Electronic Supplementary Information (ESI)

Artificial intelligence: the silver bullet for sustainable materials development

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1. General

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1.3. List of variables and abbreviations

The list of variables and abbreviations used in this research are detailed in **Table S1** and **S2**.

Table S1. Variables, explanation, and unit.

Variables	Explanation	Unit
c_e	Concentration of electrolyte	M
$c_{e,n}$	Normalized concentration of electrolyte	-
c_l	Concentration of linker	M
$c_{l,n}$	Normalized concentration of linker	-
V	Applied voltage	V
V_n	Normalized applied voltage	-
t	Reaction time	h
t_n	Normalized reaction time	-
Overall input parameter	The average from the combination of the normalized input parameters	-
Crystallinity	Crystallinity	%
$Crystallinity_n$	Normalized crystallinity	-
Purity	Purity	%
$Purity_n$	Normalized purity	-
Yield	Yield	%
$Yield_n$	Normalized yield	-
E-factor	E-factor	kg kg^{-1}
$E\text{-}factor_n$	Normalized E-factor	-
Energy	Energy	kWh kg^{-1}
$Energy_n$	Normalized energy	-
Carbon footprint	Carbon footprint	kg kg^{-1}
$Carbon\ footprint_n$	Normalized carbon footprint	-
$f_{c_{e,n}}$	Normalized factor contribution of electrolyte concentration	-
$f_{c_{e,n}}$	Normalized factor contribution of applied voltage	-
$f_{c_{e,n}}$	Normalized factor contribution of reaction time	-
$f_{c_{e,n}}$	Normalized factor contribution of linker concentration	-
m_e	Mass of electrolyte salt	g
m_l	Mass of linker	g
d_i	Individual desirability function	-
D	Global desirability function	-

Table S2. Abbreviation and explanation.

Abbreviation	Explanation
AI	Artificial intelligence
ANOVA	Analysis of variance
BET	Brunauer–Emmett–Teller theory for surface area measurement
CCC	Central composite circumscribed
CCF	Central composite design face-centered
CCI	Central composite inscribed
DoE	Design of experiment
TGA	Thermogravimetric Analysis
ML	Machine learning
MOF	Metal organic framework
RF	Random forest algorithm
SEM	Scanning electron microscopy
SSD	Squared standard deviation
SVM	Support vector machine algorithm
XRD	X-ray diffraction
ZIF	Zeolitic imidazolate framework

2. From experimental design to artificial intelligence

Synergistic combination of Design of experiments (DoE) and Artificial intelligence (AI) methodologies in the optimization of hyperdimensional systems is presented in **Fig. S1**. This flow chart is an extension of **Fig. 1**. The red dashed box covers the steps in performing DoE. The blue dashed box contains the steps in developing a surrogate function where the support vector machine (SVM) was chosen based on the validation result. The yellow dashed box contains the steps of implementing an evolutionary algorithm. In AI Module 1, the SVM generates excessive virtual datasets followed by the desirability function to find the optimum condition. AI module 2 implements an evolutionary algorithm on 50 initial virtual datasets from the SVM to be followed by the desirability function for final optimization.

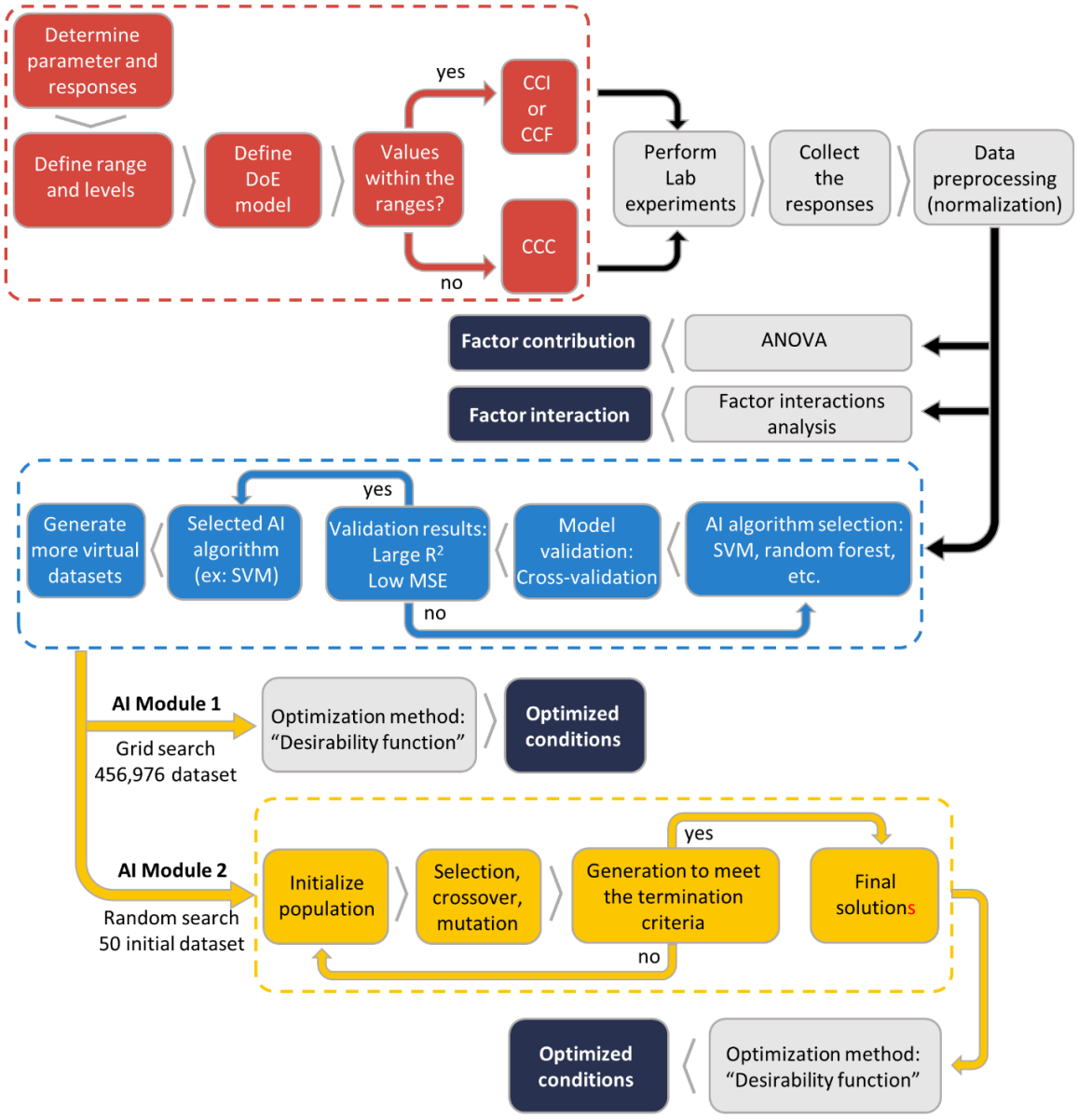

Fig. S1. General flow-chart. Synergistic combination of Design of Experiments (DoE) and Artificial Intelligence (AI) methodologies in the optimization of hyperdimensional systems. An extension from **Fig. 1**. DoE = design of experiment, CCI = central composite inscribed, CCC = central composite circumscribed, CCF = central composite face-centered, ANOVA = analysis of variance, SVM = support vector machine, MSE = mean squared error.

2.1. Data presentation and normalization

The setting parameters and their factor levels are presented in **Table S3**. CCF-DoE methodology from the four parameters and three factor levels generated 27 experiments, including repetition, as tabulated in **Table S4**.

Table S3. Setting parameters. The minimum, maximum, and intermediate values of the input parameters.

Parameters	Unit	Factor levels		
c_e	M	0.01	0.155	0.3
V	V	2	11	20
t	h	0.2	0.6	1
c_l	M	0.5	0.125	0.2

Table S4. Experimental parameters and responses. [a] the reactions did not produce any precipitate at all, [b] the reactions generated amorphous products. The obtained minimum[c] and maximum[d] values of each parameter and response are given at the end of the table.

Entries	Parameters				Product quality			Process sustainability		
	C_e (M)	V (V)	t (h)	C_l (M)	Crystallinity (%)	Purity (%)	Yield (%)	E-factor (kg kg^{-1})	Energy (kWh kg^{-1})	Carbon footprint (kg kg^{-1})
1[a]	0.010	2	0.2	0.50	0.00	0.00	0.00	na.	na.	na.
2[b]	0.300	2	0.2	0.50	0.00	0.00	0.00	na.	na.	na.
3[b]	0.010	20	0.2	0.50	0.00	0.00	0.00	na.	na.	na.
4[b]	0.300	20	0.2	0.50	0.00	0.00	0.00	na.	na.	na.
5	0.010	2	1.0	0.50	29.78	75.16	33.97	24.01	0.84	51.33
6[b]	0.300	2	1.0	0.50	0.00	0.00	0.00	na.	na.	na.
7[b]	0.010	20	1.0	0.50	0.00	0.00	0.00	na.	na.	na.
8	0.300	20	1.0	0.50	6.51	23.12	16.60	13.29	28.23	43.46
9[a]	0.010	2	0.2	2.00	0.00	0.00	0.00	na.	na.	na.
10	0.300	2	0.2	2.00	88.46	92.83	15.04	251.80	3.09	491.92
11	0.010	20	0.2	2.00	70.96	94.17	24.30	105.16	19.52	239.67
12	0.300	20	0.2	2.00	40.14	74.41	18.23	145.03	25.45	302.03
13[a]	0.010	2	1.0	2.00	0.00	0.00	0.00	na.	na.	na.
14	0.300	2	1.0	2.00	19.99	39.40	27.03	56.37	1.75	110.93
15	0.010	20	1.0	2.00	100.00	97.60	77.00	6.71	6.14	19.16
16	0.300	20	1.0	2.00	26.92	61.04	51.03	11.90	9.20	30.32
17	0.155	11	0.6	1.25	51.83	66.55	43.24	16.14	5.91	36.47
18	0.155	11	0.6	1.25	51.02	67.48	48.39	15.78	5.71	35.60
19	0.155	11	0.6	1.25	51.45	65.47	47.21	16.10	5.98	36.45
20	0.010	11	0.6	1.25	71.30	77.65	41.62	22.90	6.35	53.76
21	0.155	2	0.6	1.25	47.56	66.02	39.15	40.23	1.20	80.33
22	0.155	20	0.6	1.25	42.32	77.39	71.05	8.39	6.64	21.71
23	0.155	11	0.2	1.25	67.27	87.09	45.55	38.16	5.69	79.75
24	0.155	11	1.0	1.25	43.28	64.36	59.59	7.12	4.35	17.41
25	0.155	11	0.6	0.50	12.84	48.91	43.09	6.50	6.02	16.20
26	0.155	11	0.6	2.00	35.09	77.48	65.62	14.61	3.95	32.75
27	0.300	11	0.6	1.25	13.88	49.21	41.24	17.57	6.29	37.32
min[c]	0.01	2	0.20	0.50	0.00	0.00	0.00	6.50	0.84	16.20
max[d]	0.30	20	1.00	2.00	100.00	100.00	100.00	251.80	28.23	491.92

It can be seen from **Table S3** that the factor levels for each parameter can be observed. **Table S4** gives the results of 27 experiments generated by the CCF-DoE methodology and the experiments'

corresponding responses. To facilitate the comparison of the obtained data during graphical visualization, both parameters and responses were also presented in the normalized format, which allowed all the data to be shown on the same scale. A min-max normalization strategy,[1] where the lowest and the highest of the actual value is weighted as zero and one, respectively, was applied in this work. The min-max equation is shown in equation (S1):

$$x_n = \frac{(x - x_{min})}{(x_{max} - x_{min})} \quad \text{(S1)}$$

where x_n is the normalized value, x is the actual value of a parameter or response, x_{min} is the minimum actual value of this parameter or response, and x_{max} is the maximum actual value of this parameter or response. The overall input parameter is defined as the average from the combination of the normalized input parameters, which is calculated according to equation (S1-b):

$$Overall\ input\ parameter = \frac{(c_{e,n} + c_{l,n} + t_n + V_n)}{4} \quad \text{(S1-b)}$$

where $c_{e,n}$, $c_{l,n}$, t_n and V_n is normalized concentration of electrolyte, normalized concentration of linker, normalized reaction time, and normalized applied voltage, respectively.

The normalized datasets are presented in **Table S5**. The normalized parameters and responses are also presented in bar charts to facilitate comparison, as presented in **Fig. S2-S4**.

Table S5. Normalized experimental parameters and responses. [a] the reactions did not produce any precipitate at all, [b] the reactions generated amorphous products.

Entries	Normalized parameters				Overall input parameters	Normalized product quality			Normalized process sustainability		
	$c_{e,n}$	V_n	t_n	$c_{l,n}$		$Crystallinity_n$	$Purity_n$	$Yield_n$	$E\text{-}factor_n$	$Energy_n$	$Carbon\ footprint_n$
1[a]	0.0	0.0	0.0	0.0	0.00	0.000	0.000	0.000	na.	na.	na.
2[b]	1.0	0.0	0.0	0.0	0.25	0.000	0.000	0.000	na.	na.	na.
3[b]	0.0	1.0	0.0	0.0	0.25	0.000	0.000	0.000	na.	na.	na.
4[b]	1.0	1.0	0.0	0.0	0.50	0.000	0.000	0.000	na.	na.	na.
5	0.0	0.0	1.0	0.0	0.25	0.298	0.752	0.340	0.071	0	0.074
6[b]	1.0	0.0	1.0	0.0	0.50	0.000	0.000	0.000	na.	na.	na.
7[b]	0.0	1.0	1.0	0.0	0.50	0.000	0.000	0.000	na.	na.	na.
8	1.0	1.0	1.0	0.0	0.75	0.065	0.231	0.166	0.028	1	0.057
9[a]	0.0	0.0	0.0	1.0	0.25	0.000	0.000	0.000	na.	na.	Na.
10	1.0	0.0	0.0	1.0	0.50	0.885	0.928	0.150	1	0.082	1
11	0.0	1.0	0.0	1.0	0.50	0.710	0.942	0.243	0.402	0.682	0.47
12	1.0	1.0	0.0	1.0	0.75	0.401	0.744	0.182	0.565	0.899	0.601
13[a]	0.0	0.0	1.0	1.0	0.50	0.000	0.000	0.000	na.	na.	Na.
14	1.0	0.0	1.0	1.0	0.75	0.200	0.394	0.270	0.203	0.033	0.199
15	0.0	1.0	1.0	1.0	0.75	1.000	0.976	0.770	0.001	0.194	0.006
16	1.0	1.0	1.0	1.0	1.00	0.269	0.610	0.510	0.022	0.306	0.03
17	0.5	0.5	0.5	0.5	0.50	0.518	0.666	0.432	0.039	0.185	0.043
18	0.5	0.5	0.5	0.5	0.50	0.510	0.675	0.484	0.038	0.178	0.041
19	0.5	0.5	0.5	0.5	0.50	0.514	0.655	0.472	0.039	0.188	0.043
20	0.0	0.5	0.5	0.5	0.37	0.713	0.776	0.416	0.067	0.201	0.079
21	0.5	0.0	0.5	0.5	0.37	0.476	0.66	0.391	0.138	0.013	0.135
22	0.5	1.0	0.5	0.5	0.62	0.423	0.774	0.710	0.008	0.212	0.012
23	0.5	0.5	0.0	0.5	0.37	0.673	0.871	0.456	0.129	0.177	0.134
24	0.5	0.5	1.0	0.5	0.62	0.433	0.644	0.596	0.003	0.128	0.003
25	0.5	0.5	0.5	0.0	0.37	0.128	0.489	0.431	0	0.189	0
26	0.5	0.5	0.5	1.0	0.62	0.351	0.775	0.656	0.033	0.114	0.035
27	1.0	0.5	0.5	0.5	0.62	0.139	0.492	0.412	0.045	0.199	0.044

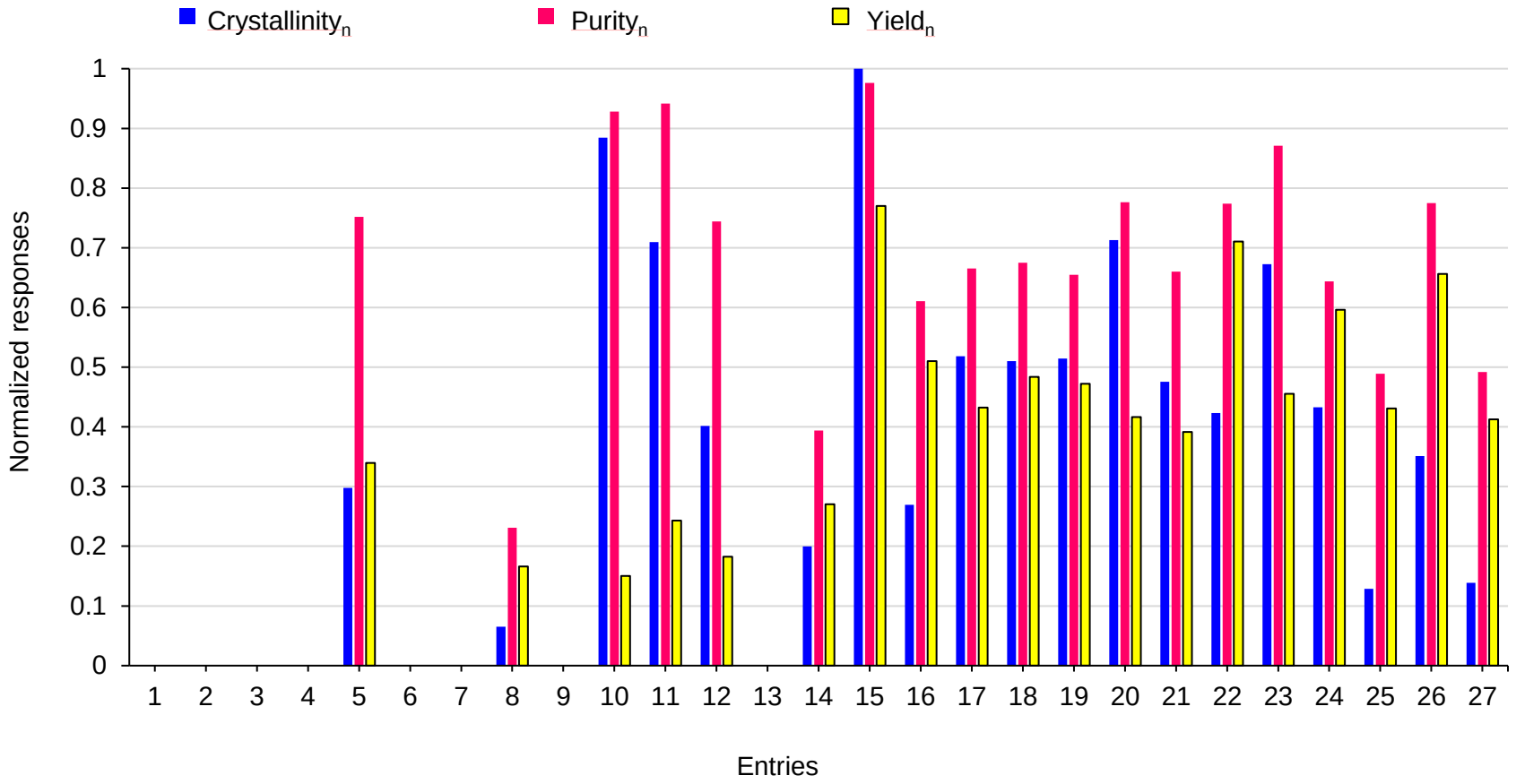

Fig. S2. Normalized product quality responses (refer to **Table S5**). The corresponding true product quality values are shown in **Table S4**.

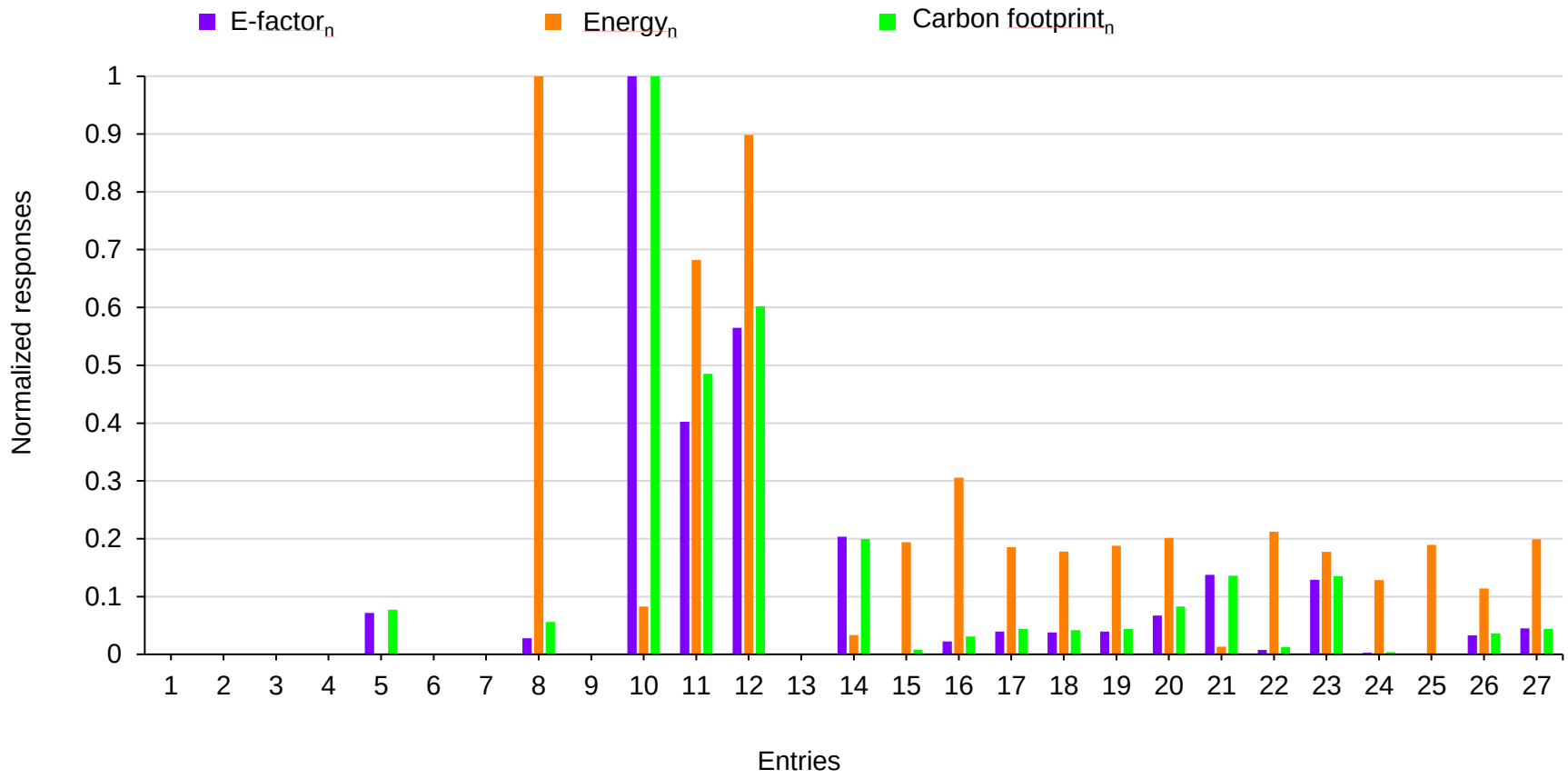

Fig. S3. Normalized process sustainability responses (refer to **Table S5**). The corresponding true product quality values are shown in **Table S4**.

