Supplementary Information:

Disentangling Multifactorial Impacts on Cathode Thermochemical Properties with Explainable Machine Learning

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I. DETAILS OF INPUT/OUTPUT VARIABLES

A. Input features

Detailed descriptions of all input feature variables classified into testing parameters, sample conditions, and material properties, are listed in Tab. S1.

TABLE S1: Input feature descriptions

Category	**Feature name**	**Description**
Testing parameters	Cutoff voltage	The voltage to which the cathode material sample to be tested is charged.
	DSC scanning speed	Temperature rising speed in the DSC test.
Sample conditions	Crucible type	Whether the test is carried out in an open or closed crucible.
	Crystal morphology	Whether the tested material is monocrystal or polycrystal.
	Active substance ratio	The mass ratio of active cathode materials in the tested cathode sample.
	Electrolyte content	The ratio of electrolyte weights to cathode weights for the DSC test.
	Electrolyte composition	The degree of electrolyte composition changes. The blank group is $LiPF_6$ in EC:(DEC/EMC/DMC)=3:7 to 1:1, labeled 0. For those with composition change not exceeding 10% or only the EC/DMC ratio adjusted, it is considered a low-degree modification and labeled 1. If more than 10% or an additive is added based on the adjusted EC/DMC ratio, it is considered highly modified and labeled 2. Samples without electrolytes are labeled 3.
	Electrolyte modification	Quantification of new chemical substances introduced. The blank group is $LiPF_6$ in EC:(DEC/EMC/DMC)=3:7 to 1:1, labeled 0. If the EC/DMC content is changed or the additive is still free of fluorine esters, it is considered a low-degree modification and labeled 1. Those containing non-esters or functional groups such as fluorine or phosphate are considered highly modified and labeled 2. Samples without electrolytes are labeled 3.
	Reversible capacity	The reversible specific discharge capacity of the cathode material.

Material properties	Surface modification	Whether the material has been surface modified.
	Bulk phase doping	Whether the material has been doped in the bulk phase.
	Anion doping	Whether the fluorine or sulfur anions are introduced in the material.
	Pristine Li content	The stoichiometric ratio of Li in the pristine material.
	Ni content	The stoichiometric ratio of Ni in the material.
	Co content	The stoichiometric ratio of Co in the material.
	Mn content	The stoichiometric ratio of Mn in the material.
	EN-TM ave (wt.)	The average electronegativity of transition metal species, weighted by their stoichiometric ratios.
	EN-TM std (wt.)	The electronegativity standard deviation of transition metal species, weighted by their stoichiometric ratios.
	EN-TM ave (unwt.)	The average electronegativity of transition metal species, unweighted by their stoichiometric ratios.
	EN-TM std (unwt.)	The electronegativity standard deviation of transition metal species, unweighted by their stoichiometric ratios.
	Radii-TM ave (wt.)	The average ionic radii of transition metal species, weighted by their stoichiometric ratios.
	Radii-TM std (wt.)	The ionic radii standard deviation of transition metal species, weighted by their stoichiometric ratios.
	Radii-TM ave (unwt.)	The average ionic radii of transition metal species, unweighted by their stoichiometric ratios.
	Radii-TM std (unwt.)	The ionic radii standard deviation of transition metal species, unweighted by their stoichiometric ratios.
	Φ-TM ave (wt.)	The average ionic potential of transition metal species, weighted by their stoichiometric ratios.
	Φ-TM std (wt.)	The ionic potential standard deviations of transition metal species, weighted by their stoichiometric ratios.
	Φ-TM ave (unwt.)	The average ionic potential of transition metal species, unweighted by their stoichiometric ratios.
	Φ-TM std (unwt.)	The ionic potential standard deviations of transition metal species, unweighted by their stoichiometric ratios.

Some additional notes need to be addressed:

1. The transition metals mentioned in the last 12 features refer to cation species occupying transition metal sites in the layered cathode materials. Therefore, some modification elements, such as boron and phosphorous, are also included in this scope, even though they are not genuine transition metals.
2. When calculating the ionic radii and ionic potentials, we assume the transition metal ions to be at their highest oxidation state when the material is delithiated. This is because the tested cathode material samples are usually taken from a charged cell that is at its highest thermal runaway risk.
3. Logarithmic transitions are applied to DSC scanning speed and electrolyte content for a more uniform data distribution that will benefit regression analysis. All the feature values are standardized with a mean value of 0 and a standard deviation of 1 before doing the regression.

B. Output characteristics

Detailed descriptions of all output characteristics are shown in Tab. S2.

TABLE S2. Output characteristic descriptions

Characteristic name	**Description**
Onset temperature	The temperature at the intersection of the starting tangent of the first significant (human-judged) heat release peak and the baseline.
Peak temperature	The temperature at which the heat release power is at its maximum in the first significant heat release peak.
Max power	The maximum heat release power along the entire thermal analysis curve (minus baseline).

