.034667493	0.145950336
N_2	0	0	0	0	0
O_2	0	0	0	0	0
NTRIET	0	0	0	0	0
TREA	0.272147902	0.675828684	0	1.93337E-23	0
AMMON-01	0	0	0	0	0
HCOOH	0.115362142	0.286480416	0	0.249869277	0.852815581
TREA-FO	0	0	0	0	0
N-ISO-01	0	0	1	0.71546323	5.7956E-226

Table S13. Heat and mass balance of Electrochemical reduction pathway

Stream Name	**Units**	**CO_2**	**water/KOH**	**R-100 to V-100**	**O_2**	**Solvent**
Description						
From				R-100	R-100	

To		R-100	R-100	V-100		E-100
Stream Class		CONVEN	CONVEN	CONVEN	CONVEN	CONVEN
Temperature	C	25	25	25	25	40
Pressure	bar	1	1	1	81.00000278	1
Enthalpy Flow	cal/sec	-7637021.468	-155085860.9	-165358205.7	2.31099E-11	-27461733.5
Average MW		44.0098	18.08527022	19.92982037	31.9988	86.1338
Mole Flows	kmol/hr	292.3261806	8163.360481	8561.018596	187.004873	1620
H_2O	kmol/hr	0	8148.360481	7780.472928	0	0
CO_2	kmol/hr	292.3261806	0	391.4228634	0	0
HCOOH	kmol/hr	0	0	260.6421236	0	0
O_2	kmol/hr	0	0	0	187.004873	0
CO	kmol/hr	0	0	6.122193882	0	0
H_2	kmol/hr	0	0	107.3584858	0	0
KOH	kmol/hr	0	15.00000049	15.00000049	0	0
2-MET-01	kmol/hr	0	0	0	0	1620
Mole Fractions						
H_2O		0	0.998162521	0.908825608	0	0
CO_2		1	0	0.045721529	0	0
HCOOH		0	0	0.030445223	0	0
O_2		0	0	0	1	0
CO		0	0	0.000715124	0	0
H_2		0	0	0.012540387	0	0
KOH		0	0.001837479	0.001752128	0	0
2-MET-01		0	0	0	0	1
Mass Fractions						
H_2O		0	0.99429962	0.821520089	0	0
CO_2		1	0	0.100964049	0	0
HCOOH		0	0	0.070309821	0	0
O_2		0	0	0	1	0

CO	0	0	0.001005073	0	0
H_2	0	0	0.001268447	0	0
KOH	0	0.00570038	0.004932521	0	0
2-MET-01	0	0	0	0	1

Stream Name	**Units**	**V-100 to E-100**	**E-100 to D-100**	**D-100 to D-101**	**D-101**
Description					
From		V-100	E-100	D-100	D-101
To		E-100	D-100	D-101	
Stream Class		CONVEN	CONVEN	CONVEN	CONVEN
Temperature	C	40	45.88817494	114.8619566	25
Pressure	bar	1	1.1	5.1	4
Enthalpy Flow	cal/sec	-152321371.8	-50598505.48	-11326836.34	-8431871.724
Average MW		19.03372859	58.65799418	32.8046018	41.49039323
Mole Flows	kmol/hr	7946.903461	2707.793959	514.4808522	324.1229369
H_2O	kmol/hr	7662.107075	926.5722468	248.322803	57.96489617
CO_2	kmol/hr	14.23911279	14.01372085	5.80619E-50	0
HCOOH	kmol/hr	255.3933502	255.1616312	250.916971	250.9169625
O_2	kmol/hr	0	0	0	0
CO	kmol/hr	0.037354884	0.023749338	4.43649E-64	0
H_2	kmol/hr	0.126567157	0.070387709	3.38629E-74	0
KOH	kmol/hr	15.00000049	14.99999804	15.24107824	15.24107824
2-MET-01	kmol/hr	0	1496.952225	3.3195E-28	0
Mole Fractions					
H_2O		0.964162597	0.342187131	0.482666754	0.178836144
CO_2		0.001791781	0.005175328	1.12855E-52	0
HCOOH		0.032137467	0.094232292	0.487709056	0.774141333

O_2	0	0	0	0
CO	4.70056E-06	8.77073E-06	8.62323E-67	0
H_2	1.59266E-05	2.59945E-05	6.58196E-77	0
KOH	0.001887528	0.005539564	0.029624189	0.047022523
2-MET-01	0	0.55283092	6.45214E-31	0
Mass Fractions				
H_2O	0.912572598	0.105093893	0.26506576	0.077651306
CO_2	0.004142958	0.003882934	1.51404E-52	0
HCOOH	0.077711983	0.073938862	0.684268052	0.858762198
O_2	0	0	0	0
CO	6.91743E-06	4.18821E-06	7.36299E-67	0
H_2	1.6868E-06	8.93344E-07	4.04469E-78	0
KOH	0.005563857	0.005298524	0.050666188	0.063586497
2-MET-01	0	0.811780705	1.69411E-30	0

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