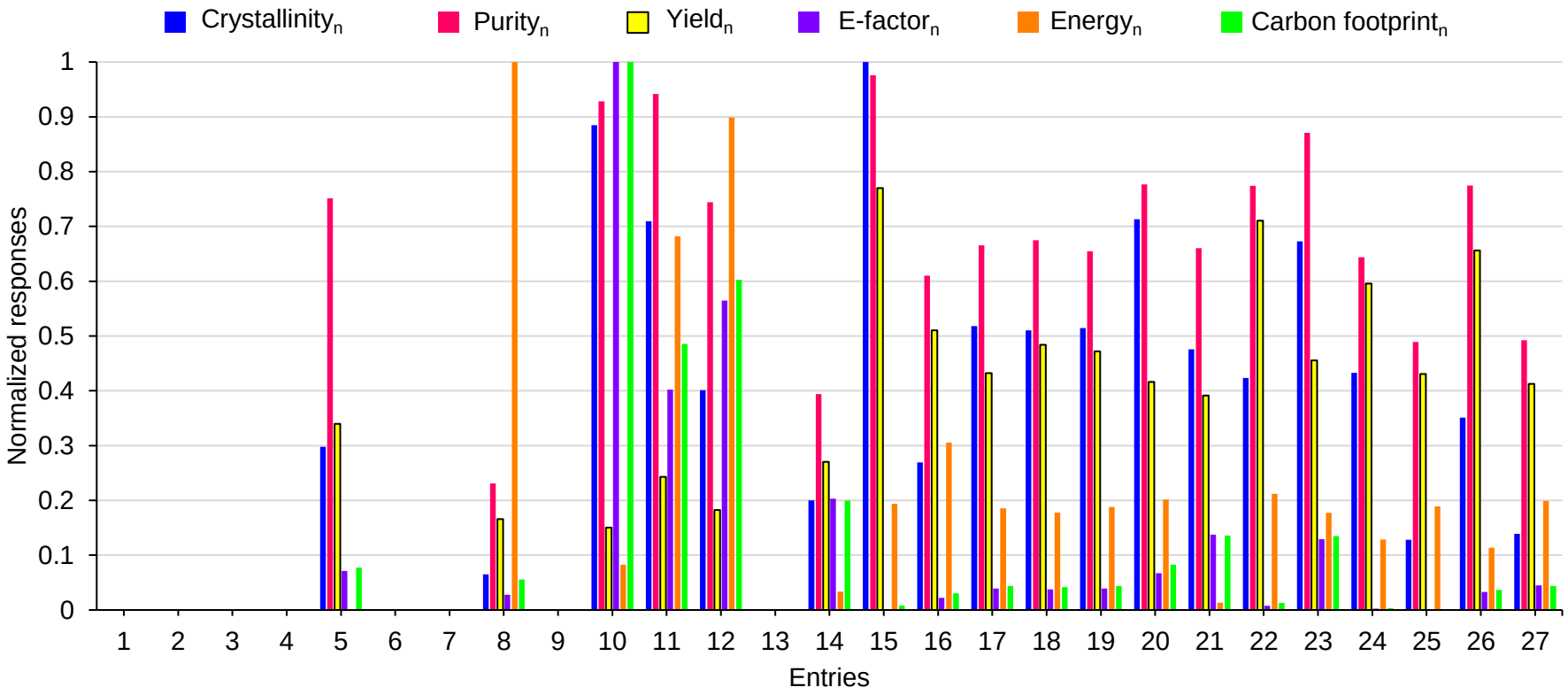

Fig. S4. Normalized product quality and process sustainability responses (refer to **Table S5**). The corresponding true values are shown in **Table S4**.

2.2. ANOVA analysis for determining factor contributions

To find out the extent of the impact of the input variables on each response, the sum of squared deviation (SSD) was calculated. The SSD of parameter i is defined by equation (S2):

$$SSD = \sum_{j=1}^{N} (\bar{x}_{i,j} - \bar{x}_{grand})^2 \quad \text{(S2)}$$

where $\bar{x}_{i,j}$ is the mean value of one response when input variable i is at level j, $\bar{x}_{grand}$ is the grand mean value of one response. There are four different input variables at three different levels, and therefore N = 3 in equation (S2) (see **Table S3**). The percentage contribution of normalized input variable i can be obtained from equation (S3):

$$f_{i,n} = \frac{SSD}{\sum_{i=1}^{K} SSD} \quad \text{(S3)}$$

where K is the total number of input variables, and n indicates normalization.

The factor contribution is presented relative to the total contribution (normalized), as shown in **Table S6** and illustrated in **Fig. S5**. The factor contribution is also presented in Pareto charts, as depicted in **Fig. S6-S7**. The Pareto chart gives information on individual parameter contribution as well as cumulative contribution.

Table S6. Factor contributions for each response in a normalized format. For a visual comparison of the data, see the radar chart under **Fig. S5**.

	$Crystallinity_n$	$Purity_n$	$Yield_n$	$E\text{-}factor_n$	$Energy_n$	$Carbon\ footprint_n$
$f_{c_{e,n}}$	0.15	0.19	0.25	0.12	0.14	0.13
f_{V_n}	0.16	0.20	0.23	0.18	0.55	0.16
f_{T_n}	0.08	0.14	0.25	0.53	0.19	0.55
$f_{c_{l,n}}$	0.61	0.47	0.27	0.17	0.12	0.16
SUM	1.00	1.00	1.00	1.00	1.00	1.00

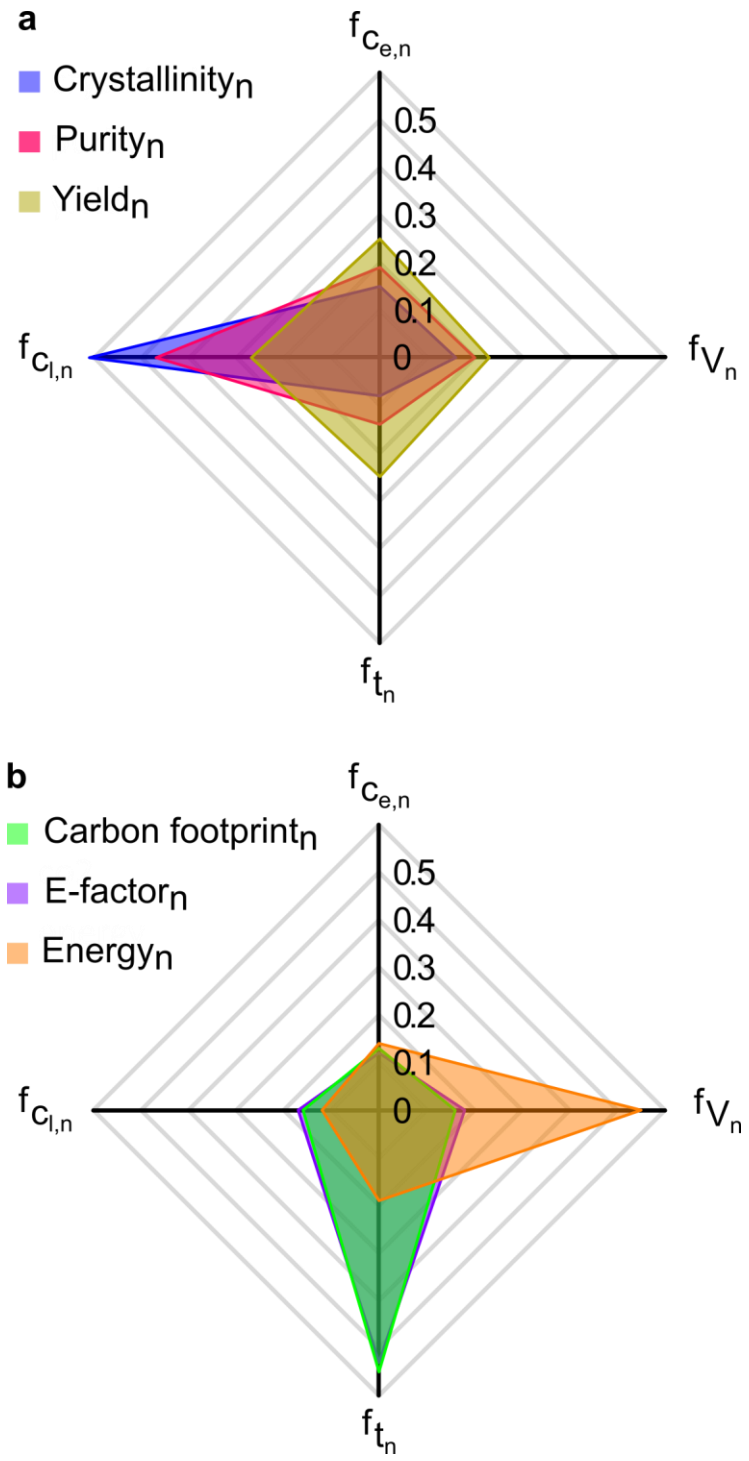

Fig. S5. Factor contributions. Factor contribution for each response under product quality (**a**) and process sustainability (**b**). The actual factor contributions values are provided in **Table S6**.

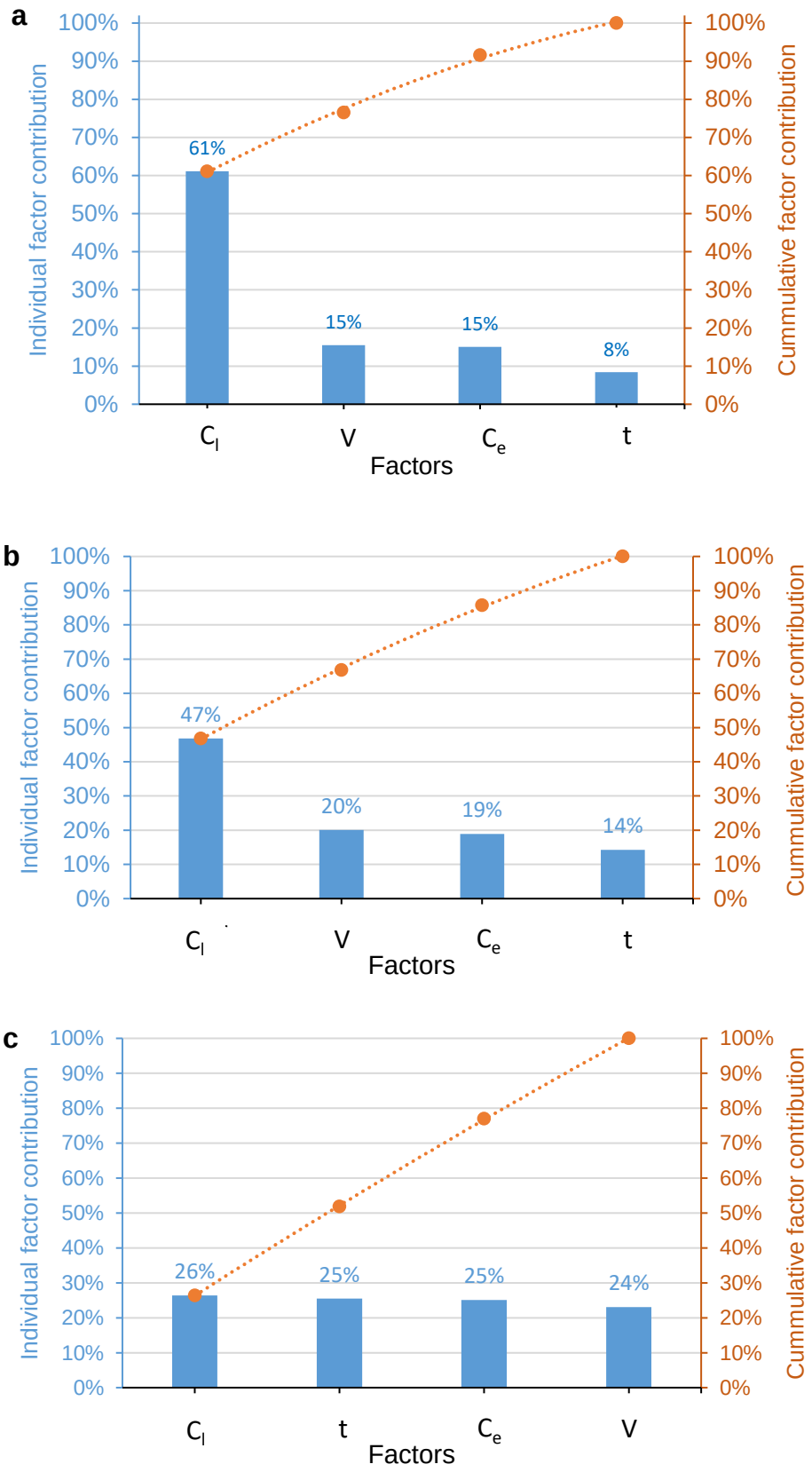

Fig. S6. Pareto charts product quality. The factor contribution of the product quality metrics, namely crystallinity (**a**), purity (**b**), and yield (**c**). The blue axis represents individual factor contribution; the orange axis represents cumulative factor contribution.

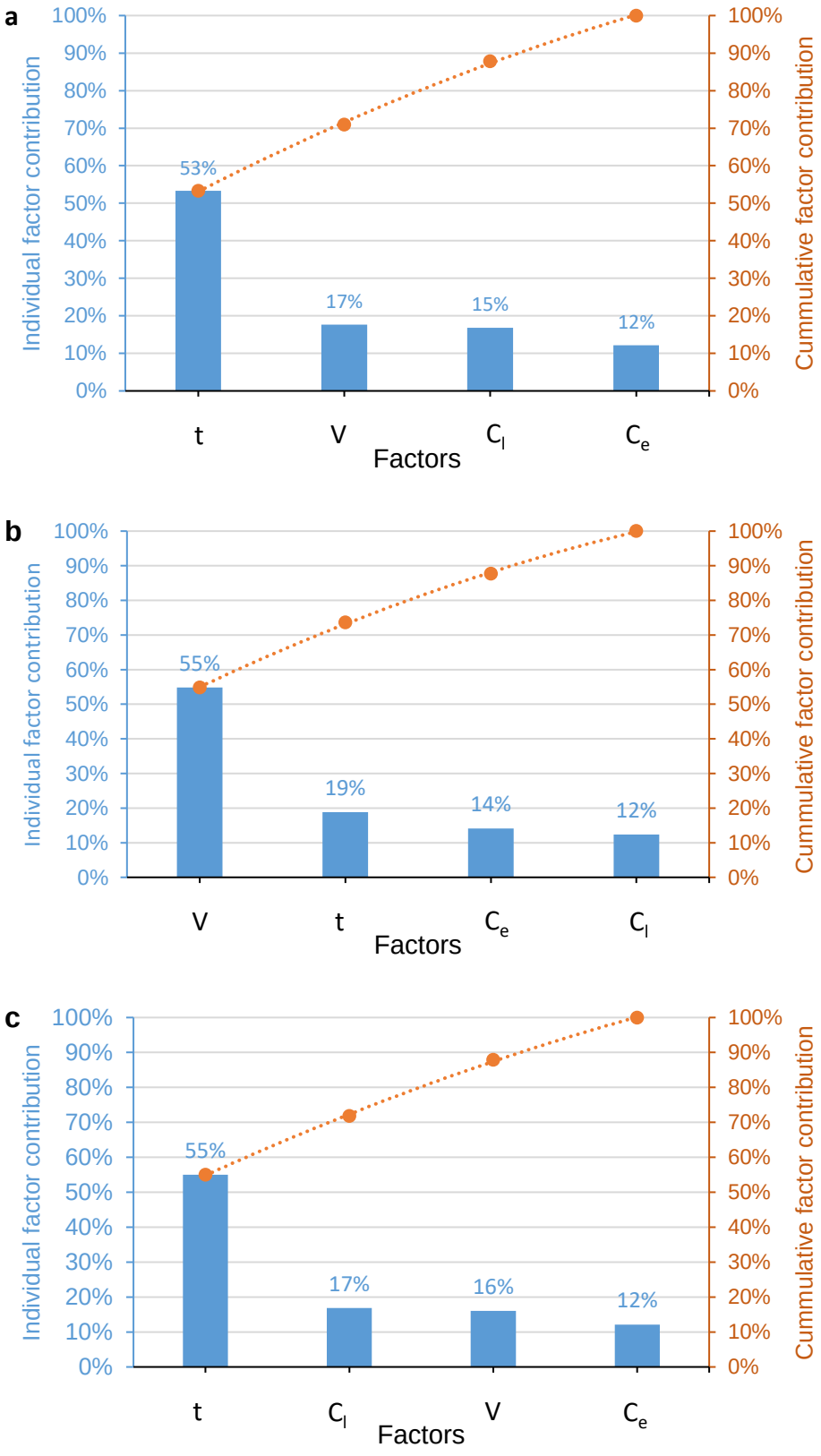

Fig. S7. Pareto charts process sustainability. The factor contribution of the process sustainability metrics, namely E-factor (**a**), energy (**b**), and carbon footprint (**c**). The blue axis represents individual factor contribution, orange axis represents cumulative factor contribution.

2.3. Individual correlation (main effect) calculations

The individual correlation plot (also called the main effect) is used to compare the relative strength of the effects of the parameters. The sign of a main effect informs the direction of the effect, and the magnitude of a main effect indicates the strength of the effect. The methodology was adapted from the literature.[2] To determine the main effect, the lowest and the highest values in each parameter were grouped, and then the responses from each dataset (lowest dataset and highest dataset) were averaged. Individual correlation between each parameter and each response was computed. The results are presented in **Fig. 2e**.

An example is given in **Fig. S8** to calculate the main effect of the electrolyte on crystallinity. The plot between electrolyte concentration and crystallinity is also presented, demonstrating that an increase in electrolyte concentration from 0.01 M to 0.3 M causes an approx. 10% decrease in the average crystallinity. By applying the same calculation principle, the individual correlation between each parameter and each response was computed. The results are presented in **Fig. 2e**.

Entries	c_e (M)	V (V)	t (h)	C_l (M)	Crystallinity (%)
1	0.01	2	0.2	0.5	0.00
3	0.01	20	0.2	0.5	0.00
5	0.01	2	1	0.5	29.78
7	0.01	20	1	0.5	0.00
9	0.01	2	0.2	2	0.00
11	0.01	20	0.2	2	70.96
13	0.01	2	1	2	0.00
15	0.01	20	1	2	100.00
20	0.01	11	0.6	1.25	71.30
17	0.155	11	0.6	1.25	51.83
18	0.155	11	0.6	1.25	51.02
19	0.155	11	0.6	1.25	51.45
21	0.155	2	0.6	1.25	47.56
22	0.155	20	0.6	1.25	42.32
23	0.155	11	0.2	1.25	67.27
24	0.155	11	1	1.25	43.28
25	0.155	11	0.6	0.5	12.84
26	0.155	11	0.6	2	35.09
2	0.3	2	0.2	0.5	0.00
4	0.3	20	0.2	0.5	0.00
6	0.3	2	1	0.5	0.00
8	0.3	20	1	0.5	6.51
10	0.3	2	0.2	2	88.46
12	0.3	20	0.2	2	40.14
14	0.3	2	1	2	19.99
16	0.3	20	1	2	26.92
27	0.3	11	0.6	1.25	13.88

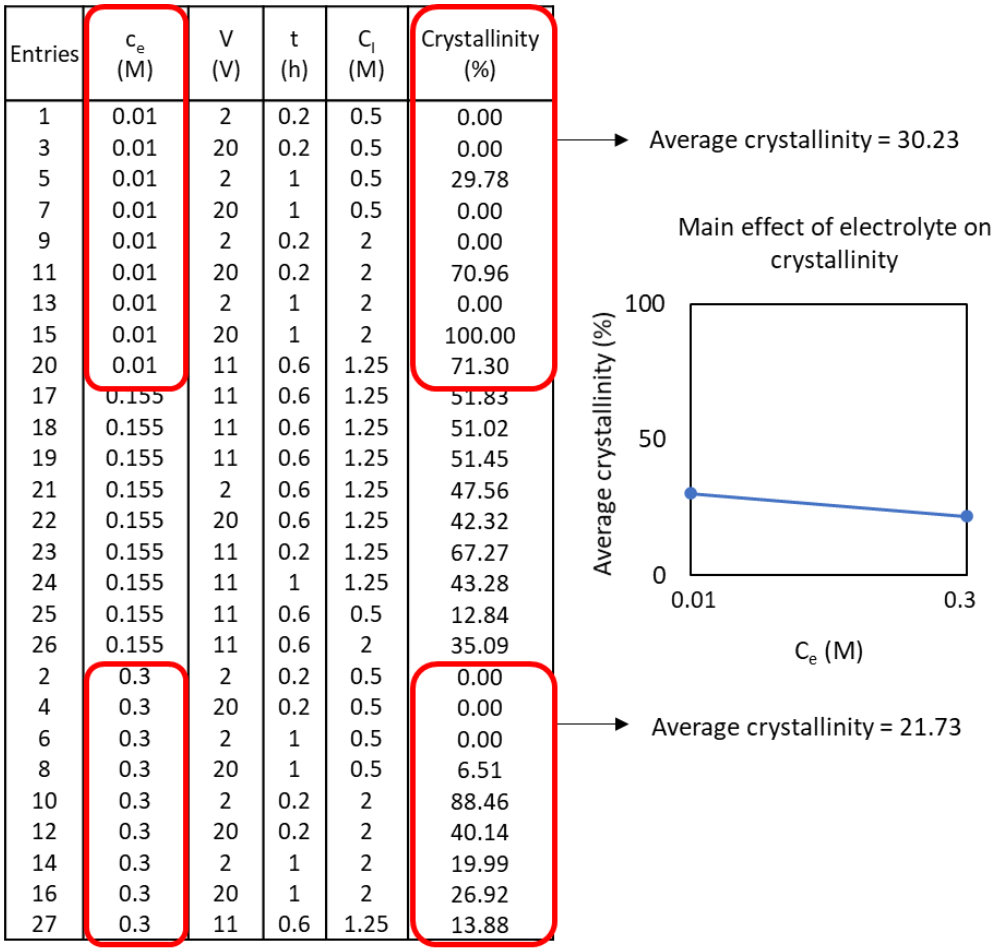

Fig. S8. Calculation and plot for the individual correlation of the electrolyte on the crystallinity.

2.4. Two-factor-responses interactions calculations

The interaction of parameters can influence the quality of the final product. The two-factor-responses interactions for all factors and responses are given in **Fig. S10-S15**. As an example, the interaction between electrolyte concentration (C_e) and voltage (V) on crystallinity is given in **Fig. S9**. To study the interaction between C_e and V, it is important to calculate the average crystallinity values for the four possible combinations of C_e at the lowest value (Low C_e), C_e at the highest value (High C_e), V at the lowest value (Low V), and V at the highest value (High V). Parallel lines in the interaction plot indicate no interaction, and non-parallel lines indicate parameter interactions. This methodology was adapted from the literature.[2]

a

Entries	c_e (M)	V (V)	t (h)	c_l (M)	Crystallinity (%)
1	0.01	2	0.2	0.5	0.00
5	0.01	2	1	0.5	29.78
9	0.01	2	0.2	2	0.00
13	0.01	2	1	2	0.00
2	0.3	2	0.2	0.5	0.00
6	0.3	2	1	0.5	0.00
10	0.3	2	0.2	2	88.46
14	0.3	2	1	2	19.99
3	0.01	20	0.2	0.5	0.00
7	0.01	20	1	0.5	0.00
11	0.01	20	0.2	2	70.96
15	0.01	20	1	2	100.00
4	0.3	20	0.2	0.5	0.00
8	0.3	20	1	0.5	6.51
12	0.3	20	0.2	2	40.14
16	0.3	20	1	2	26.92
17	0.155	11	0.6	1.25	51.83
18	0.155	11	0.6	1.25	51.02
19	0.155	11	0.6	1.25	51.45
20	0.01	11	0.6	1.25	71.30
21	0.155	2	0.6	1.25	47.56
22	0.155	20	0.6	1.25	42.32
23	0.155	11	0.2	1.25	67.27
24	0.155	11	1	1.25	43.28
25	0.155	11	0.6	0.5	12.84
26	0.155	11	0.6	2	35.09
27	0.3	11	0.6	1.25	13.88

b

Combination	Average crystallinity (%)
Low c_e with low V	7.5
High c_e with low V	27.1
Low c_e with high V	42.7
High c_e with high V	18.4

c

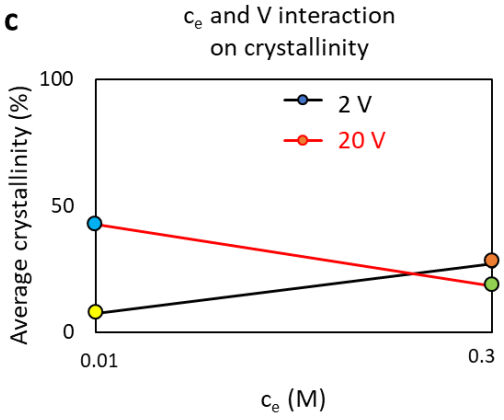

Fig. S9. Two-factor-responses interactions. (**a**) Experimental table with a classification of different electrolyte and voltage combinations, where yellow indicates the combination of the lowest electrolyte with the highest voltage, orange indicates the combination of the highest electrolyte with the lowest voltage, blue indicates the combination of the lowest electrolyte with the highest voltage, and green indicates the combination of the highest electrolyte with the highest voltage. (**b**) the average crystallinity of each combination. (**c**) the interaction plot between electrolyte and voltage to crystallinity is shown.

As shown in **Fig. S9.c**, at low voltage (2 V), increasing the electrolyte concentration increases the average crystallinity by about 20%. Meanwhile, at high voltage (20 V), increasing the electrolyte concentration decreases the average crystallinity by around 24%. **Fig. S9** shows that to get the highest

crystallinity, the combination of high voltage and low electrolyte concentration is desired (indicated by blue color).

By applying the same principle as described in **Fig. S9**, the factor interaction can be calculated for all the parameter combinations and responses. The two-factor-responses interaction plots are described in **Fig. S10-S15**, where parallel lines indicate no interaction and non-parallel lines indicate parameter interactions.