II. DETAILS OF FEATURE SELECTION

A. Correlation coefficients

According to the calculated Spearman correlation coefficients demonstrated in main text Fig. 1a and the criterion of |Spearman coefficient|>0.8, as well as the principle of "maintaining simpler and more fundamental features", the eliminated features in this step are:

1. Electrolyte modification
2. EN-TM ave (wt.)
3. EN-TM std (wt.)
4. Radii-TM ave (wt.)
5. Φ-TM ave (wt.)
6. Φ-TM std (wt.)
7. Φ-TM ave (unwt.)
8. Φ-TM std (unwt.)

B. Null importance

According to the null importance feature screening strategy described in our main text, features whose actual importance falls below the first quartile (Q_1) of null importance values are eliminated. We therefore calculate the null importance scores of each feature variable to every output characteristic according to the formula below:

$$Score = \ln \frac{Importance_{Actual} + 1e^{-10}}{Q_1(Importances_{Null}) + 1}$$

Here the e^{-10} in the numerator prevents the logarithm of zero, and the 1 in the denominator prevents division by zero. The expression ensures that the features to be discarded get negative scores. We list the calculated scores of each feature variable to every output characteristic in Tab. S3. The split and gain scores are calculated respectively based on the split and gain importance. In our practice, only those features getting both negative split and gain scores are discarded, colored red in Tab. S3.

The eliminated non-important features for three thermochemical characteristics are summarized below:

1. For **onset temperature**:
   (a) DSC scanning speed
   (b) Active substance ratio
   (c) Bulk phase doping
   (d) Anion doping
   (e) Radii-TM ave (unwt.)

2. For **peak temperature**:
    (a) Bulk phase doping
    (b) Anion doping
3. For **max power**:
    (a) Anion doping
    (b) Mn content
    (c) EN-TM ave (unwt.)
    (d) EN-TM std (unwt.)
    (e) Radii-TM ave (unwt.)
    (f) Radii-TM std (unwt.).

TABLE S3. Feature null importance scores

Feature name	Null importance score					
	Onset temperature		Peak temperature		Max power	
	Split	Gain	Split	Gain	Split	Gain
Cutoff voltage	0.263111	1.559504	-0.1202	0.48759	0.057158	0.13869
DSC scanning speed	-0.21072	-0.32365	0.265376	0.794231	0.980829	2.578186
Crucible type	0.413833	0.266848	0.775839	2.425989	0.265703	3.063481
Crystal morphology	0.09646	-0.0387	-0.1431	0.251787	-0.0685	0.028662
Active substance ratio	-0.07496	-0.12471	0.35315	0.845383	0.182824	1.407127
Electrolyte content	0.030283	-0.11579	-0.33361	0.483575	0.652897	3.622218
Electrolyte composition	0.712458	0.975264	0.724238	2.587193	-0.3514	2.601453
Reversible capacity	-0.28338	0.614095	-0.24256	0.35015	-0.11401	0.288674
Surface modification	0.318141	0.376506	-0.09937	0.21979	-0.10536	0.026612
Bulk phase doping	-0.07522	-0.29504	-0.29865	-0.02434	-0.14147	0.26752
Anion doping	-23.0259	-23.0259	-23.0259	-23.0259	-23.0259	-23.0259
Pristine Li content	-0.1226	1.069188	0.510826	1.491625	0.231802	0.122433
Ni content	0.088193	1.295852	0.959929	3.268181	0.27029	0.905089
Co content	-0.61856	0.713477	0.001714	2.006067	0.528566	1.561223
Mn content	0.909163	3.719137	0.240336	1.667375	-0.04652	-0.12195
EN-TM ave (unwt.)	-0.05763	0.975004	-0.15415	0.010162	-0.64265	-1.09932
EN-TM std (unwt.)	0.541341	1.595916	0.323787	0.846341	-0.21131	-0.34964
Radii-TM std (wt.)	0.278816	2.211943	-0.25464	1.357604	0.364476	0.694274
Radii-TM ave (unwt.)	-0.30673	-0.26339	-0.17768	0.017749	-0.51941	-0.26557
Radii-TM std (unwt.)	0.280539	0.964863	0.559616	1.146372	-0.17768	-0.16003

III. TRAINING DETAILS

Five-sixth of the data are randomly chosen for model training, with the rest one-sixth for testing. We exhaustively search the SVR model parameters using the `GridSearchCV` tool in the `scikit-learn` library. 5-fold cross-validation is used to assess the performance of each hyperparameter combination included in the parameter grid defined below:

```
param_grid = {
    "C": [10**x for x in range(-1, 3)],
    "kernel": ["linear", "poly", "rbf", "sigmoid"],
    "gamma": ["auto"],
}
```

The best hyperparameter combination for each output characteristic are:

- For **onset temperature**: {`"C": 100, "gamma": "auto", "kernel": "rbf"`}
- For **peak temperature**: {`"C": 100, "gamma": "auto", "kernel": "rbf"`}
- For **max power**: {`"C": 10, "gamma": "auto", "kernel": "rbf"`}

IV. DATA SOURCES

The raw training and testing data are collected from the literature listed below.[1–187]

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