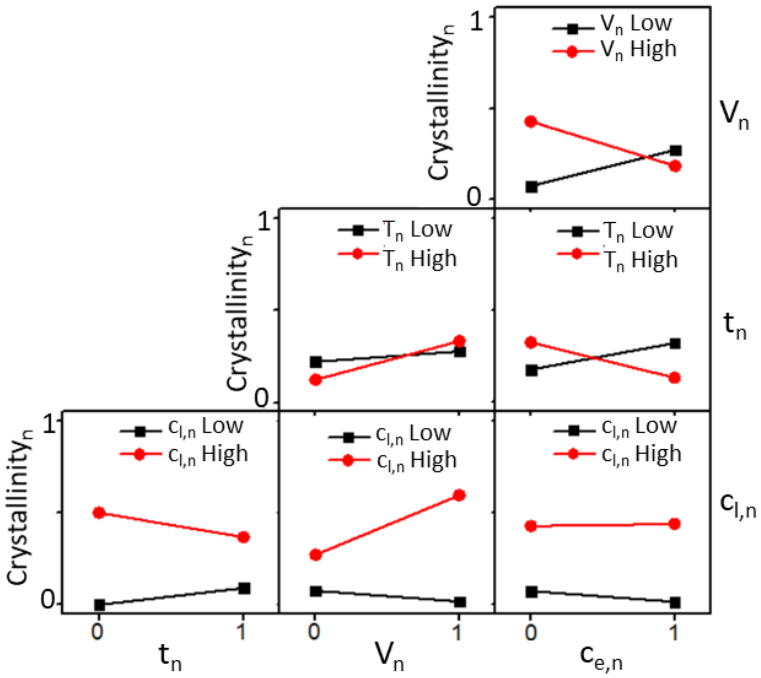

Fig. S10. Two-factor-responses interactions on crystallinity. Interaction between two factors and its effect on crystallinity.

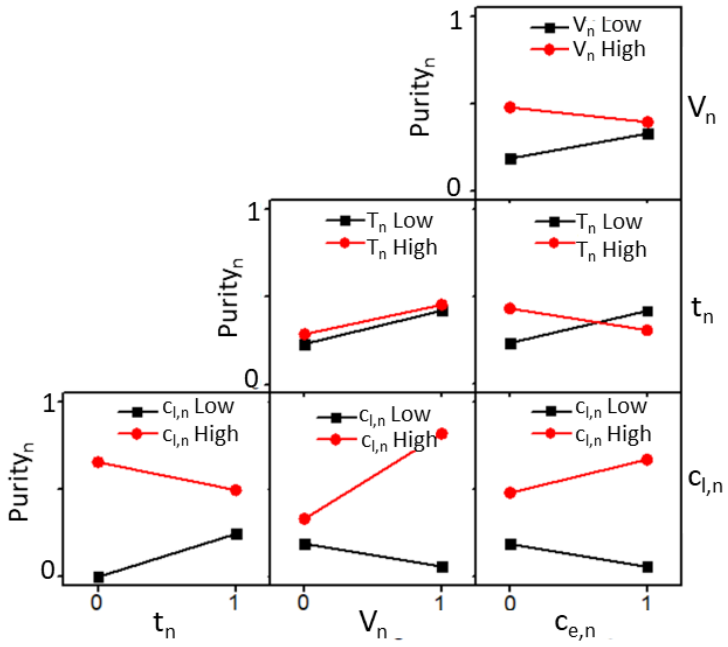

Fig. S11. Two-factor-responses interactions on purity. Interaction between two factors and its effect on purity.

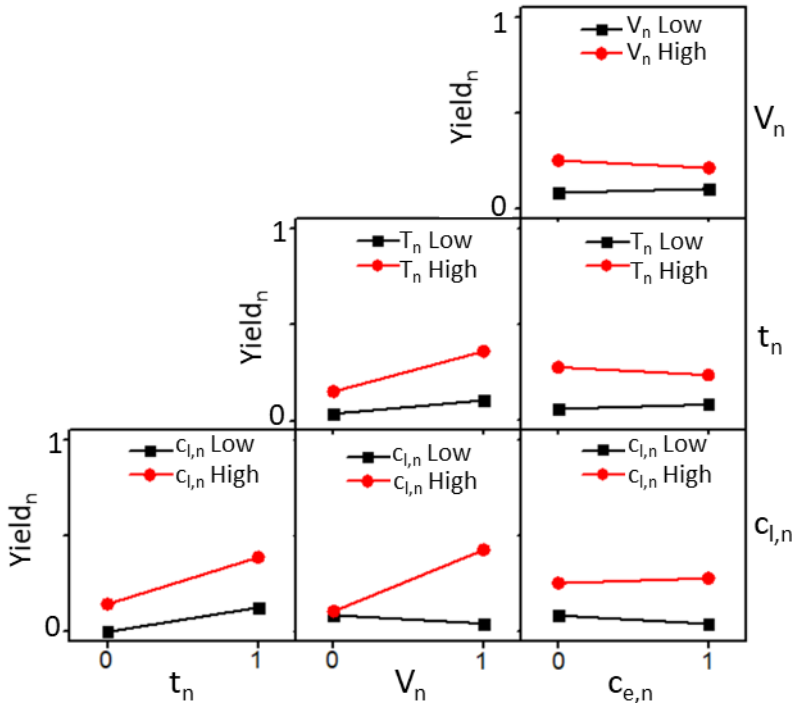

Fig. S12. Two-factor-responses interactions on yield. Interaction between two factors and its effect on yield.

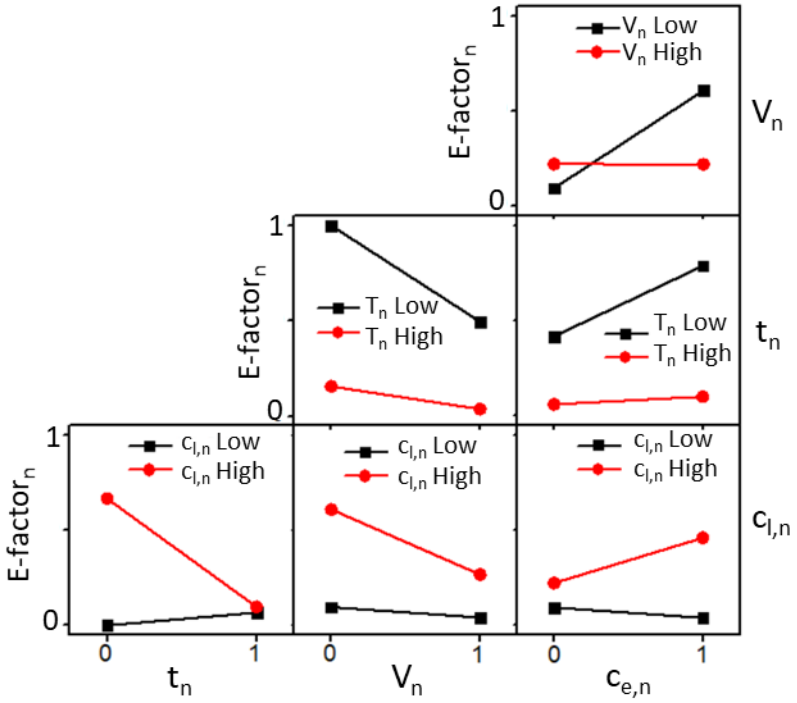

Fig. S13. Two-factor-responses interactions on E-factor. Interaction between two factors and its effect on E-factor.

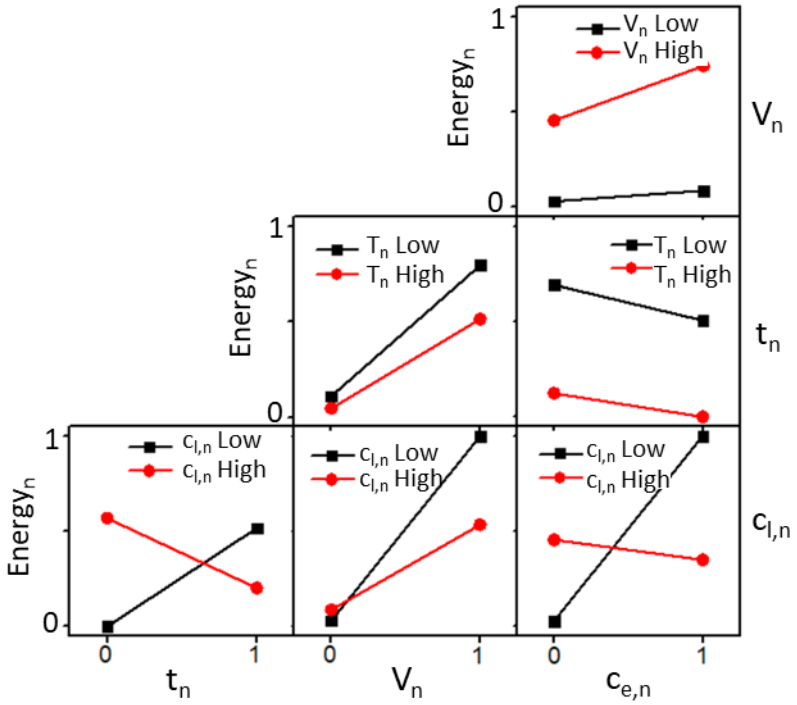

Fig. S14. Two-factor-responses interactions on energy. Interaction between two factors and its effect on energy.

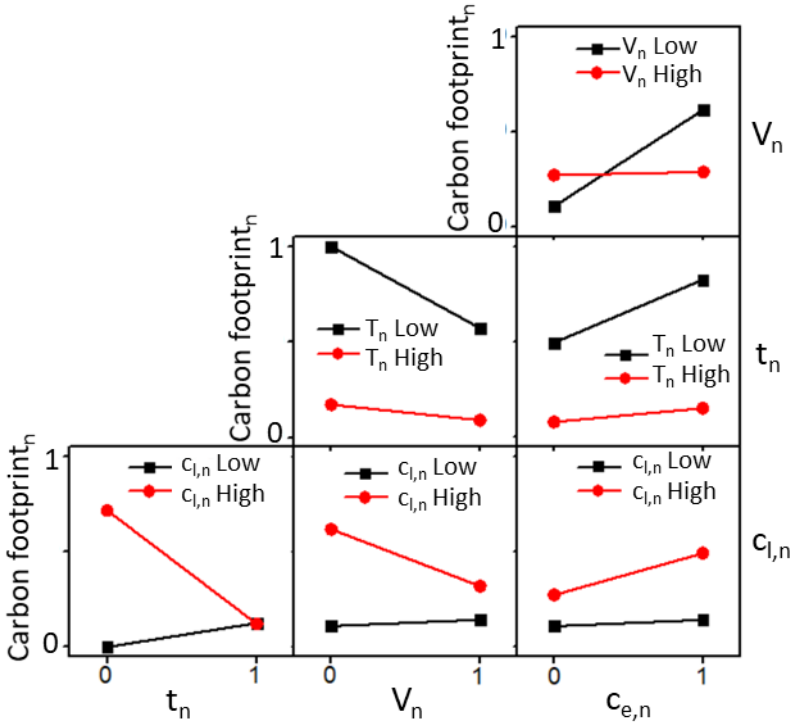

Fig. S15. Two-factor-responses interactions on carbon footprint. Interaction between two factors and its effect on carbon footprint.

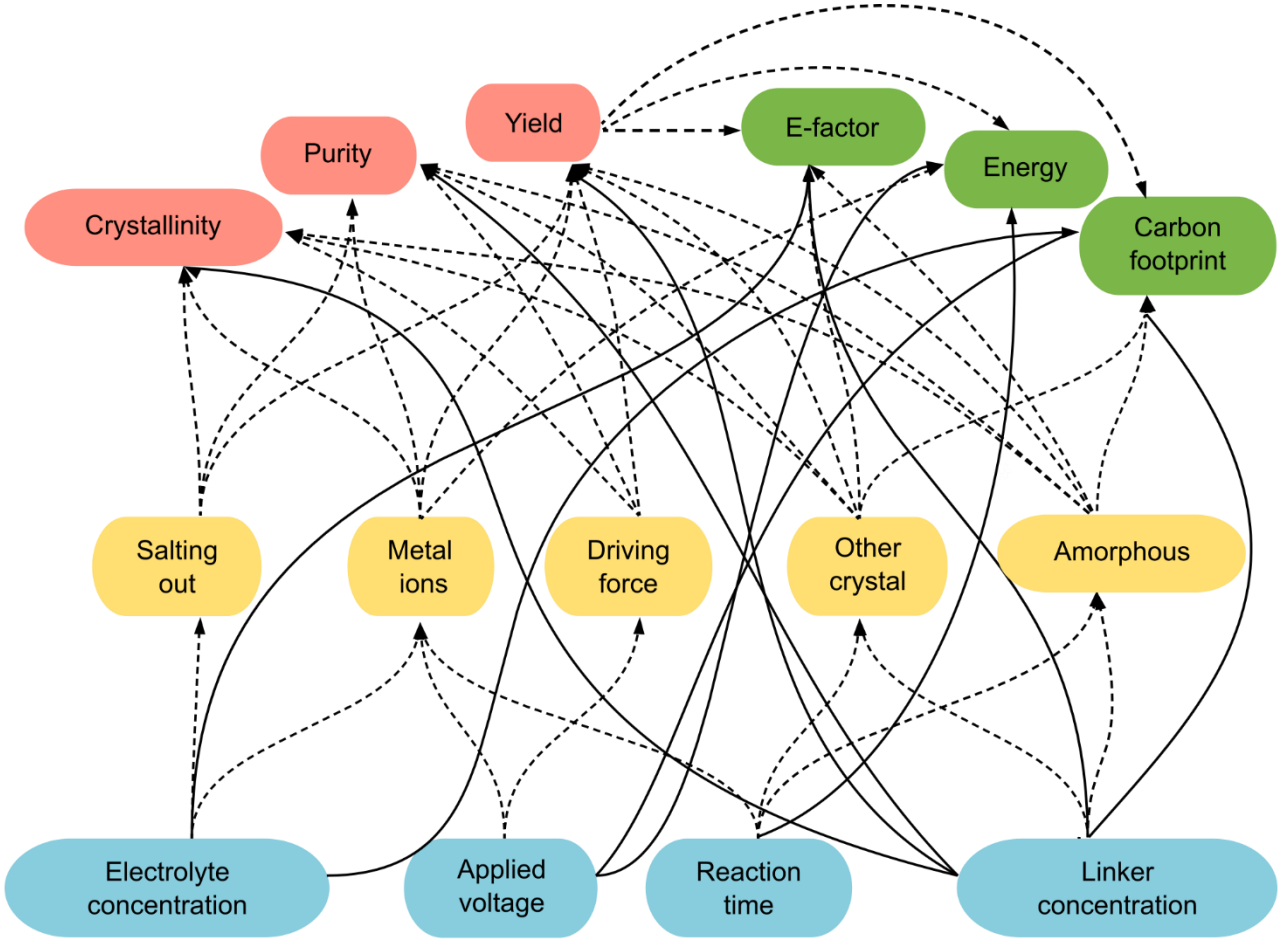

Fig. S16. Mind map of the multivariable correlations in the electrochemical synthesis of ZIF-8. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

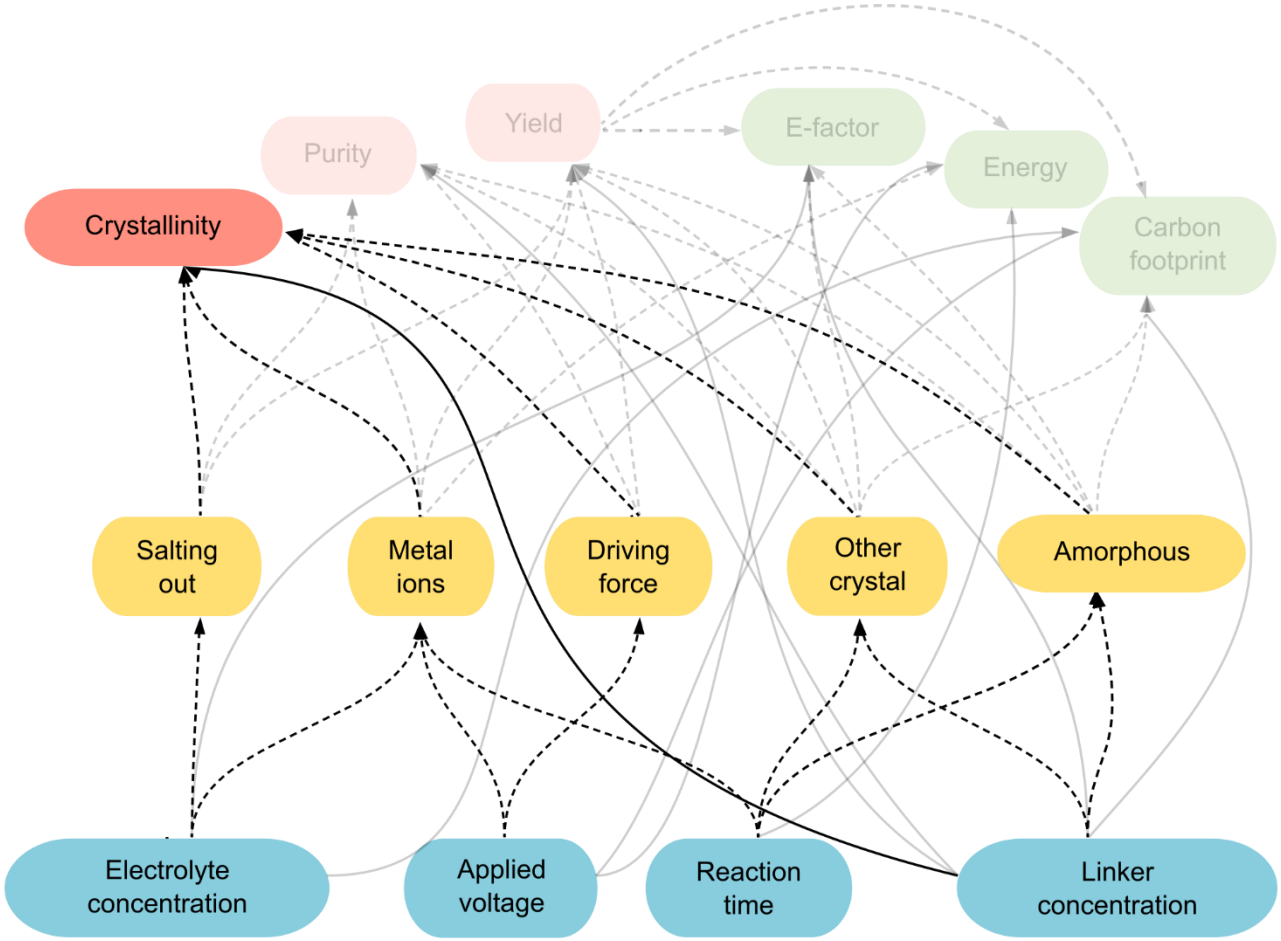

Fig. S17. Mind map of the multivariable correlations on crystallinity. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

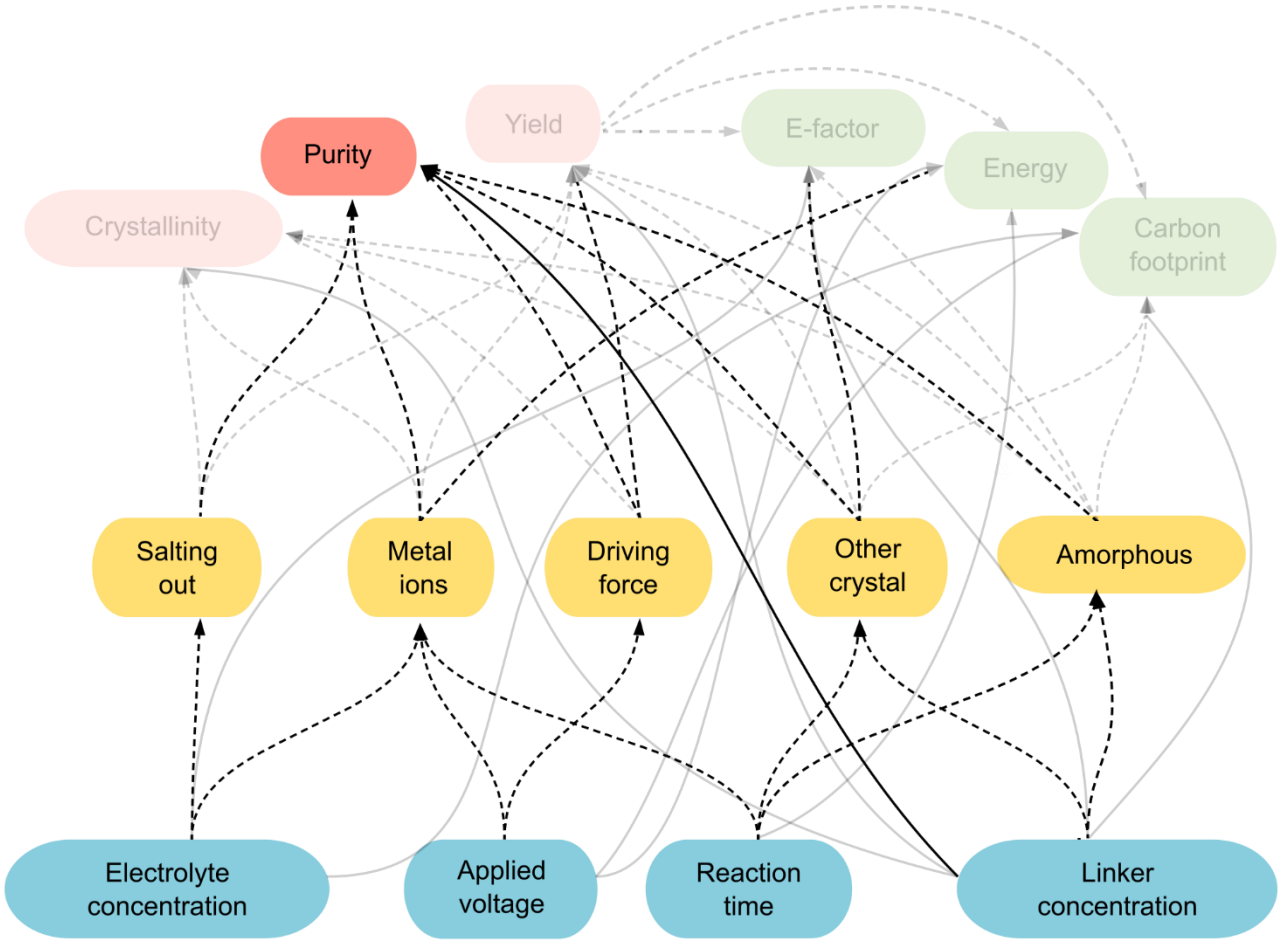

Fig. S18. Mind map of the multivariable correlations on purity. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

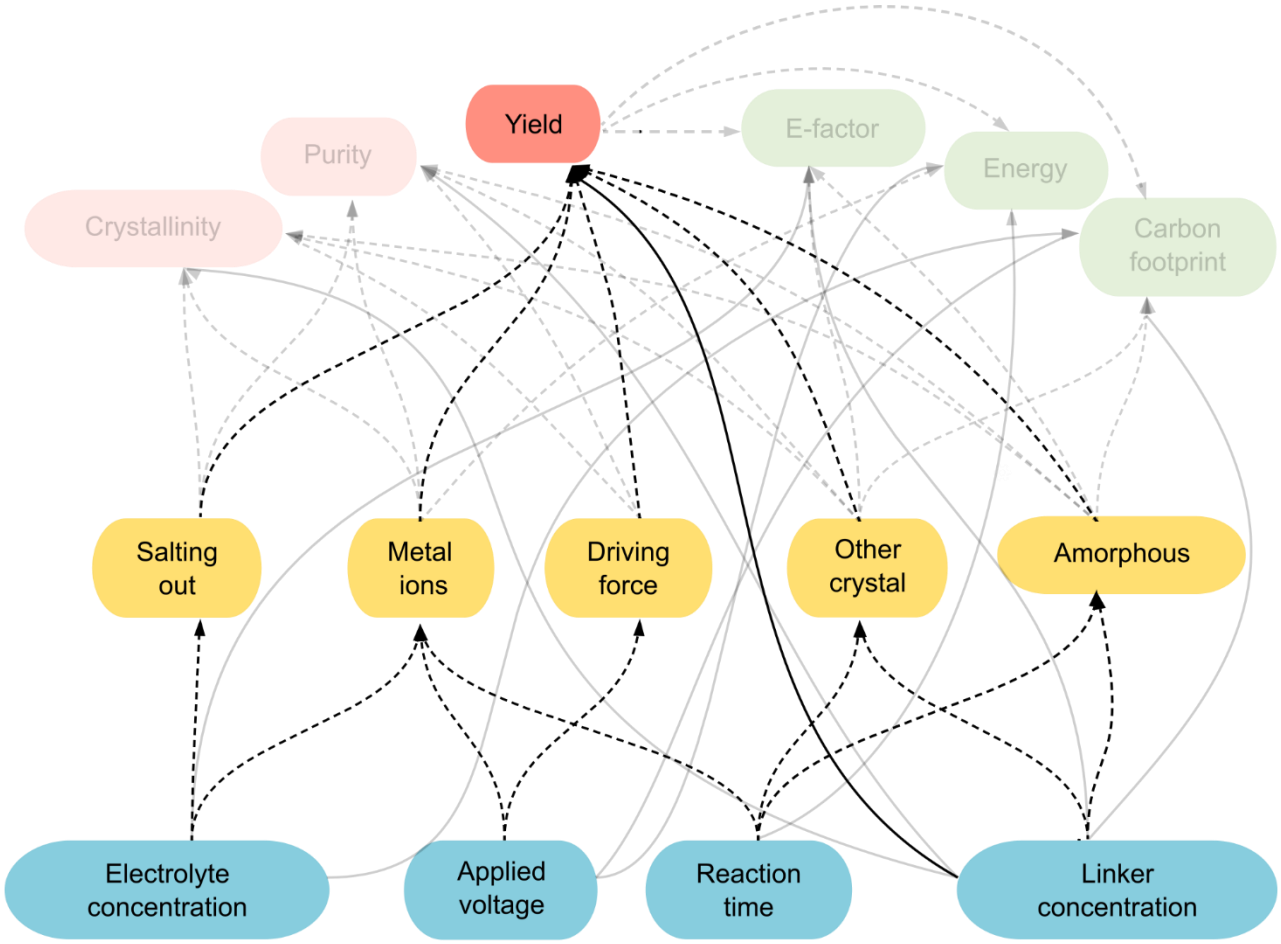

Fig. S19. Mind map of the multivariable correlations on yield. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

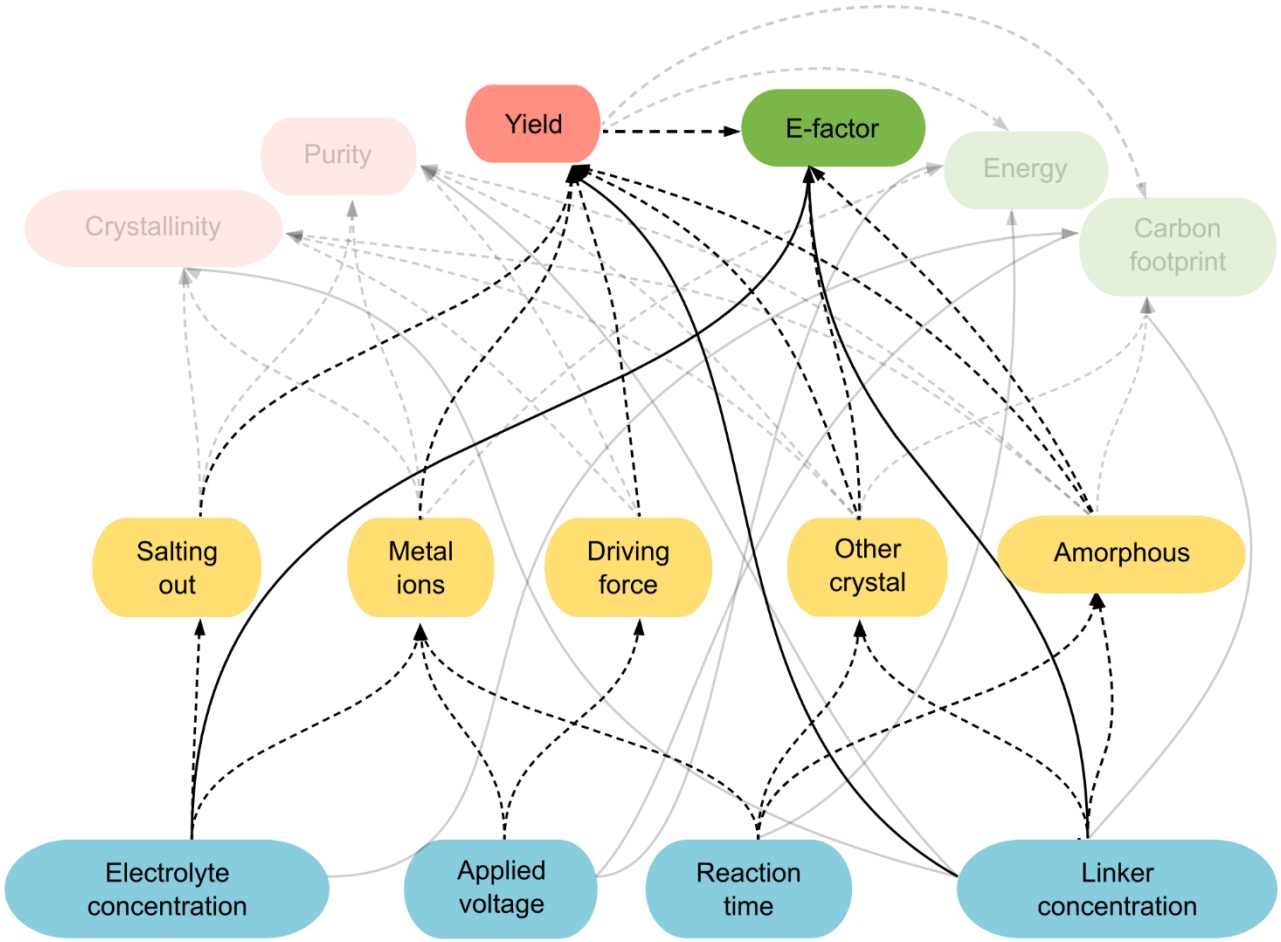

Fig. S20. Mind map of the multivariable correlations on E-factor. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

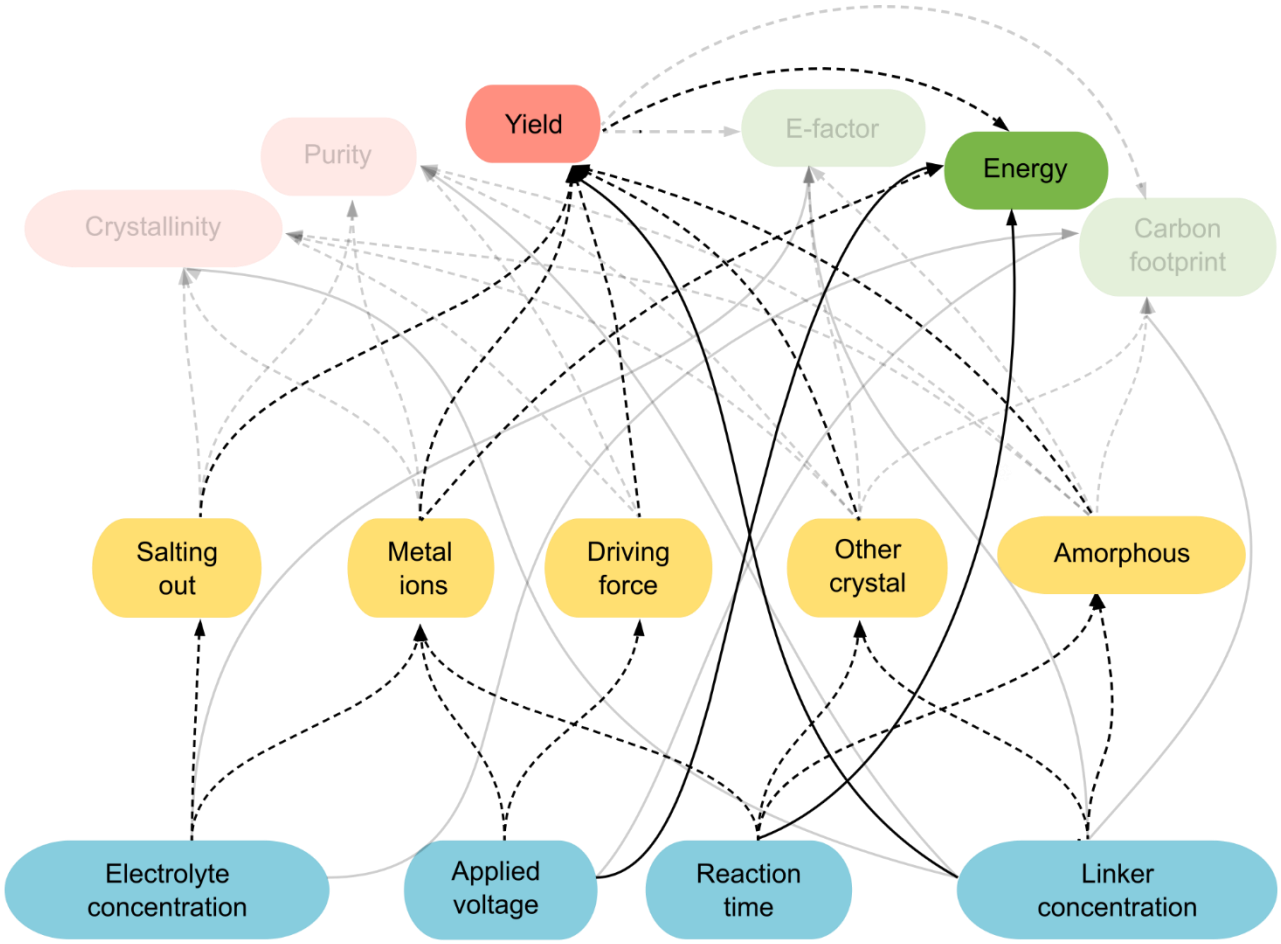

Fig. S21. Mind map of the multivariable correlations on energy. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

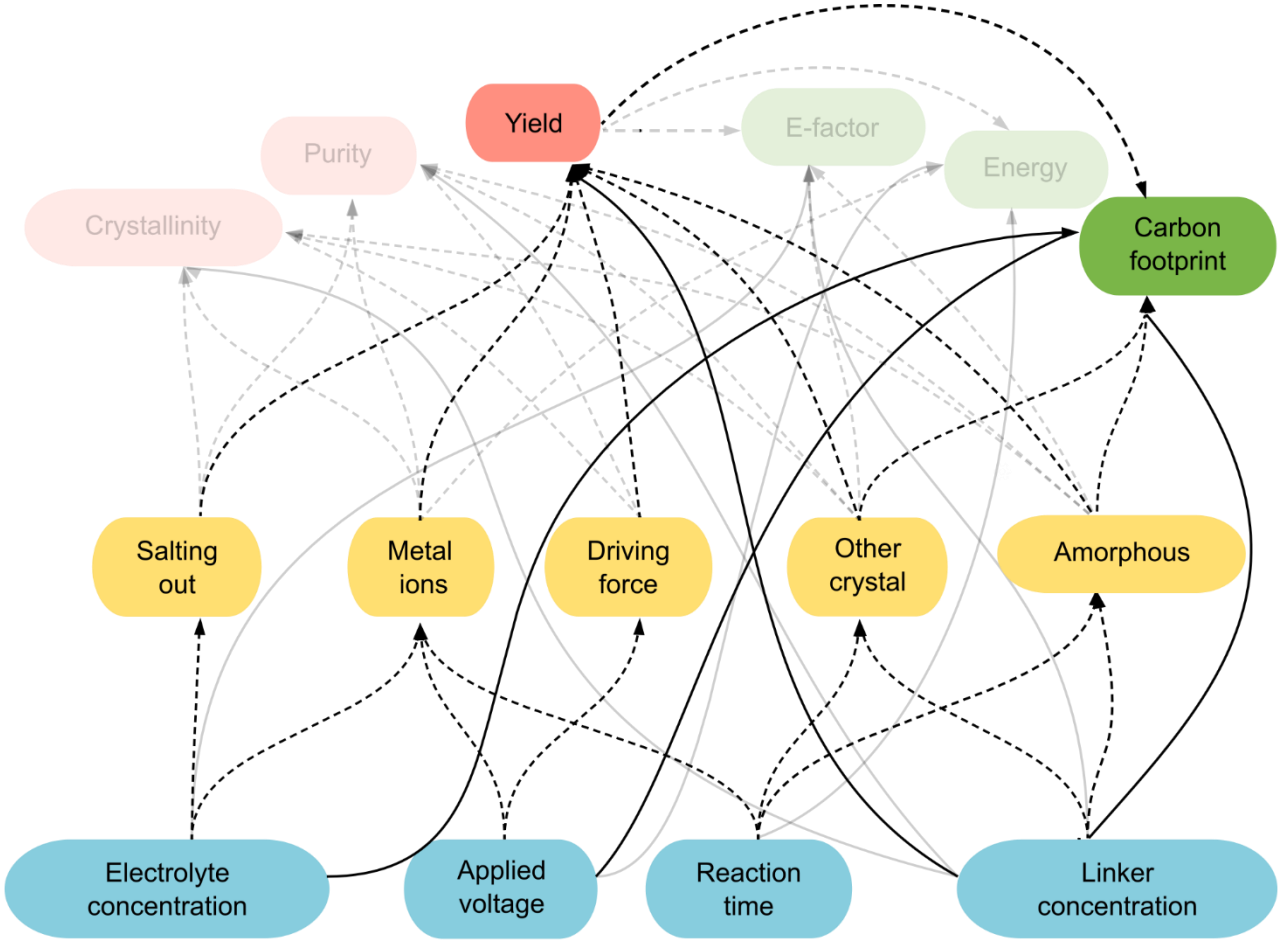

Fig. S22. Mind map of the multivariable correlations on carbon footprint. Input parameters are represented in blue, product quality in red, and process sustainability in green. The intermediate correlations are shown in yellow. The solid line indicates a direct parameter-response effect; the dashed line indicates an indirect parameter-response effect.

2.5. Determination of crystallinity

The percentage of relative crystallinity was calculated in a manner consistent with previous studies,[3–5] through a comparison of the integral-breadth of the most intense XRD peak ($2\theta \approx 7.5°$). The calculation process is illustrated in **Fig. S23**. The baseline and the integral breadth were determined by the Origin software using Pseudo-Voigt fitting by applying equation (S4). The highest integral-breadth was achieved by a sample from Entry 15, which was then used as a reference to calibrate the crystallinity of other samples using equation (S5), where β_i is the integral breadth of experiment i and β_{15} is the highest integral breadth of Entry 15.

$$\text{Integral-breadth } (\beta) = \frac{\text{peak area}}{\text{peak height}} \quad \text{(S4)}$$

$$\text{Crystallinity} = \frac{\beta_i}{\beta_{15}} \times 100\% \quad \text{(S5)}$$

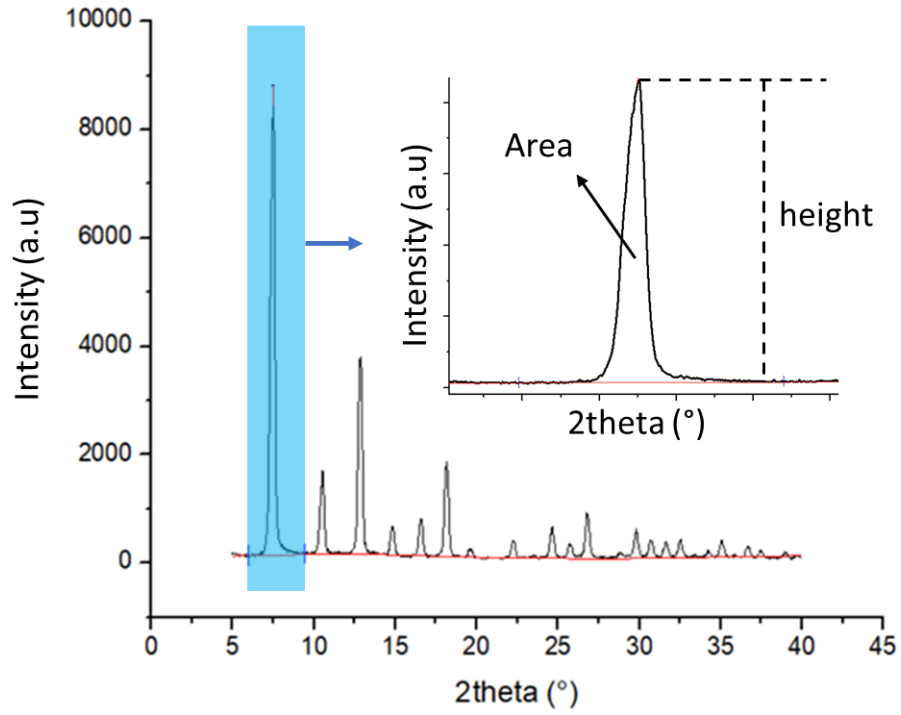

Fig. S23. Determination of relative crystallinity. An example of integral breadth determination from a sample from Entry 15, which has the highest integral breadth value.

2.6. Determination of purity

In contrast to the relative crystallinity, where the degree of crystallinity or the quality of the crystalline part among the products is compared, the purity evaluates the ratio of the desired ZIF-8 products to the whole products that may be present in the yield, including amorphous and other possible crystalline materials.

The percentage of purity was determined by XRD measurements by dividing the area under the peaks corresponding to the XRD pattern of ZIF-8 with the whole integral of the XRD pattern, following equation (S6). An example of the purity calculation from Entry 15 is given in **Fig. S24**.

$$\text{Purity} = \frac{\text{(peak area of ZIF-8)}}{\text{(total peak area in XRD)}} \times 100\% \qquad \text{(S6)}$$

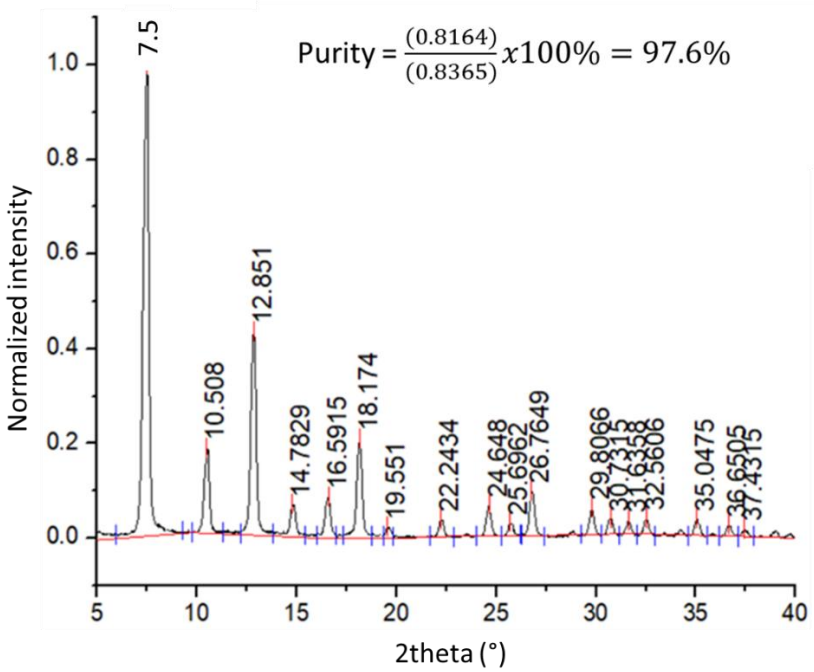

Fig. S24. Determination of purity. An example of purity determination of a sample from Entry 15.

2.7. Determination of yield

Yield is defined as the ratio between the amount of experimental and theoretical products, as shown by equation (S7).

$$\text{Yield} = \left(\frac{\text{experimental product (g)}}{\text{theoretical product (g)}}\right) \times 100\% \qquad \text{(S7)}$$

An example of yield determination based on Entry 15 in **Table S4** is given as the following. Stoichiometric of the reaction formation of ZIF-8 is presented in the reaction scheme below;

$$2C_4H_6N_2 + Zn \rightarrow C_8H_{10}N_4Zn$$
(ZIF-8)

mass of Zn consumed is 0.126 g

$$\text{mol of Zn consumed} = \frac{\text{mass of Zn (g)}}{\text{Ar of Zn (g mol}^{-1})} = \frac{0.126\text{ g}}{65.4\text{ (g mol}^{-1})} = 0.002\text{ mol}$$

as mol of zinc consumed is equivalent to mol of ZIF-8 produced, the theoretical amount of ZIF-8 produced based on zinc as the limiting reactant can be calculated by the following:

$$\begin{aligned}\text{theoretical product (ZIF-8)} &= \text{mol of ZIF-8} \times \text{Molecular mass of ZIF-8}\\ &= 0.002\text{ mol} \times 227.58\text{ g mol}^{-1}\\ &= 0.445\text{ g}\end{aligned}$$

experimental product (ZIF-8) = 0.337 g

following equation (S7), the yield from Entry 15 in **Table S4** is:

$$\text{Yield} = \left(\frac{0.337\text{ g}}{0.445\text{ g}}\right) \times 100\% = 77\%$$

2.8. Determination of E-factor

E-factor is defined as the mass ratio between the amount of waste and the product, as shown in equation (S8).

$$\text{E-factor} = \frac{\text{waste (kg)}}{\text{product (kg)}} \qquad \text{(S8)}$$

As per convention, waste is defined as the amount of all reagents minus the weight of the product, excluding water. Based on the reported synthetic details, the approximate E-factors were calculated by excluding the amount of water used as solvents and also during the washing procedures.[6,7] An example of E-factor calculation for Entry 15 is detailed by the following:

Electrolyte (KCl) = 0.01 M → 0.0112 g
Linker ($C_4H_6N_2$) = 2 M → 2.46 g
Zn electrode = 0.126 g
ZIF-8 product = 0.337 g

$$2C_4H_6N_2 + Zn + KCl \rightarrow C_8H_{10}N_4Zn + \text{waste}$$
$$(2.460\ \text{g}) + (0.126\ \text{g}) + (0.0112\ \text{g}) \rightarrow (0.337\ \text{g}) + (2.260\ \text{g})$$

Total amount of reactants = (2.460 + 0.126 + 0.0112) = 2.597 g
Amount of waste = Total amount of reactants - amount of final product
= (2.597 – 0.337) = 2.260 g

$$\text{E-factor} = \frac{\text{waste (kg)}}{\text{product (kg)}} = \frac{0.00226\ \text{(kg)}}{0.000337\ \text{(kg)}} = 6.69\ \text{kg kg}^{-1}$$

The complete datasets of E-factor calculation results are tabulated in **Table S7**.

Table S7. Experimental runs, reaction conditions, and E-factor.

Entries	c_e (M)	V (V)	t (h)	c_l (M)	Zn consumed (g)	m_l (g)	m_e (g)	Total reactant (g)	ZIF-8 product (g)	Waste (g)	E-factor (kg kg^{-1})
1	0.01	2	0.2	0.50	0.000	0.62	0.011	0.626	0.000	0.626	na.
2	0.30	2	0.2	0.50	0.017	0.62	0.335	0.967	0.000	0.967	na.
3	0.01	20	0.2	0.50	0.029	0.62	0.011	0.655	0.000	0.655	na.
4	0.30	20	0.2	0.50	0.030	0.62	0.335	0.980	0.000	0.980	na.
5	0.01	2	1.0	0.50	0.013	0.62	0.011	0.639	0.025	0.614	24.01
6	0.30	2	1.0	0.50	0.052	0.62	0.335	1.002	0.000	1.002	na.
7	0.01	20	1.0	0.50	0.146	0.62	0.011	0.772	0.000	0.772	na.
8	0.30	20	1.0	0.50	0.130	0.62	0.335	1.080	0.075	1.005	13.29
9	0.01	2	0.2	2.00	0.000	2.46	0.011	2.471	0.000	2.471	na.
10	0.30	2	0.2	2.00	0.021	2.46	0.335	2.816	0.011	2.805	251.80
11	0.01	20	0.2	2.00	0.028	2.46	0.011	2.499	0.023	2.476	105.16
12	0.30	20	0.2	2.00	0.030	2.46	0.335	2.825	0.019	2.806	145.03
13	0.01	2	1.0	2.00	0.000	2.46	0.011	2.471	0.000	2.471	na.
14	0.30	2	1.0	2.00	0.053	2.46	0.335	2.848	0.049	2.799	56.37
15	0.01	20	1.0	2.00	0.126	2.46	0.011	2.597	0.336	2.260	6.71
16	0.30	20	1.0	2.00	0.127	2.46	0.335	2.922	0.226	2.696	11.90
17	0.16	11	0.6	1.25	0.068	1.54	0.173	1.779	0.103	1.675	16.14
18	0.16	11	0.6	1.25	0.067	1.54	0.173	1.778	0.105	1.672	15.78
19	0.16	11	0.6	1.25	0.069	1.54	0.173	1.780	0.104	1.676	16.10
20	0.01	11	0.6	1.25	0.047	1.54	0.011	1.596	0.066	1.529	22.90
21	0.16	2	0.6	1.25	0.031	1.54	0.173	1.742	0.042	1.700	40.23
22	0.16	20	0.6	1.25	0.077	1.54	0.173	1.788	0.190	1.598	8.39
23	0.16	11	0.2	1.25	0.028	1.54	0.173	1.739	0.044	1.695	38.16
24	0.16	11	1.0	1.25	0.108	1.54	0.173	1.819	0.224	1.595	7.12
25	0.16	11	0.6	0.50	0.077	0.62	0.173	0.865	0.115	0.750	6.50
26	0.16	11	0.6	2.00	0.076	2.46	0.173	2.709	0.173	2.536	14.61
27	0.30	11	0.6	1.25	0.073	1.54	0.335	1.946	0.104	1.841	17.57

2.9. Determination of energy consumption

Energy consumption during electrochemical synthesis can be calculated by considering the amount of zinc consumed and the applied voltage. The methodology to calculate energy consumption is adapted from literature.[8,9]

An example of calculating energy consumption for electrochemical production ZIF-8 is determined for sample Entry 15 using the following:

1. Determine the amount of applied voltage and corresponding Zinc electrode that is dissolved.
   Voltage = 20 V
   Zinc consumed = 0.0019 mol

2. Determine the charges involved
   1 mol e^- = 96500 Coulomb

   $Zn \rightarrow Zn^{2+} + 2e^-$
   0.0019 mol × 2 mol e^- = 0.00385 mol e^-
   Total charges=mol e^-×charges (S9)
   $Total\ charges = 0.00385 \times 96500 = 371.95\ Coulomb$

3. Determine the energy
   $$\text{Energy (kWh)} = \frac{\text{Voltage (V)} \times \text{charge (C)} \times 0.278}{10^6} \quad \text{(S10)}$$
   $$\text{Energy (kWh)} = \frac{20\ \text{V} \times 371.95\ \text{Coulomb} \times 0.278}{10^6} = 0.00207\ \text{kWh}$$

4. Energy per unit product
   Total product = 0.336 g
   $$\text{Energy per unit product (kWh kg}^{-1}) = \frac{\text{Energy (kWh)}}{\text{total product (kg)}} \quad \text{(S11)}$$
   $$\text{Energy per unit product } \left(\text{kWh kg}^{-1}\right) = \frac{0.00207\ \text{kWh}}{0.000336\ \text{kg}} = 6.1\ \text{kWh kg}^{-1}$$

The complete datasets of Energy calculation results are tabulated in **Table S8**.

Table S8. Experimental runs, reaction conditions, and energy consumption.

Entries	c_e (M)	V (V)	t (h)	c_l (M)	Zn consumed (g)	ZIF-8 product (g)	Zn (mmol)	e^- (mmol)	Charge (Coulomb)	Energy (Wh)	Energy consumption (kWh kg^{-1})
1	0.01	2	0.2	0.50	0.000	0.000	0.000	0.000	0.00	0.000	na.
2	0.30	2	0.2	0.50	0.017	0.000	0.260	0.520	50.18	0.027	na.
3	0.01	20	0.2	0.50	0.029	0.000	0.444	0.887	85.61	0.476	na.
4	0.30	20	0.2	0.50	0.030	0.000	0.459	0.918	88.56	0.492	na.
5	0.01	2	1.0	0.50	0.013	0.025	0.199	0.398	38.38	0.021	0.84
6	0.30	2	1.0	0.50	0.052	0.000	0.795	1.591	153.50	0.085	na.
7	0.01	20	1.0	0.50	0.146	0.000	2.233	4.466	430.99	2.396	na.
8	0.30	20	1.0	0.50	0.130	0.075	1.988	3.977	383.76	2.133	28.23
9	0.01	2	0.2	2.00	0.000	0.000	0.000	0.000	0.00	0.000	na.
10	0.30	2	0.2	2.00	0.021	0.011	0.321	0.642	61.99	0.034	3.09
11	0.01	20	0.2	2.00	0.028	0.023	0.428	0.857	82.66	0.459	19.52
12	0.30	20	0.2	2.00	0.030	0.019	0.459	0.918	88.56	0.492	25.45
13	0.01	2	1.0	2.00	0.000	0.000	0.000	0.000	0.00	0.000	na.
14	0.30	2	1.0	2.00	0.053	0.049	0.811	1.621	156.45	0.087	1.75
15	0.01	20	1.0	2.00	0.126	0.336	1.927	3.854	371.95	2.068	6.14
16	0.30	20	1.0	2.00	0.127	0.226	1.942	3.885	374.90	2.084	9.2
17	0.16	11	0.6	1.25	0.068	0.103	1.040	2.080	200.73	0.613	5.91
18	0.16	11	0.6	1.25	0.067	0.105	1.025	2.050	197.78	0.604	5.71
19	0.16	11	0.6	1.25	0.069	0.104	1.055	2.111	203.69	0.622	5.98
20	0.01	11	0.6	1.25	0.047	0.066	0.719	1.438	138.74	0.424	6.35
21	0.16	2	0.6	1.25	0.031	0.042	0.474	0.948	91.51	0.050	1.2
22	0.16	20	0.6	1.25	0.077	0.190	1.178	2.355	227.30	1.263	6.64
23	0.16	11	0.2	1.25	0.028	0.044	0.428	0.857	82.66	0.252	5.69
24	0.16	11	1.0	1.25	0.108	0.224	1.652	3.304	318.81	0.974	4.35
25	0.16	11	0.6	0.50	0.077	0.115	1.178	2.355	227.30	0.695	6.02
26	0.16	11	0.6	2.00	0.076	0.173	1.162	2.325	224.35	0.686	3.95
27	0.30	11	0.6	1.25	0.073	0.104	1.117	2.233	215.49	0.659	6.29

2.10. Determination of Carbon footprint

For the calculation of the carbon footprint, the energy used for the equipment and the waste generated were converted into equivalent CO_2. For the energy source, the CO_2 coefficient of 0.79 kg CO_2 kWh^{-1} was chosen for electrical energy provided by the Saudi Arabian national grid.[15] The calculation to estimate the equivalent CO_2 for the incineration of the chemical waste took into account the carbon content of the linker 2-methylimidazole ($C_4H_6N_2$) and KCl incineration, following the methodology as reported in the literature,[10] and database from CCalC2.[16] An example of the calculation for sample Entry 15 is given below.

 # CO_2 equivalent from linker ($C_4H_6N_2$) incineration:

$C_4H_6N_2 + 7.5O_2 \rightarrow 4CO_2 + 2NO_2 + 3H_2O$

CO_2 coefficient factor $= \frac{4 \text{ mol } CO_2}{1 \text{ mol } C_4H_6N_2} \times \left(\frac{44 \text{ g mol}^{-1}}{82 \text{ g mol}^{-1}}\right) = 2.14 \text{ g g}^{-1} = 2.14 \text{ kg kg}^{-1}$

Total linker waste = 2.2 g

CO_2 equivalent from linker inceneration = $0.0022 \times 2.14 = 0.0048$ kg kg^{-1}

Total ZIF-8 product = 0.337 g

CO_2 equivalent from linker inceneration per product = $\frac{0.0048}{0.000337} = 14.29$ kg kg^{-1}

 # CO_2 equivalent from electrolyte (KCl) incineration:

KCl waste = 0.0112 g

From CCalC2 software, incineration of 0.0112 g KCl is equivalent to 0.0166 kg kg^{-1}

#CO_2 equivalent from electricity grid = 0.79 Kg CO_2/kWh

Energy consumption per kg ZIF-8 = 6.14 kWh kg^{-1}

CO_2 equivalent from ZIF-8 production = $6.14 \times 0.79 = 4.85$ kg kg^{-1}

TOTAL carbon footprint = CO_2 from waste + CO_2 from electricity

= 14.29 + 0.0166 + 4.85

= 19.15 kg kg^{-1}

The complete datasets of Energy calculation results are tabulated in **Table S9**.

Table S9. Experimental runs, reaction conditions, and carbon footprint.

Entries	c_e (M)	V (V)	t (h)	c_l (M)	Zn consumed (g)	Linker (g)	Electrolyte (g)	Total reactant (g)	ZIF-8 product (g)	Linker waste (g)	Electrolyte waste (g)	*CO_2 from linker (kg kg^{-1})	*CO_2 from electrolyte (kg kg^{-1})	Energy consumption (kWh kg^{-1})	*CO_2 from energy (kg kg^{-1})	Total *CO_2 from ZIF-8 synthesis (kg kg^{-1})
1	0.010	2	0.2	0.50	0.000	0.615	0.011	0.626	0.000	0.615	0.011	na.	na.	na.	na.	na.
2	0.300	2	0.2	0.50	0.017	0.615	0.335	0.967	0.000	0.632	0.335	na.	na.	na.	na.	na.
3	0.010	20	0.2	0.50	0.029	0.615	0.011	0.655	0.000	0.644	0.011	na.	na.	na.	na.	na.
4	0.300	20	0.2	0.50	0.030	0.615	0.335	0.980	0.000	0.645	0.335	na.	na.	na.	na.	na.
5	0.010	2	1.0	0.50	0.013	0.615	0.011	0.639	0.026	0.602	0.011	50.45	0.22	0.84	0.66	51.33
6	0.300	2	1.0	0.50	0.052	0.615	0.335	1.002	0.000	0.667	0.335	na.	na.	na.	na.	na.
7	0.010	20	1.0	0.50	0.146	0.615	0.011	0.772	0.000	0.761	0.011	na.	na.	na.	na.	na.
8	0.300	20	1.0	0.50	0.130	0.615	0.335	1.080	0.076	0.669	0.335	18.95	2.21	28.23	22.30	43.46
9	0.010	2	0.2	2.00	0.000	2.460	0.011	2.471	0.000	2.460	0.011	na.	na.	na.	na.	na.
10	0.300	2	0.2	2.00	0.021	2.460	0.335	2.816	0.011	2.470	0.335	474.46	15.02	3.09	2.44	491.92
11	0.010	20	0.2	2.00	0.028	2.460	0.011	2.499	0.024	2.464	0.011	224.01	0.24	19.52	15.42	239.67
12	0.300	20	0.2	2.00	0.030	2.460	0.335	2.825	0.019	2.471	0.335	273.28	8.65	25.45	20.11	302.03
13	0.010	2	1.0	2.00	0.000	2.460	0.011	2.471	0.000	2.460	0.011	na.	na.	na.	na.	na.
14	0.300	2	1.0	2.00	0.053	2.460	0.335	2.848	0.050	2.463	0.335	106.18	3.37	1.75	1.38	110.93
15	0.010	20	1.0	2.00	0.126	2.460	0.011	2.597	0.337	2.249	0.011	14.30	0.02	6.14	4.85	19.16
16	0.300	20	1.0	2.00	0.127	2.460	0.335	2.922	0.226	2.361	0.335	22.31	0.74	9.20	7.27	30.32
17	0.155	11	0.6	1.25	0.068	1.538	0.173	1.779	0.104	1.502	0.173	30.96	0.83	5.91	4.67	36.47
18	0.155	11	0.6	1.25	0.067	1.538	0.173	1.778	0.106	1.499	0.173	30.28	0.82	5.71	4.51	35.60
19	0.155	11	0.6	1.25	0.069	1.538	0.173	1.780	0.104	1.503	0.173	30.89	0.83	5.98	4.73	36.45
20	0.010	11	0.6	1.25	0.047	1.538	0.011	1.596	0.067	1.518	0.011	48.65	0.08	6.35	5.02	53.76
21	0.155	2	0.6	1.25	0.031	1.538	0.173	1.742	0.042	1.527	0.173	77.33	2.05	1.20	0.95	80.33
22	0.155	20	0.6	1.25	0.077	1.538	0.173	1.788	0.190	1.425	0.173	16.01	0.45	6.64	5.24	21.71
23	0.155	11	0.2	1.25	0.028	1.538	0.173	1.739	0.044	1.522	0.173	73.31	1.95	5.69	4.50	79.75
24	0.155	11	1.0	1.25	0.108	1.538	0.173	1.819	0.224	1.422	0.173	13.59	0.39	4.35	3.44	17.41
25	0.155	11	0.6	0.50	0.077	0.615	0.173	0.865	0.115	0.577	0.173	10.69	0.75	6.02	4.76	16.20
26	0.155	11	0.6	2.00	0.076	2.460	0.173	2.709	0.174	2.362	0.173	29.13	0.50	3.95	3.12	32.75
27	0.300	11	0.6	1.25	0.073	1.538	0.335	1.946	0.105	1.506	0.335	30.75	1.60	6.29	4.97	37.32

*equivalent CO_2 emission

2.11. Machine learning algorithm and data generation

Support Vector Machine (SVM) was chosen to fit the data in this study due to several merits. First, it is robust to outliers as the decision boundary is only determined by the support vectors, which are a subset of the training data samples close to the boundary. Second, SVM uses kernels to model non-linear hyperplane as a decision boundary as it is analogous to mapping the input data into a higher-dimensional space for an easier separation. Last, SVM suffers less from the overfitting issue when the training data set is small, compared to other regression models.

Random Forest (RF) is a widely used machine learning method, which construct multiple trees to do classification or regression. However, when we use RF for regression, the amount of data will have a large impact on the performance of RF models. In our case, we only have a small number of training records, the RF model will be easily overfitted and the prediction value will be extremely discrete, finally leading to poor performance.

All codes used in this paper are implemented by Python. Pandas5 package is used to import all the original data and export all the generated data. The support vector machine (SVM) model is implemented and trained by Scikit-learn6 package. All visualization work is accomplished by using Matplotlib7 package. Radial basis function kernel was used in SVM model due to its fitting flexibility for high-dimension data. The parameter of SVM model such as 'gamma' is crucial for its performance. A too small value of gamma leads to low accuracy, while a high value of gamma could cause overfitting problem. To choose the best parameters for SVM model such as 'gamma' and 'C', cross validation method was applied. After cross validation, gamma is set to 1, and C is set to 10. Min-max normalization method was used to scale all the input and output variables to a certain range (0~1).

Before data generation process, all the input is scaled from 0 to 1 as mentioned. The step value of 0.04 was used to generate a 4-dimension grid (for 4 input variables in this experiment). For each dimension there are (1 / 0.04) + 1= 26 steps, so the total number of generated points are 26×26×26×26 = 456,976. The machine learning model was also validated for its reliability. The validity test was also compared to other machine learning algorithm, e.g., Random Forest (RF), and the results are given in **Fig. S25** and **Fig. S26**.

2.12. Machine learning model validity test

The validity of the machine learning model was tested by comparing the prediction with the experimental results from two machine learning algorithms: Support Vector Machine (SVM) and Random Forest (RF). The better model is indicated by a higher regression coefficient (R^2) value and a lower mean square error (MSE) value. The model comparison on the product quality responses (crystallinity, purity, and yield) and process sustainability responses (E-factor, energy, and carbon footprint) is presented in **Fig. S25** and **Fig. S26**, respectively. The results show that the fittings using the SVM model are better than those of the RF model.

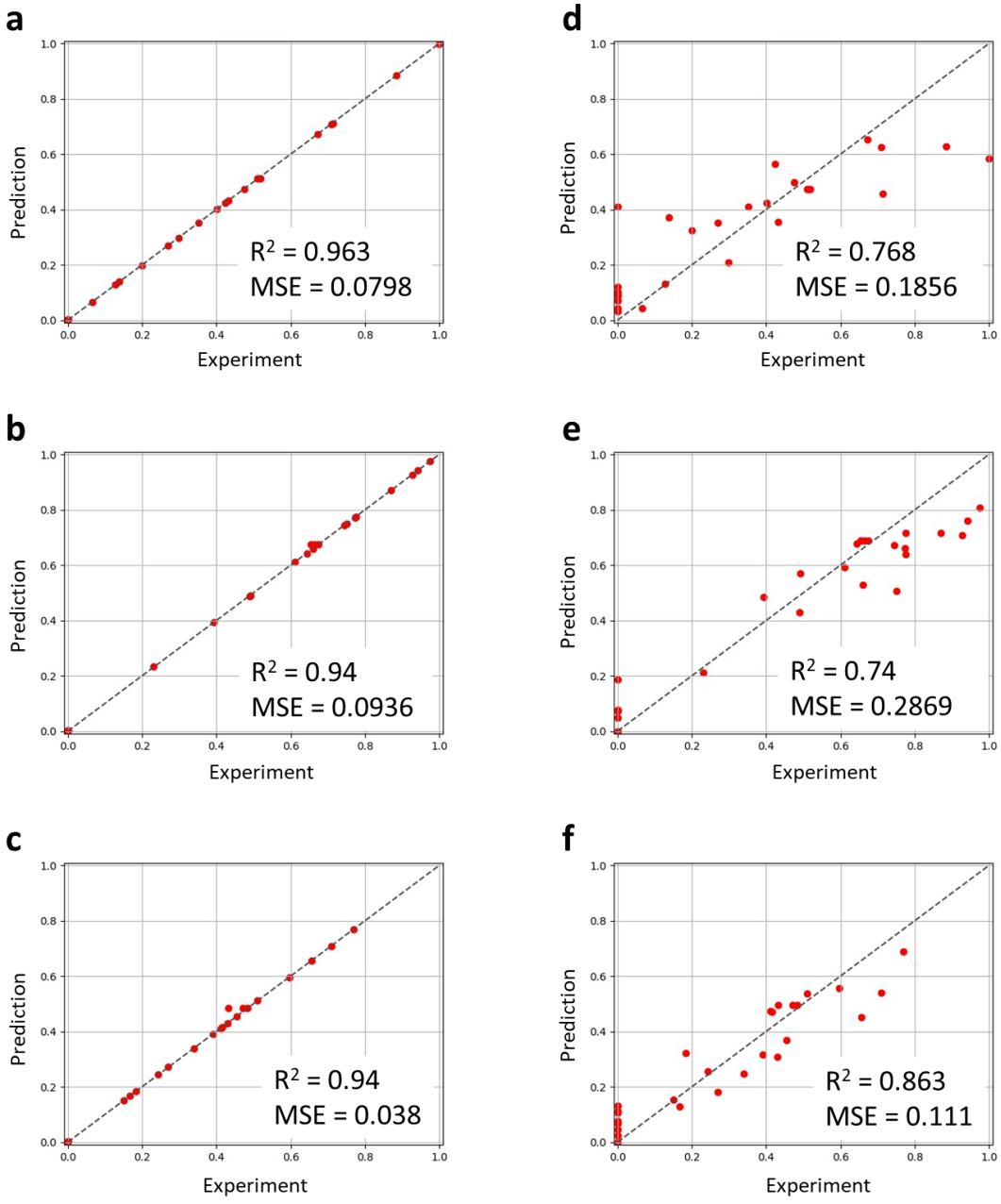

Fig. S25. Validity evaluation of the training dataset for product quality. (**a-c**) SVM and (**d-f**) RF models for crystallinity, purity, and yield.

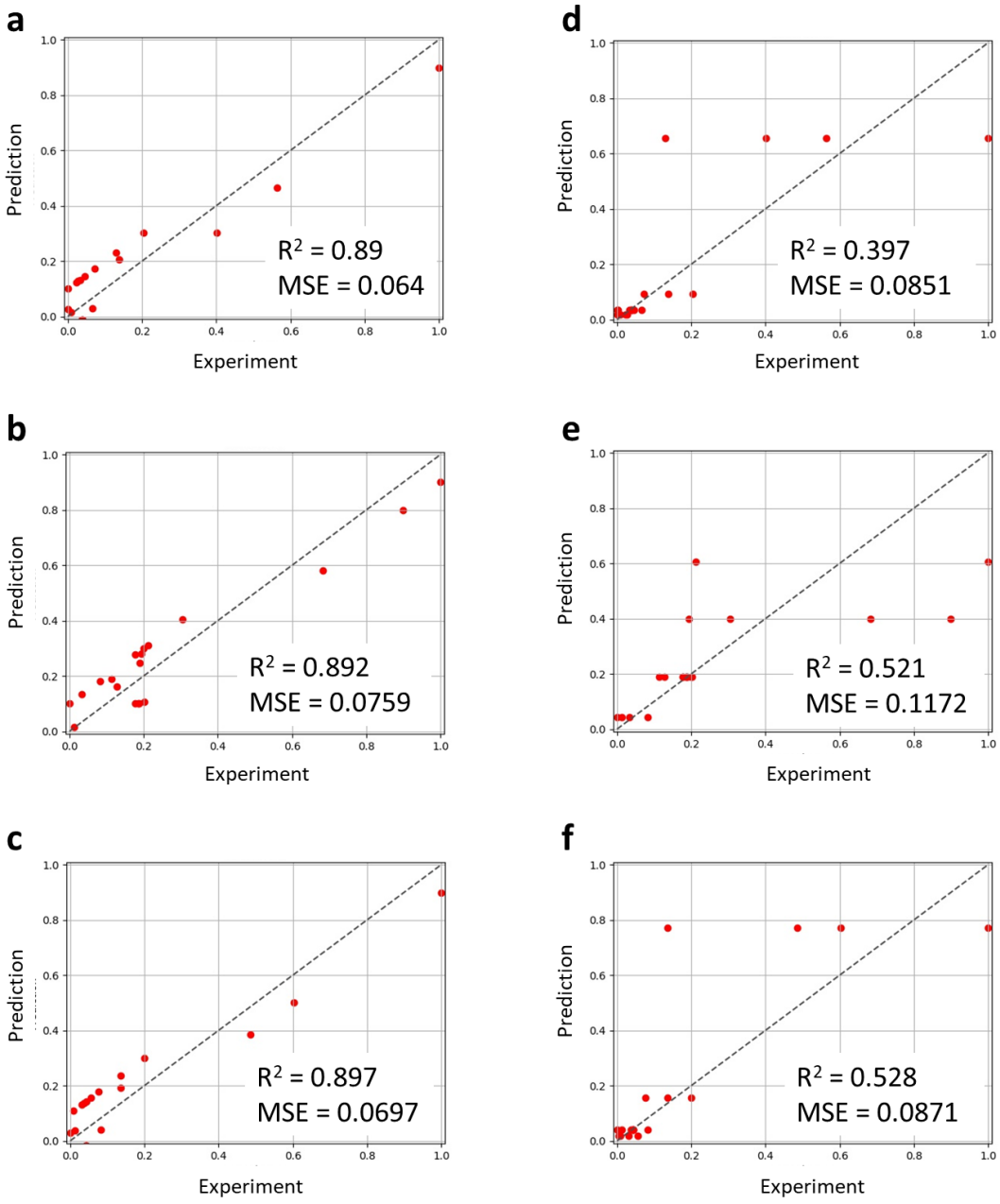

Fig. S26. Validity evaluation on the training dataset of process sustainability quality. (**a-c**) SVM and (**d-f**) RF models for E-factor, energy, and carbon footprint.

2.13. Sampling points

The sampling distributions in the Central-Composite Design that applies the face-centered model of DoE are displayed in **Fig. S27a**. The CCD model comprises the factorial parts, which are located at the corners of the design space, the axial parts that are located in the center of the design space, and the center point. The total number of experiments depends on the number of variables (k), which is calculated by 2^k+2k+n_0, where 2^k and 2k represent the factorial and axial runs, respectively, and n_0 is the number of repeated experiments at the center point.[11]

As there are four parameters to be optimized in this work (electrolyte concentration, applied voltage, reaction time, and linker concentration), and each parameter has three levels, the conventional one-factor at-a-time approach thus would have resulted in $3^4 = 81$ experiments. Nevertheless, by applying the CCF model of DoE, the total number of experiments can be reduced to 27 experiments, including 16 factorial points, six axial points, and five center points with three repetitions, to ensure reproducibility.

Fig. S27b illustrates the evolution of the sampling points generated by grid-search methods. The number of samples was expanded from the initial 27 datasets into much larger datasets. A more detailed illustration of the sampling points in the 4D spaces for both DoE and machine learning are visualized in Supplementary Video 1 and Video 2.

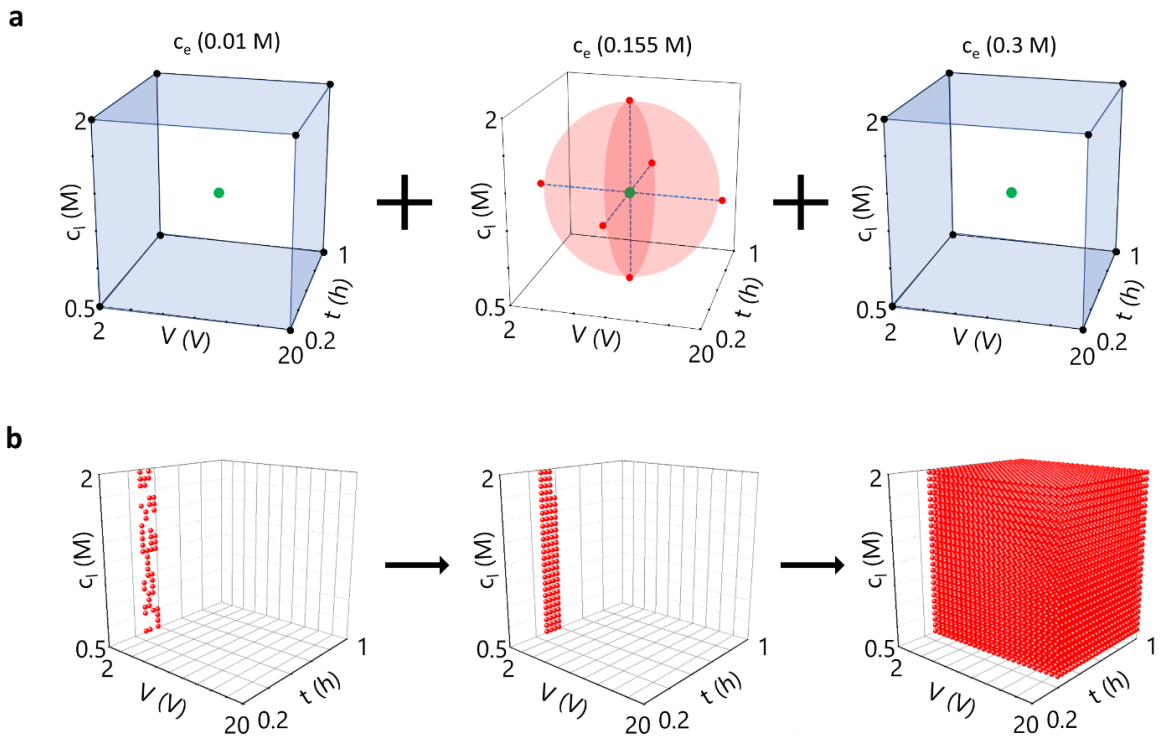

Fig. S27. Distribution of sampling points. (**a**) sampling points generated from the CCF model of DoE, (**b**) the extended sampling points distribution generated from the grid search method of machine learning.

2.14. Code for machine learning fitting and optimization

The code for the machine learning fitting and all the details of implementation were written in Python. The full code is available from the corresponding author upon reasonable request.

A. AI-module 1: SVM + Grid search + Desirability function

```
import numpy as np
import matplotlib.pyplot as plt
from sklearn.svm import SVR
from matplotlib import cm
import pandas as pd
import matplotlib
import time
import geatpy as ea

data = pd.read_excel('CCF design Rifan - 20200303.xlsx')
data = data.iloc[:,[2,3,4,5,6,7,8,9]].dropna()

data_train = data.iloc[1:,:]

data_train = data_train.iloc[:,[0,1,2,3,4]]

ori_features = data_train.iloc[:,:-1].values

minmin = np.min(ori_features,axis=0)
maxmax = np.max(ori_features,axis=0)
features = (ori_features -minmin)/(maxmax-minmin)
labels_4 = data_train.iloc[:,-1].values / 100
labels_5 = data.iloc[1:,5].values / 100
labels_6 = data.iloc[1:,6].values / 100

clf_4 = SVR(gamma=1,C=10,epsilon=0.001)
clf_5 = SVR(gamma=1,C=10,epsilon=0.001)
clf_6 = SVR(gamma=1,C=10,epsilon=0.001)
```

```
clf_4 = clf_4.fit(features, labels_4)
clf_5 = clf_5.fit(features, labels_5)
clf_6 = clf_6.fit(features, labels_6)

data = pd.read_excel('sustainability.xlsx')
data = data.iloc[:,[1,2,3,4,5,6,7]].dropna()
data = data.iloc[1:,:]
data_1 = data.iloc[:,[0,1,2,3,4]]
data_2 = data.iloc[:,[0,1,2,3,5]]
data_3 = data.iloc[:,[0,1,2,3,6]]
ori_features = data_1.iloc[:,:-1].values
minmin = np.min(ori_features,axis=0)
maxmax = np.max(ori_features,axis=0)
features = (ori_features -minmin)/(maxmax-minmin)

labels = data_1.iloc[:,-1].values
label_min = np.min(labels)
label_max = np.max(labels)
labels_1 = (labels - label_min) / (label_max - label_min)

labels = data_2.iloc[:,-1].values
label_min = np.min(labels)
label_max = np.max(labels)
labels_2 = (labels - label_min) / (label_max - label_min)

labels = data_3.iloc[:,-1].values
label_min = np.min(labels)
label_max = np.max(labels)
labels_3 = (labels - label_min) / (label_max - label_min)

clf_1 = SVR(gamma=1,C=1)
clf_2 = SVR(gamma=1,C=1)
clf_3 = SVR(gamma=1,C=1)

clf_1 = clf_1.fit(features, labels_1)
clf_2 = clf_2.fit(features, labels_2)
clf_3 = clf_3.fit(features, labels_3)
```

```
def aim_max(a,b,c,d):
    feature = np.concatenate([a,b,c,d],-1)
    return clf_4.predict(feature) + clf_5.predict(feature) + clf_6.predict(feature)

def aim_min(a,b,c,d):
    feature = np.concatenate([a,b,c,d],-1)
    return clf_1.predict(feature) + clf_2.predict(feature) + clf_3.predict(feature)

class MyProblem(ea.Problem):
    def __init__(self):
        name = 'MyProblem'
        M = 10
        maxormins = [-1] * M
        Dim = 4
        varTypes = [0] * Dim
        lb = [0] * Dim
        ub = [1] * Dim
        lbin = [1] * Dim
        ubin = [1] * Dim
        ea.Problem.__init__(self, name, M, maxormins, Dim, varTypes, lb, ub, lbin, ubin)
    def aimFunc(self, pop):
        x1 = pop.Phen[:, [0]]
        x2 = pop.Phen[:, [1]]
        x3 = pop.Phen[:, [2]]
        x4 = pop.Phen[:, [3]]

        pop.ObjV = np.zeros((pop.Phen.shape[0], self.M))
        pop.ObjV[:,[0]] = np.clip(clf_4.predict(np.concatenate([x1,x2,x3,x4],-1)).reshape(-1,1),0,1)
        pop.ObjV[:,[1]] = np.clip(clf_5.predict(np.concatenate([x1,x2,x3,x4],-1)).reshape(-1,1),0,1)
        pop.ObjV[:,[2]] = np.clip(clf_6.predict(np.concatenate([x1,x2,x3,x4],-1)).reshape(-1,1),0,1)
        pop.ObjV[:,[3]] = -np.clip(clf_1.predict(np.concatenate([x1,x2,x3,x4],-1)).reshape(-1,1),0,1)
        pop.ObjV[:,[4]] = -np.clip(clf_2.predict(np.concatenate([x1,x2,x3,x4],-1)).reshape(-1,1),0,1)
        pop.ObjV[:,[5]] = -np.clip(clf_3.predict(np.concatenate([x1,x2,x3,x4],-1)).reshape(-1,1),0,1)
        pop.ObjV[:,[6]] = -x1
        pop.ObjV[:,[7]] = -x2
        pop.ObjV[:,[8]] = -x3
```

```
        pop.ObjV[:,[9]] = -x4

    def calReferObjV(self):
        ObjV1 = np.ones((100))
        ObjV2 = np.ones((100))
        ObjV3 = np.ones((100))
        ObjV4 = np.zeros((100))
        ObjV5 = np.zeros((100))
        ObjV6 = np.zeros((100))
        ObjV7 = np.zeros((100))
        ObjV8 = np.zeros((100))
        ObjV9 = np.zeros((100))
        ObjV10 = np.zeros((100))
        globalBestObjV = np.array([ObjV1, ObjV2,ObjV3,ObjV4,ObjV5,ObjV6,ObjV7,ObjV8,ObjV9,ObjV10]).T
        print(globalBestObjV.shape)
        return globalBestObjV

if __name__ == "__main__":
    problem = MyProblem()
    Encoding = 'RI'
    NIND = 50
    Field = ea.crtfld(Encoding, problem.varTypes, problem.ranges, problem.borders)
    population = ea.Population(Encoding, Field, NIND)
    myAlgorithm = ea.moea_NSGA3_templet(problem, population)
    myAlgorithm.drawing = 0
    myAlgorithm.MAXGEN = 50

    NDSet = myAlgorithm.run()
    NDSet.save()

    PF = problem.getReferObjV()
    myAlgorithm.drawing = 1
    if PF is not None:
        metricName = [['IGD']]
        [NDSet_trace, Metrics] = ea.indicator.moea_tracking(myAlgorithm.pop_trace, PF, metricName, problem.maxormins)
```

```
    ea.trcplot(Metrics, labels = metricName, titles = metricName)
result = pd.read_csv('./Result/ObjV.csv',header=None).values
result = np.clip(result,-1,1)
rank_list = []
for i in result:
    value = 1
    for j in i:
        if j >= 0:
            value *= j
        elif j < 0:
            value *= (1 + j)
    rank_list.append(value ** 0.1)
max_index = rank_list.index(max(rank_list))
paras = pd.read_csv('./Result/Phen.csv',header=None).values
f_out = open('final_result.txt','w')
f_out.write('input parameters:' + str(paras[max_index]) + '\n\n')
f_out.write('output responses:' + str([abs(i) for i in result[max_index]]) + '\n')
f_out.close()
```

B. AI-module 2: SVM + NSGA-II + Desirability function

```
import numpy as np
import matplotlib.pyplot as plt
from sklearn.svm import SVR
from matplotlib import cm
import pandas as pd
import matplotlib
import time
import geatpy as ea
import psutil
import os

#start = time.clock()

data = pd.read_excel('CCF design Rifan - 20200303.xlsx')
data = data.iloc[:,[2,3,4,5,6,7,8,9]].dropna()

data_train = data.iloc[1:,:]

data_train = data_train.iloc[:,[0,1,2,3,4]]

ori_features = data_train.iloc[:,:-1].values

minmin = np.min(ori_features,axis=0)
maxmax = np.max(ori_features,axis=0)
features = (ori_features -minmin)/(maxmax-minmin)
labels_4 = data_train.iloc[:,-1].values / 100
labels_5 = data.iloc[1:,5].values / 100
labels_6 = data.iloc[1:,6].values / 100

clf_4 = SVR(gamma=1,C=10,epsilon=0.001)
clf_5 = SVR(gamma=1,C=10,epsilon=0.001)
clf_6 = SVR(gamma=1,C=10,epsilon=0.001)

clf_4 = clf_4.fit(features, labels_4)
clf_5 = clf_5.fit(features, labels_5)
```

```
clf_6 = clf_6.fit(features, labels_6)

data = pd.read_excel('sustainability.xlsx')
data = data.iloc[:,[1,2,3,4,5,6,7]].dropna()
data = data.iloc[1:,:]
data_1 = data.iloc[:,[0,1,2,3,4]]
data_2 = data.iloc[:,[0,1,2,3,5]]
data_3 = data.iloc[:,[0,1,2,3,6]]
ori_features = data_1.iloc[:,:-1].values
minmin = np.min(ori_features,axis=0)
maxmax = np.max(ori_features,axis=0)
features = (ori_features -minmin)/(maxmax-minmin)

labels = data_1.iloc[:,-1].values
label_min = np.min(labels)
label_max = np.max(labels)
labels_1 = (labels - label_min) / (label_max - label_min)

labels = data_2.iloc[:,-1].values
label_min = np.min(labels)
label_max = np.max(labels)
labels_2 = (labels - label_min) / (label_max - label_min)

labels = data_3.iloc[:,-1].values
label_min = np.min(labels)
label_max = np.max(labels)
labels_3 = (labels - label_min) / (label_max - label_min)

clf_1 = SVR(gamma=1,C=1)
clf_2 = SVR(gamma=1,C=1)
clf_3 = SVR(gamma=1,C=1)

clf_1 = clf_1.fit(features, labels_1)
clf_2 = clf_2.fit(features, labels_2)
clf_3 = clf_3.fit(features, labels_3)

result = []
```

```
grid = np.arange(0,1.04,0.04)
max_value = -10
for i in grid:
    for j in grid:
        for k in grid:
            for l in grid:
                value = 1
                a = np.clip(clf_4.predict(np.array([i,j,k,l]).reshape(1,-1)),0,1)
                b = np.clip(clf_5.predict(np.array([i,j,k,l]).reshape(1,-1)),0,1)
                c = np.clip(clf_6.predict(np.array([i,j,k,l]).reshape(1,-1)),0,1)
                d = np.clip(clf_1.predict(np.array([i,j,k,l]).reshape(1,-1)),0,1)
                e = np.clip(clf_2.predict(np.array([i,j,k,l]).reshape(1,-1)),0,1)
                f = np.clip(clf_3.predict(np.array([i,j,k,l]).reshape(1,-1)),0,1)
                one_result = [a,b,c,d,-e,-f,-i,-j,-k,-l]
                for ele in one_result:
                    if ele >= 0:
                        value *= j
                    elif ele < 0:
                        value *= (1 + j)
                value = value ** 0.1
                if value > max_value:
                    max_value = value
                    final_result = one_result
print(final_result)
#end = time.clock()
#print('time:',end-start)
```

3. Response surface plot

3.1. Response surface plot of product quality

The response surface plots from the SVM fits of the parameters on product quality can be visualized in the 4D diagram shown in **Fig. S28**, where the interaction of the four input parameters to each product quality response can be observed.

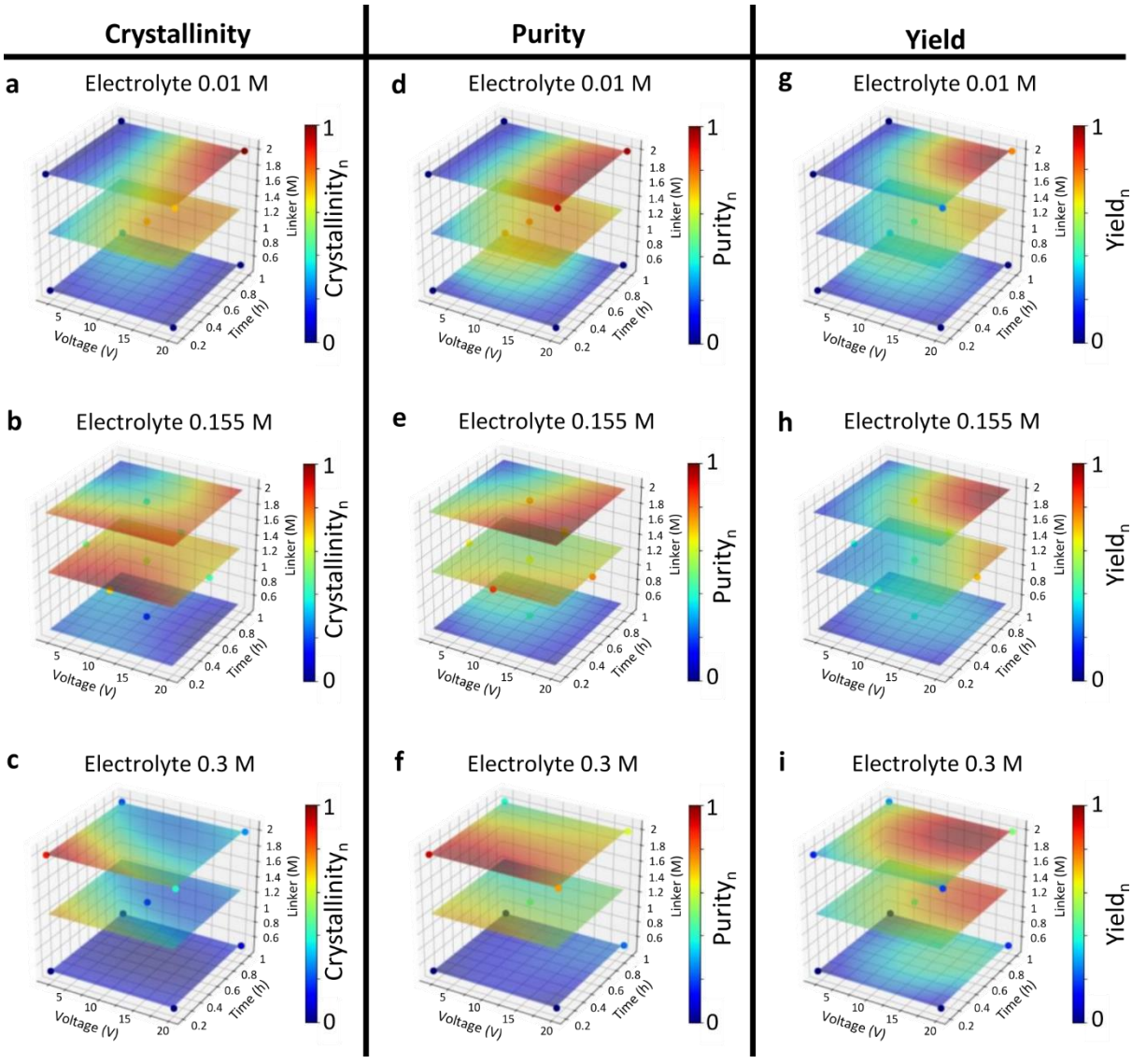

Fig. S28. 4D response surface for product quality. Response surface plots from the SVM fits of the parameters on crystallinity (**a-c**), purity (**d-f**), and yield (**g-i**). C_e is electrolyte concentration, V is applied voltage, T is reaction time, C_l is linker concentration. The values of the responses are normalized, where zero and one are the lowest and the highest, respectively (refer to **Table S3**). The color bar indicates the degree of the responses, where dark red and dark blue indicate the highest and the lowest response values, respectively. The sampling points generated from the DoE are placed as the dots in each surface plot. The 4D plot consists of 456,976 data points generated through the predictive machine learning through the SVM surrogate function.

Fig. S28a-c shows that as the electrolyte concentration increased, the crystallinity decreased when the voltage increased. This suggests the existence of an interaction between the electrolyte and voltage towards crystallinity. The same observation applies between reaction time and electrolyte concentration.

In **Fig. S28d-f**, the response surface of the parameters towards purity is presented. A similar trend is observed as that for the crystallinity, where interaction between the electrolyte with voltage and reaction time present. This implies that the same scenarios that maximize the crystallinity also apply to maximize purity. Both crystallinity and purity can be boosted by maximizing the voltage, reaction time, and linker concentration while minimizing the electrolyte.

Taken together, it can be interpreted that high voltage produces more zinc cations as the source of the metal node to form MOFs. A longer reaction time allows enough time for nucleation and crystal growth to occur. A high linker concentration can ensure that there is enough linker to form a framework with the metal cations. Meanwhile, excessive electrolyte concentration seems to inhibit MOFs formation due to a possible "salting out" process, as we speculated earlier. Finally, this situation decreases the MOFs crystallinity and increases the contaminants, which decreases the purity.

Fig. S28g-i presents the response surface parameters towards yield. Interestingly, these figures show a different trend to that of crystallinity and purity. These figures show that the maximum area of yield is always observed at high voltage, high linker concentration, and long reaction time, regardless of the electrolyte concentration. These observations imply that by considering parameter interactions, the electrolyte concentration seems to have the smallest effect on the yield.

3.2. Response surface plot of process quality

The response surface plots from the SVM fits of the parameters on process sustainability can be visualized in 4D diagrams, as shown in **Fig. S29**, where the interaction of four input parameters with each process sustainability response can be observed.

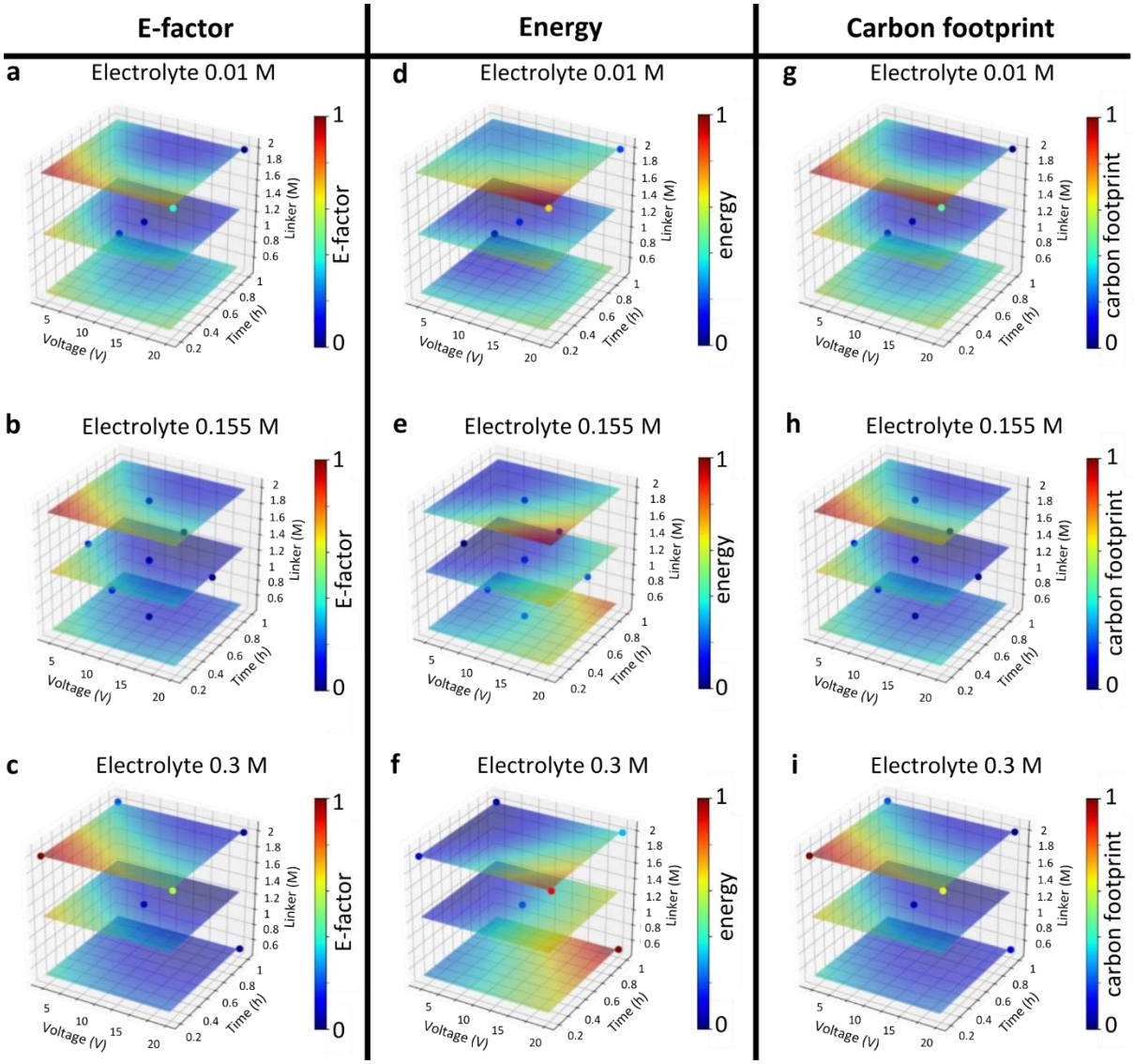

Fig. S29. 4D response surface for process sustainability. Response surface plots from the SVM fits of the parameters on E-factor (**a-c**), Energy (**d-f**), and Carbon footprint (**g-i**). C_e is electrolyte concentration, V is applied voltage, T is reaction time, C_l is linker concentration. The values of the responses are normalized, with zero and one being the lowest and the highest, respectively (refer to **Table S3**). The color bar indicates the degree of the responses, where dark red and dark blue indicate the highest and the lowest response values, respectively. The sampling points generated from the DoE are placed as the dots in each surface plot. The 4D plot consists of 456,976 data points generated through the predictive machine learning through the SVM surrogate function.

In **Fig. S29a-c**, the most desired regions with the lowest E-factors are indicated by the blue area in the surface plots. It can be seen that high linker concentration, low voltage, and short reaction time do not minimize the environmental impact. An explanation could be that at low voltage and short reaction time, there is not enough metal cations, driving force, and time to form the MOFs. As a result, excessive linker concentrations become waste, which causes increasing E-factor.

As shown in **Fig. S29d-f**, the response surface plots exhibit a different trend to that of the E-factor. A combination of high voltage, short reaction time, high linker concentration, and high electrolyte concentration are not preferred to minimize energy consumption. A plausible argument could be that increasing the electrolyte at high voltage and a short reaction time decreases the solubility of the linker, thus requiring more energy to dissolve the linker. Prolonging the reaction time while increasing the linker concentration seems to decrease the energy used during the MOFs synthesis. This is because a longer reaction time and an increased linker concentration drive the reaction to the formation of the desired MOFs product, thus utilizing the energy more efficiently and resulting in an overall decrease in the energy consumption per generated product.

Interestingly, the response surface plots of carbon footprint (**Fig. S29g-i**) are almost identical to the E-factor. The carbon footprint represents the CO_2 emissions equivalent from the generated waste and energy consumption. This similarity of the response surface between E-factor and carbon footprint can be explained because the dominant content in both E-factor and carbon footprint is the same, which is the waste from the unreacted linker.

4. Optimization strategy

An optimization approach that transforms multiple variables into one variable is known as "Derringer's Desirability function".[12–14] Desirability criteria can be set depending on the requirement, i.e., whether the observed variables should be maximized or minimized. In the first step, an individual desirability function ($d_{i,}$) for each observed variable (y_i) must be created using the suitable formula, equation (S12) and equation (S13). Desirability has values between 0 and 1, where ($d_{i,}$) = 0 indicates an undesirable response, and ($d_{i,}$) = 1 represents a completely desirable value. If the observed variables must be maximized, $d_{i,max}$ is expressed in equation (S12).

$$d_{i,max} = \begin{bmatrix} 0 & \text{if } y_i < L_i \\ \left(\frac{y_i - L_i}{U_i - L_i}\right)^s & \text{if } L_i \le y_i \le U_i \\ 1 & \text{if } y_i > U_i \end{bmatrix} \quad \text{(S12)}$$

where U_i is the highest value for the response, L_i is the lowest limit of the value, and s is a power value named "weight", set by the analyst to determine how important it is for y_i to be close to the maximum. The equation for $d_{i,min}$ when it has to be minimized is expressed in equation (S13).

$$d_{i,min} = \begin{bmatrix} 1 & \text{if } y_i < L_i \\ \left(\frac{U_i - y_i}{U_i - L_i}\right)^s & \text{if } L_i \le y_i \le U_i \\ 0 & \text{if } y_i > U_i \end{bmatrix} \quad \text{(S13)}$$

Once all the variables are transformed into desirability functions, they are combined to reveal the best joint responses in Global Desirability (D) using equation (S14):

$$D = (d_1^{r_1} \times d_2^{r_2} \times \ldots\ldots \times d_n^{r_n})^{\frac{1}{\Sigma r_i}} \quad (14)$$

where r_i is the importance of each variable relative to the other variables. In this case, since all the variables are equally important, r equals 1 for each individual desirability.

In this work, all the variables were assumed to be equally important, and two optimization objectives are presented. In the first optimization objective, only product quality (crystallinity, purity, and yield) was considered to be maximized. The results are tabulated in **Table S10**. In the second optimization objective, both product quality (crystallinity, purity, and yield) and process sustainability (E-factor, energy, carbon footprint) were considered for optimization, and the results are given in **Table 11**.

Table S10. Optimization objective 1. The goal of optimization objective 1 is to maximize the product quality (crystallinity, purity, and yield)

Variable	Goal	Lower limit	Upper limit	Result	d_i
c_e	in range	0.01	0.30	0.04	1.00
V	in range	2.00	20.00	20.00	1.00
t	in range	0.20	1.00	0.90	1.00
c_l	in range	0.50	2.00	1.94	1.00
Crystallinity	maximize	0.00	100.00	93.34	0.93
Purity	maximize	0.00	100.00	100.00	1.00
Yield	maximize	0.00	100.00	87.79	0.88
E-factor	in range	6.50	251.80	22.42	1.00
Energy	in range	0.84	28.23	7.85	1.00
Carbon footprint	in range	15.45	476.90	46.13	1.00
Global desirability					0.98

The desirability function of input parameters was set to one as the goal for these was to be in range, which implies having a desirability of one whenever the result is within the defined range. Furthermore, the calculation of the global desirability was computed, and the results are shown in **Table S11**.

Table S11. Optimization objective 2. The goal of optimization objective 2 is to maximize the product quality (crystallinity, purity, and yield) and minimize the process sustainability (E-factor, energy, and carbon footprint).

Variable	Goal	Lower limit	Upper limit	result	d_i
c_e	in range	0.01	0.30	0.07	1.00
V	in range	2.00	20.00	18.56	1.00
t	in range	0.20	1.00	0.90	1.00
c_l	in range	0.50	2.00	1.76	1.00
Crystallinity	maximize	0.00	100.00	84.58	0.85
Purity	maximize	0.00	100.00	99.96	1.00
Yield	maximize	0.00	100.00	88.30	0.88
E-factor	minimize	6.50	251.80	9.34	0.99
Energy	minimize	0.84	28.23	6.47	0.79
Carbon footprint	minimize	15.45	476.90	19.43	0.99
Global desirability					0.95

The applied methodologies (both AI Module 1 and AI Module 2) were able to generate a single optimum condition. The single solution of the optimum condition for each optimization objective with different

methodologies is compared in **Table S12**. The normalized and true value of the variables at each optimization objective, which corresponds with **Table S12**, are also represented as bar charts, as illustrated in **Fig. S30**.

Table S12. Single optimum solution. True values of all variables for each scenario with different approaches

Variable	**Objective 1**				**Objective 2**			
	Target	DoE	AI Module 1	AI Module 2	Target	DoE	AI Module 1	AI Module 2
Electrolyte (M)	in range	0.01	0.04	0.04	in range	0.01	0.07	0.07
Voltage (V)	in range	20.00	20.00	20.00	in range	20.00	18.56	18.54
Time (h)	in range	1.00	0.90	0.91	in range	1.00	0.90	0.93
Linker (M)	in range	2.00	1.94	1.94	in range	2.00	1.76	1.86
Crystallinity (%)	Max	100.00	93.34	94.71	Max	100.00	84.58	85.64
Purity (%)	Max	97.60	100.00	100.00	Max	97.60	99.96	100.00
Yield (%)	Max	77.00	87.79	86.53	Max	77.00	88.30	89.18
E-factor (kg kg^{-1})	in range	6.71	22.42	21.21	min	6.71	9.34	11.24
Energy (kWh kg^{-1})	in range	6.14	7.85	7.96	min	6.14	6.47	6.81
Carbon footprint (kg kg^{-1})	in range	19.15	46.13	52.36	min	19.15	19.43	27.03

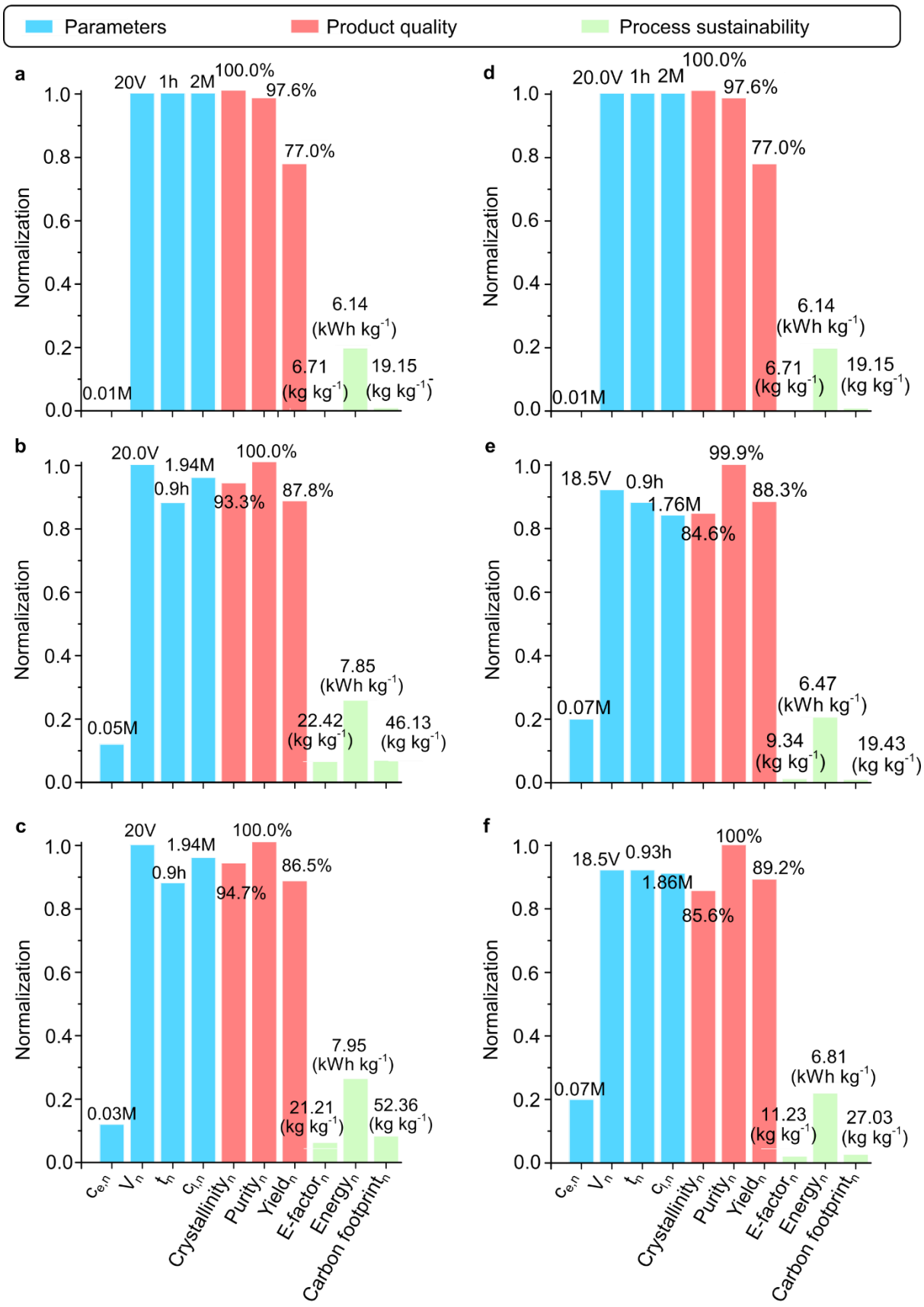

Fig. S30. Normalized and true values of the variables as results of DoE, AI Module 1 and AI Module 2 at optimization objective 1 (**a-c**)), and objective 2 (**d-f**).

5. Structure and morphology characterizations

The XRD patterns and SEM images of the electrochemical products tabulated in **Table S4** are presented in **Fig. S31-S49**.

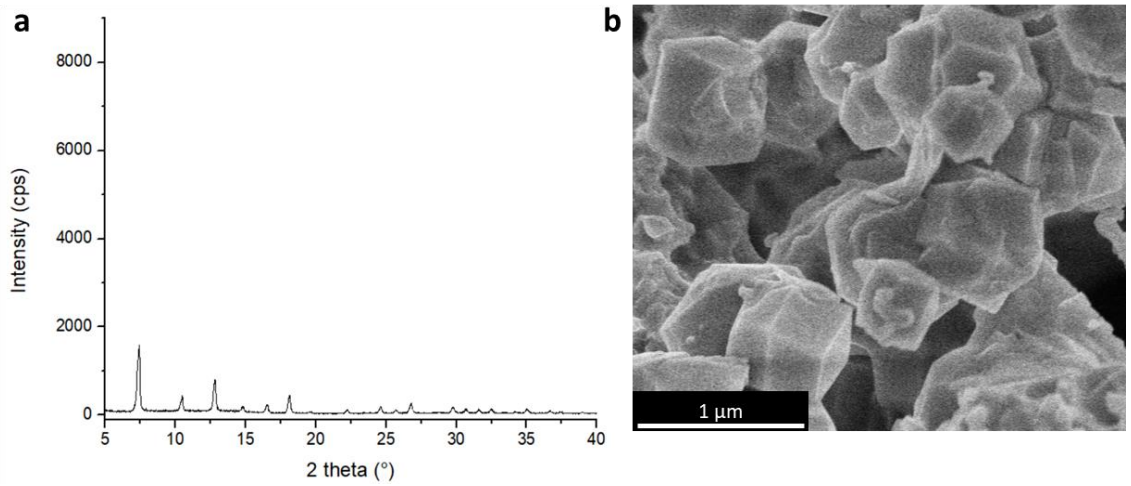

Fig. S31. XRD pattern (a) and SEM image (b) of a sample from Entry 5.

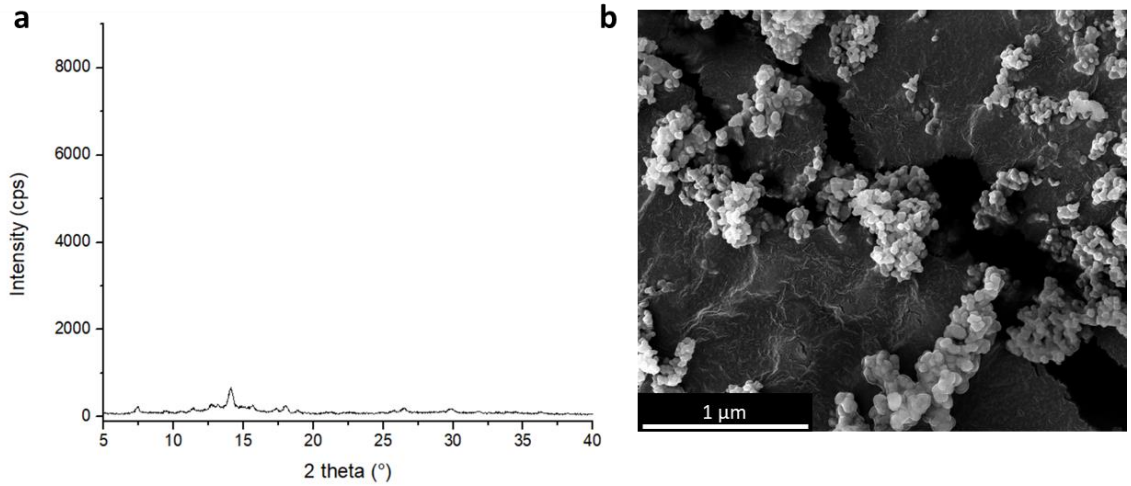

Fig. S32. XRD pattern (a) and SEM image (b) of a sample from Entry 8

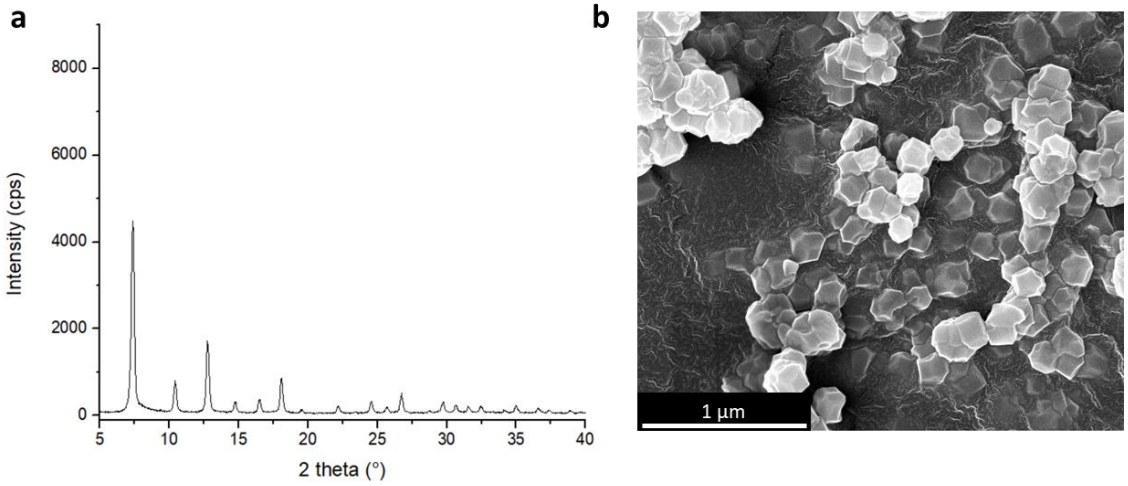

Fig. S33. XRD pattern (a) and SEM image (b) of a sample from Entry 10

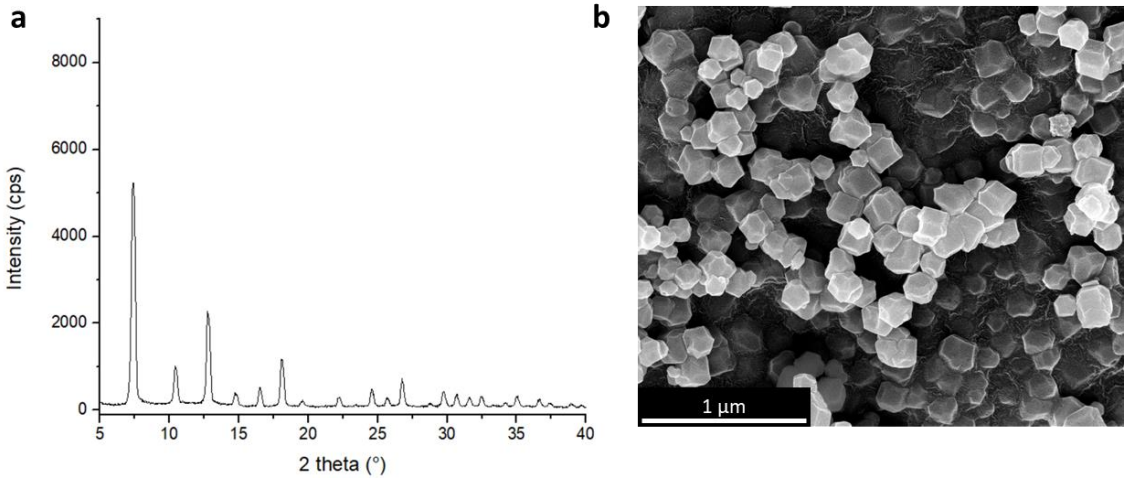

Fig. S34. XRD pattern (a) and SEM image (b) of a sample from Entry 11

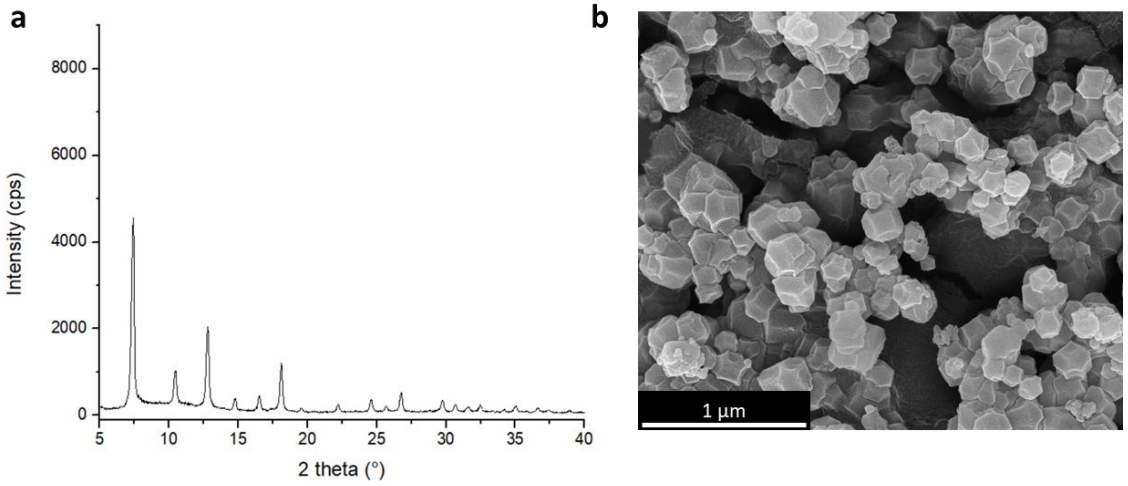

Fig. S35. XRD pattern (a) and SEM image (b) of a sample from Entry 12

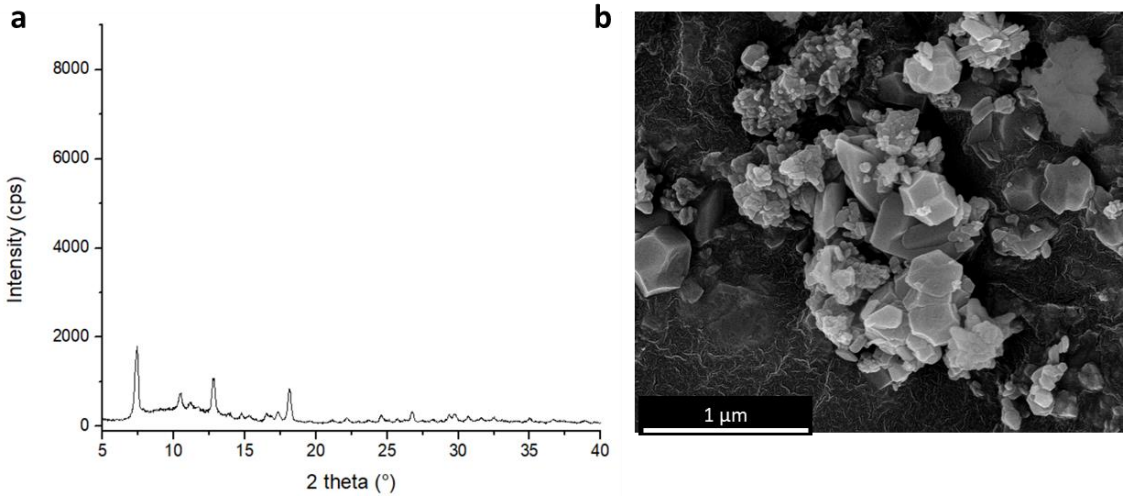

Fig. S36. XRD pattern (a) and SEM image (b) of a sample from Entry 14

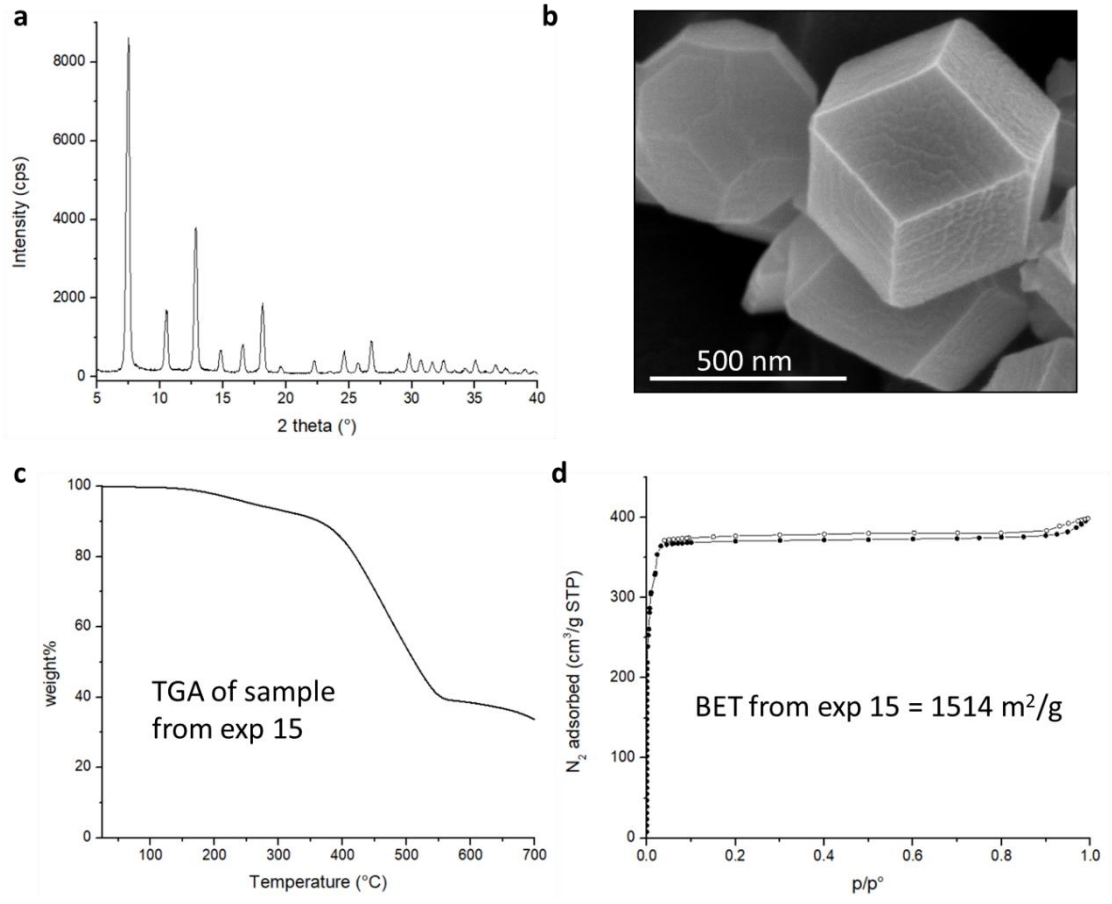

Fig. S37. XRD pattern (a), SEM image (b), TGA (c), and BET measurement (d) of a sample from Entry 15

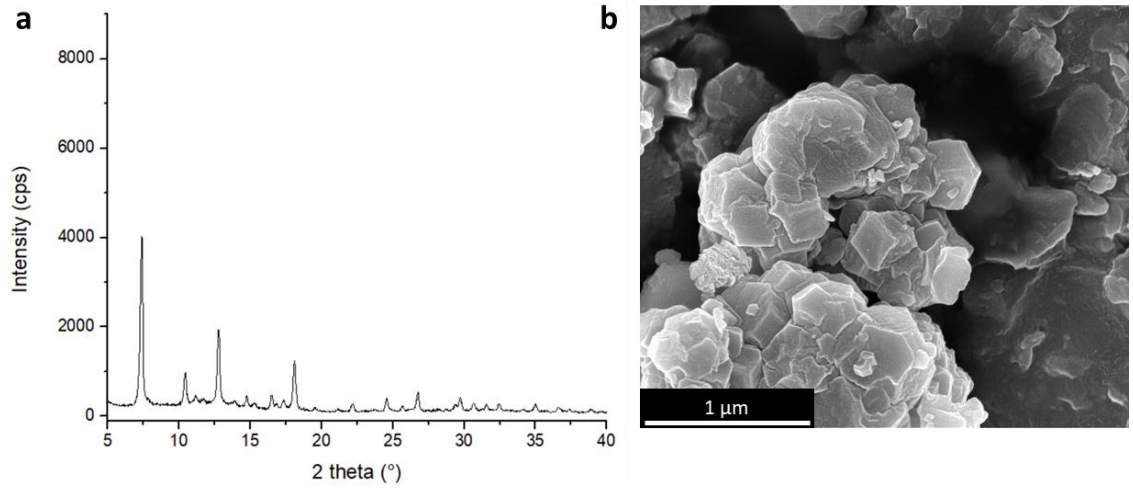

Fig. S38. XRD pattern (a) and SEM image (b) of a sample from Entry 16

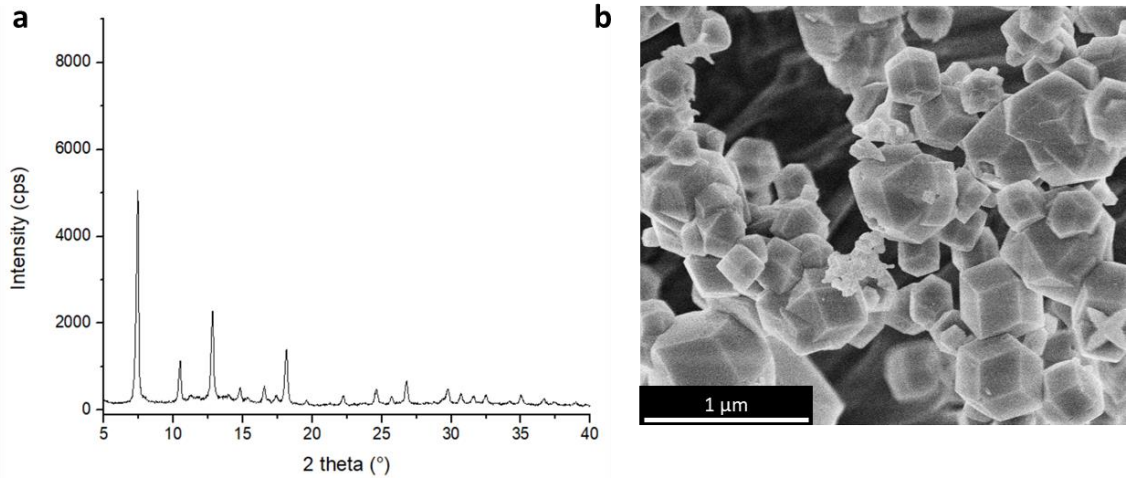

Fig. S39. XRD pattern (a) and SEM image (b) of a sample from Entry 17

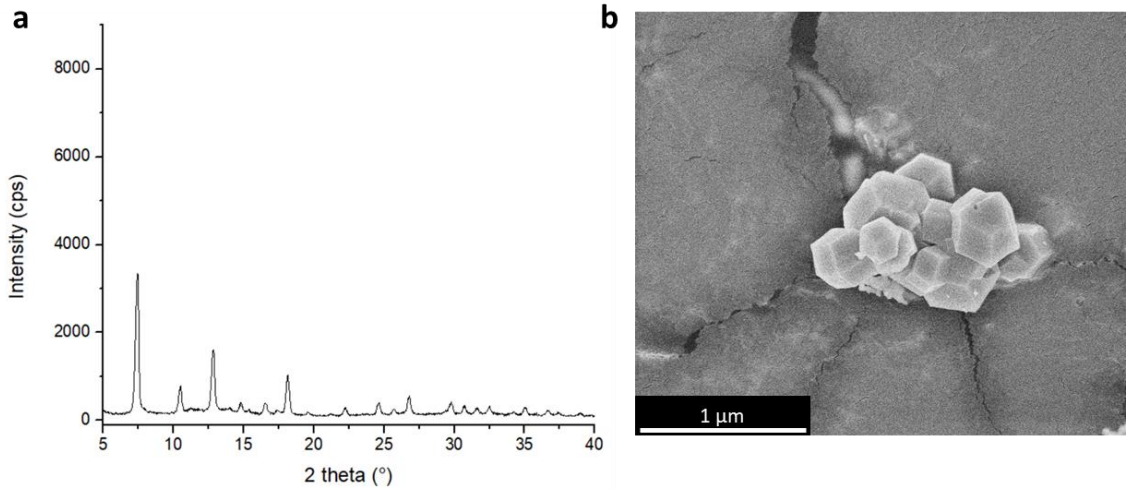

Fig. S40. XRD pattern (a) and SEM image (b) of a sample from Entry 18

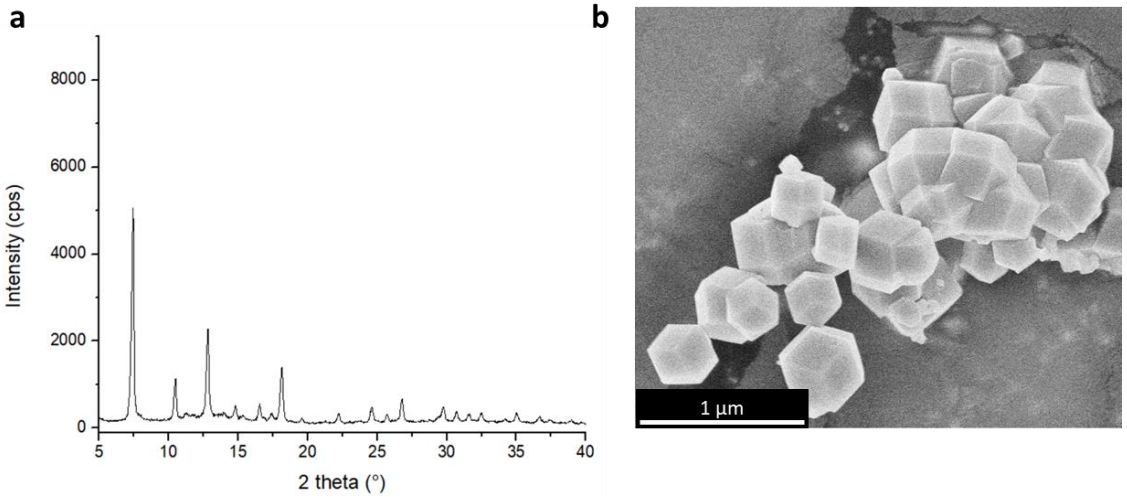

Fig. S41. XRD pattern (a) and SEM image (b) of a sample from Entry 19

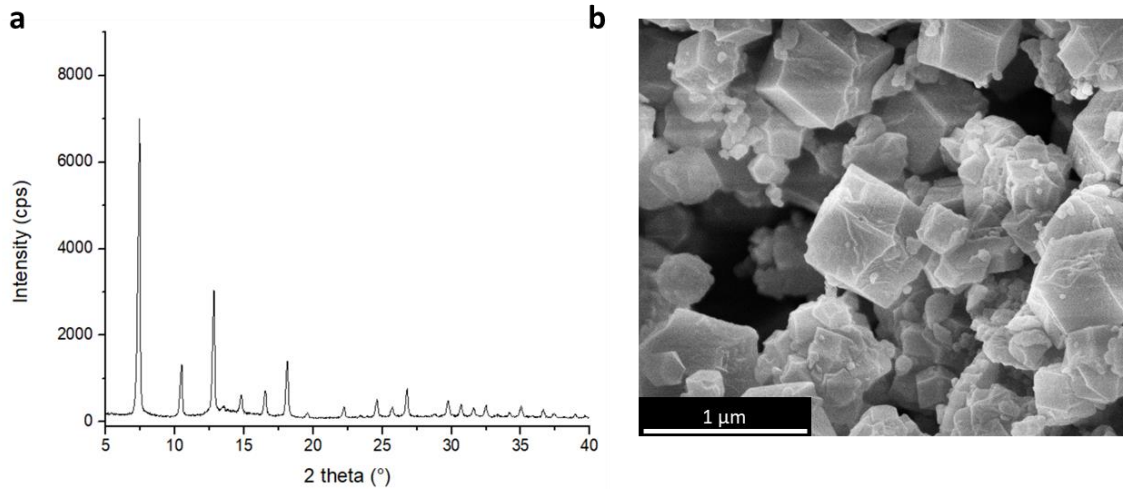

Fig. S42. XRD pattern (a) and SEM image (b) of a sample from Entry 20

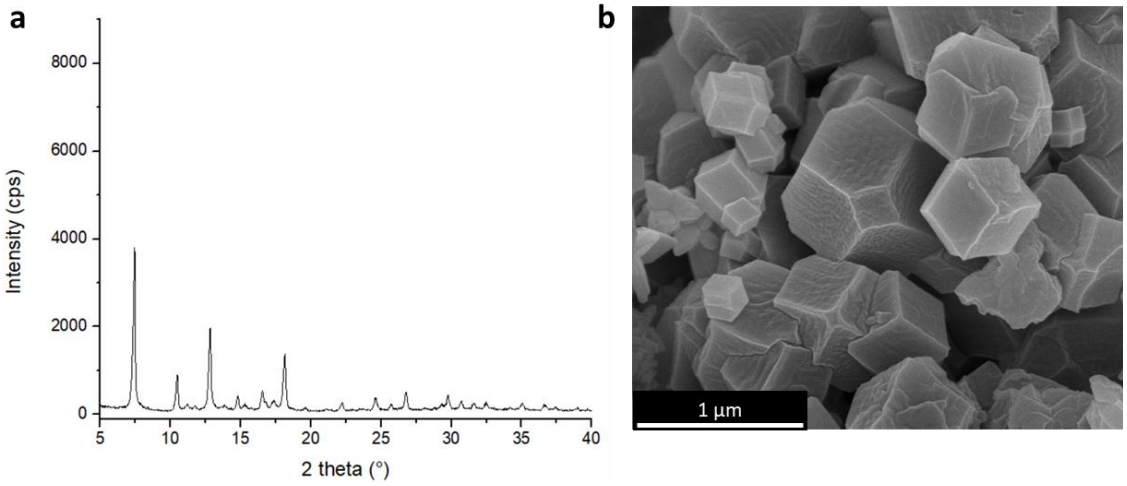

Fig. S43. XRD pattern (a) and SEM image (b) of a sample from Entry 21

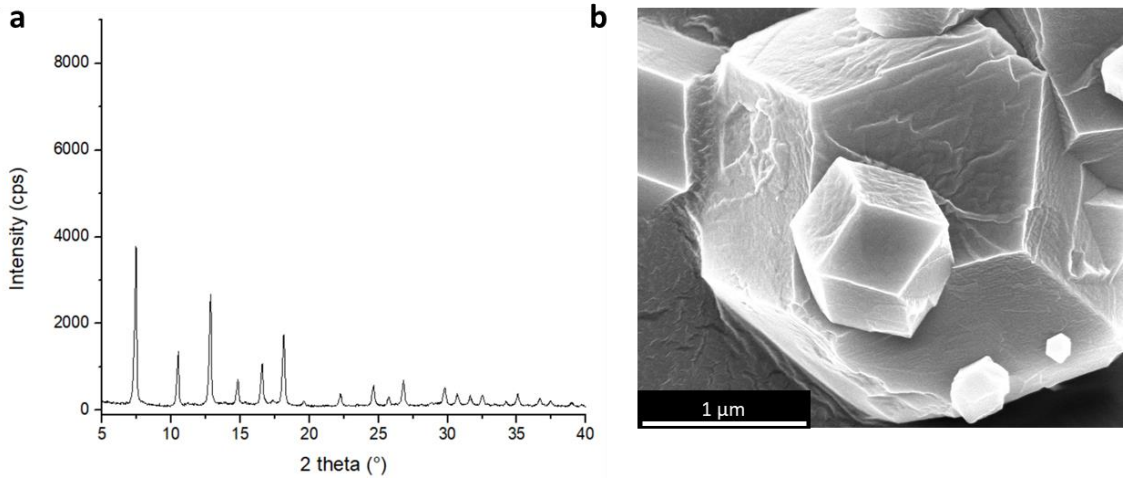

Fig. S44. XRD pattern (a) and SEM image (b) of a sample from Entry 22

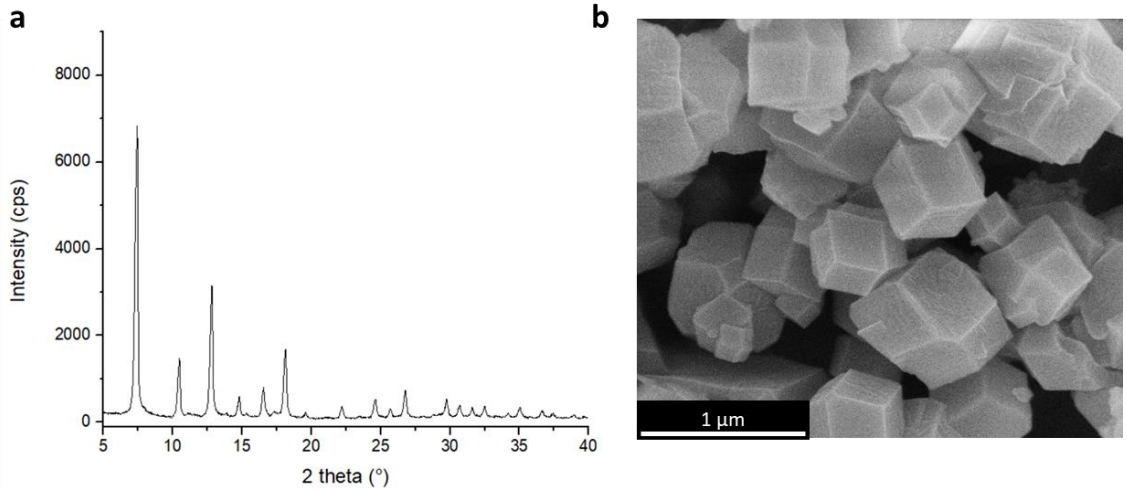

Fig. S45. XRD pattern (a) and SEM image (b) of a sample from Entry 23

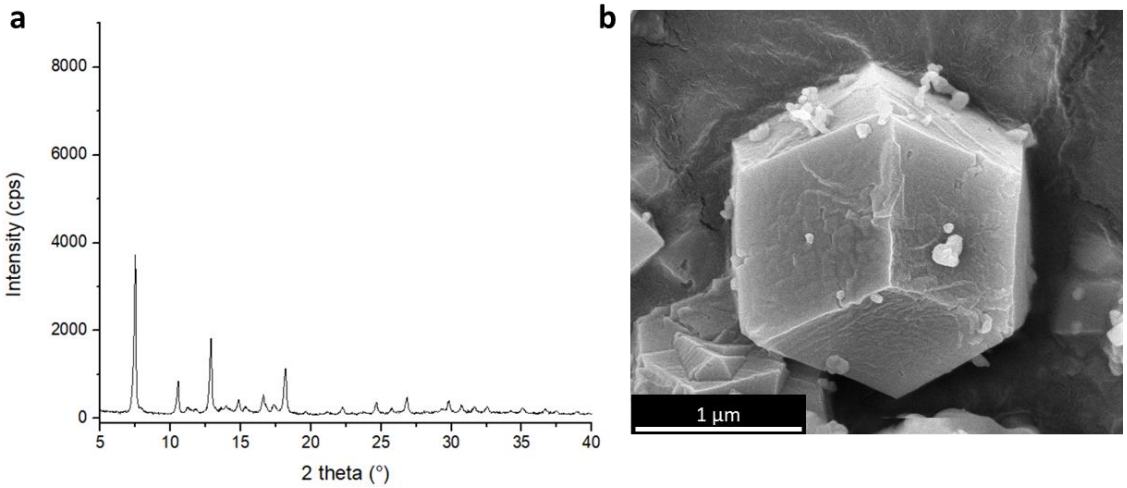

Fig. S46. XRD pattern (a) and SEM image (b) of a sample from Entry 24

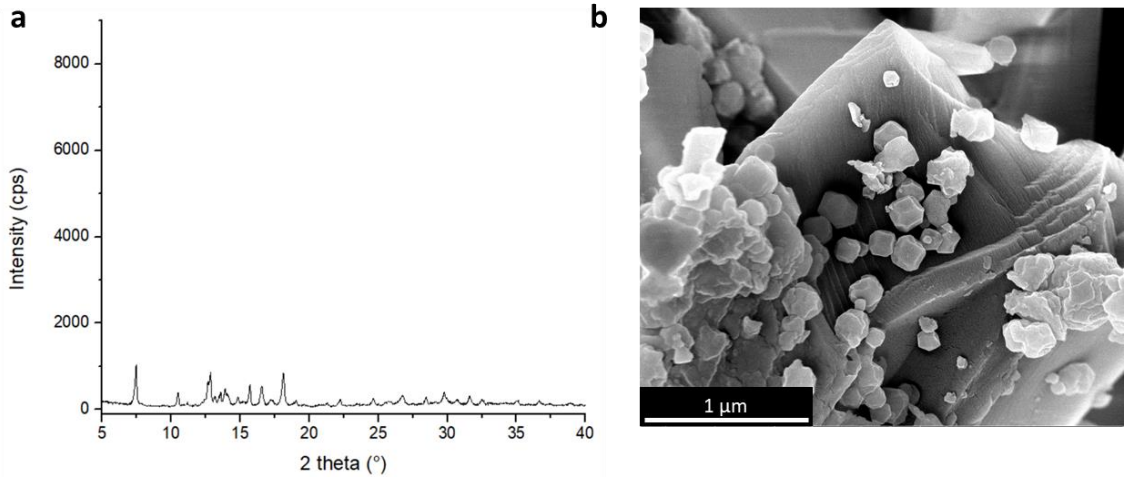

Fig. S47. XRD pattern (a) and SEM image (b) of a sample from Entry 25

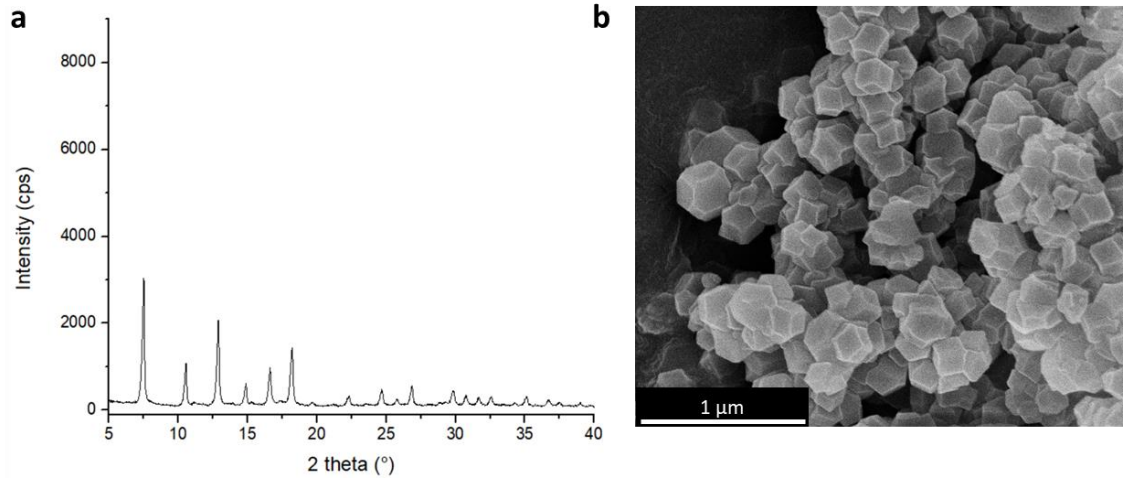

Fig. S48. XRD pattern (a) and SEM image (b) of a sample from Entry 26

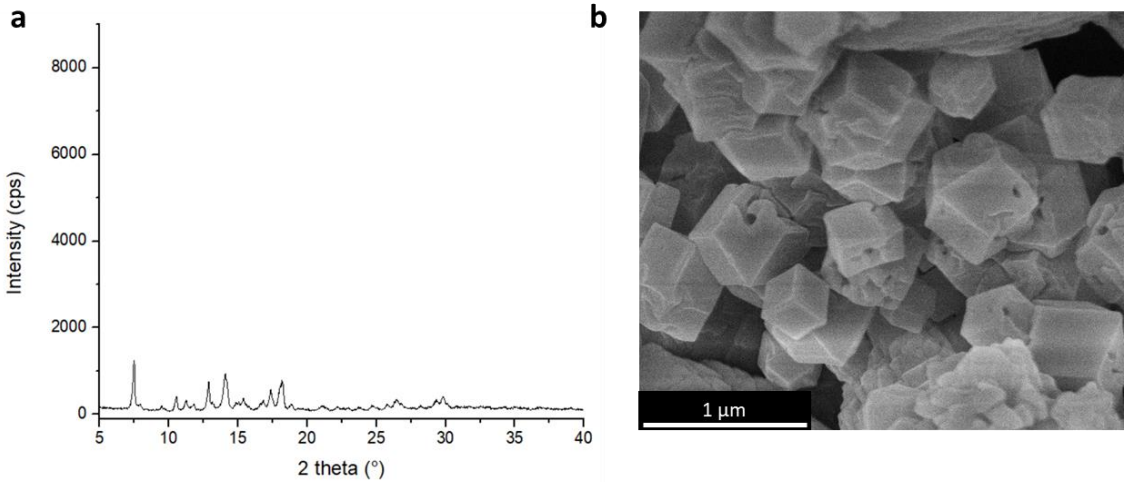

Fig. S49. XRD pattern (a) and SEM image (b) of a sample from Entry 27.

6. References

(1) Jain, S.; Shukla, S.; Wadhvani, R. Dynamic Selection of Normalization Techniques Using Data Complexity Measures. *Expert Systems with Applications* **2018**, *106*, 252–262.

(2) Antony, J. *Design of Experiments for Engineers and Scientists*; Elsevier, 2014.

(3) Baxter, E. F.; Bennett, T. D.; Cairns, A. B.; Brownbill, N. J.; Goodwin, A. L.; Keen, D. A.; Chater, P. A.; Blanc, F.; Cheetham, A. K. A Comparison of the Amorphization of Zeolitic Imidazolate Frameworks (ZIFs) and Aluminosilicate Zeolites by Ball-Milling. *Dalton Trans.* **2016**, *45* (10), 4258–4268.

(4) Bennett, T. D.; Sotelo, J.; Tan, J.-C.; A. Moggach, S. Mechanical Properties of Zeolitic Metal–Organic Frameworks: Mechanically Flexible Topologies and Stabilization against Structural Collapse. *CrystEngComm* **2015**, *17* (2), 286–289.

(5) Guillerm, V.; Ragon, F.; Dan-Hardi, M.; Devic, T.; Vishnuvarthan, M.; Campo, B.; Vimont, A.; Clet, G.; Yang, Q.; Maurin, G.; Férey, G.; Vittadini, A.; Gross, S.; Serre, C. A Series of Isoreticular, Highly Stable, Porous Zirconium Oxide Based Metal–Organic Frameworks. *Angew. Chem.* **2012**, *124* (37), 9401–9405.

(6) Sheldon, R. A. The E Factor 25 Years on: The Rise of Green Chemistry and Sustainability. *Green Chem.* **2017**, *19* (1), 18–43.

(7) Maity, P.; Gopinath, C. S.; Bhaduri, S.; Lahiri, G. K. Applications of a High Performance Platinum Nanocatalyst for the Oxidation of Alcohols in Water. *Green Chem.* **2009**, *11* (4), 554–561.

(8) Scott, K. *Sustainable and Green Electrochemical Science and Technology*, 1st ed.; John Wiley & Sons, Ltd, 2017.

(9) Vincent, C. *Modern Batteries: An Introduction to Electrochemical Power Sources*, *2nd Edition*, 2 edition.; Butterworth-Heinemann: London : New York, 1997.

(10) Treptow, R. S. Carbon Footprint Calculations: An Application of Chemical Principles. *J. Chem. Educ.* **2010**, *87* (2), 168–171.

(11) Bagherzadeh, E.; Zebarjad, S. M.; Hosseini, H. R. M.; Chagnon, P. Preparation, Optimization and Evolution of the Kinetic Mechanism of an Fe-MIL-88A Metal–Organic Framework. *CrystEngComm* **2019**, *21* (3), 544–553.

(12) Derringer, G.; Suich, R. Simultaneous Optimization of Several Response Variables. *Journal of Quality Technology* **1980**, *12* (4), 214–219.

(13) Vera Candioti, L.; De Zan, M. M.; Cámara, M. S.; Goicoechea, H. C. Experimental Design and Multiple Response Optimization. Using the Desirability Function in Analytical Methods Development. *Talanta* **2014**, *124*, 123–138.

(14) Heidari, H.; Razmi, H. Multi-Response Optimization of Magnetic Solid Phase Extraction Based on Carbon Coated Fe3O4 Nanoparticles Using Desirability Function Approach for the Determination of the Organophosphorus Pesticides in Aquatic Samples by HPLC–UV. *Talanta* **2012**, *99*, 13–21.

(15) https://ecometrica.com/assets/Electricity-specific-emission-factors-for-grid-electricity.pdf.

(16) http://www.ccalc.org.uk/