

# **Redefining the Stability of Water Oxidation Electrocatalysts: Insights from Materials Databases and Machine Learning**

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Number of pages: 17

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## Additional Information on Machine Learning Models

The machine learning models and libraries discussed in this Perspective are available at the following [GitHub link](#). These resources are intended for community use, allowing researchers to analyze their datasets or categorize materials into specific groups depending on their needs. We advise compiling materials databases from well-controlled experimental conditions and including a comprehensive set of features to improve result accuracy. Additionally, we encourage exploring other tools and models to leverage machine learning in electrocatalysis.

**Supporting note:** Constructing decision trees using gradient boosting.

The gradient boosting technique, which involves a series of decision trees, each acting as a weak learner, builds each successive tree on the deficiencies of its predecessors to enhance the overall model. However, in our study, the XGBoost model produced only one tree. Three main factors explain this outcome:

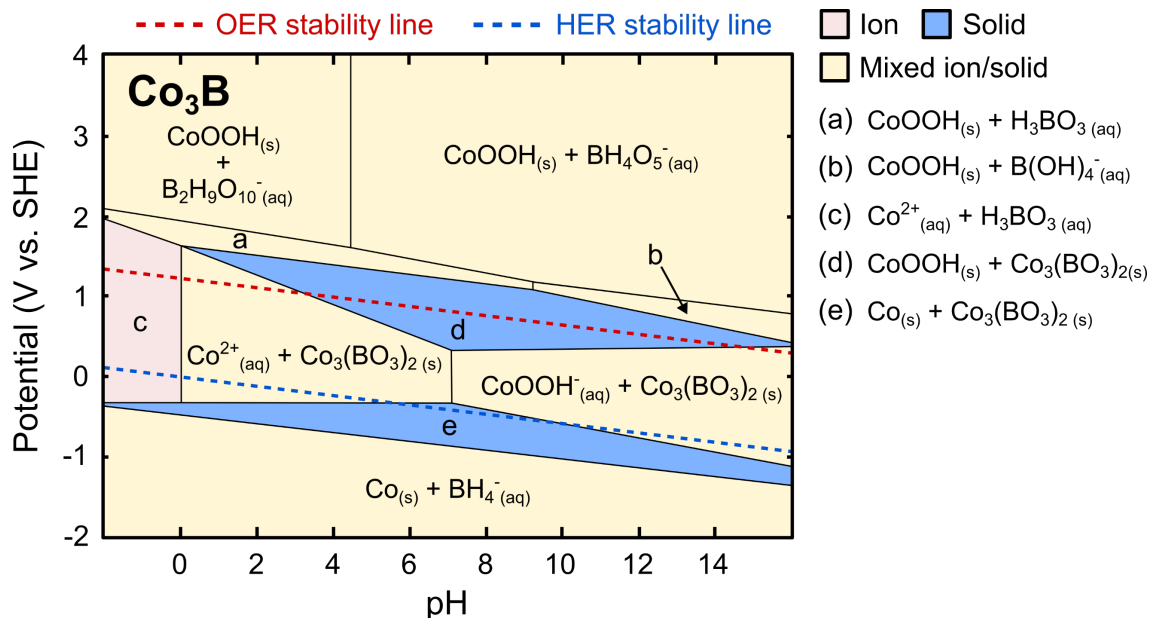
First, after extensive testing with various machine learning models, such as Linear Regression and Support Vector Machines using a Linear Kernel, we discovered that the independent and dependent variables in our dataset exhibited a simple linear relationship. In such scenarios, adding more trees would not improve model performance and might lead to overfitting.

Second, we implemented early stopping to prevent overfitting and ensure the model's effectiveness on unseen data. This technique halts the training process when the model's performance on a validation set meets a predefined threshold. In our case, this criterion was satisfied after just one tree, resulting in an XGBoost model composed of this single tree.

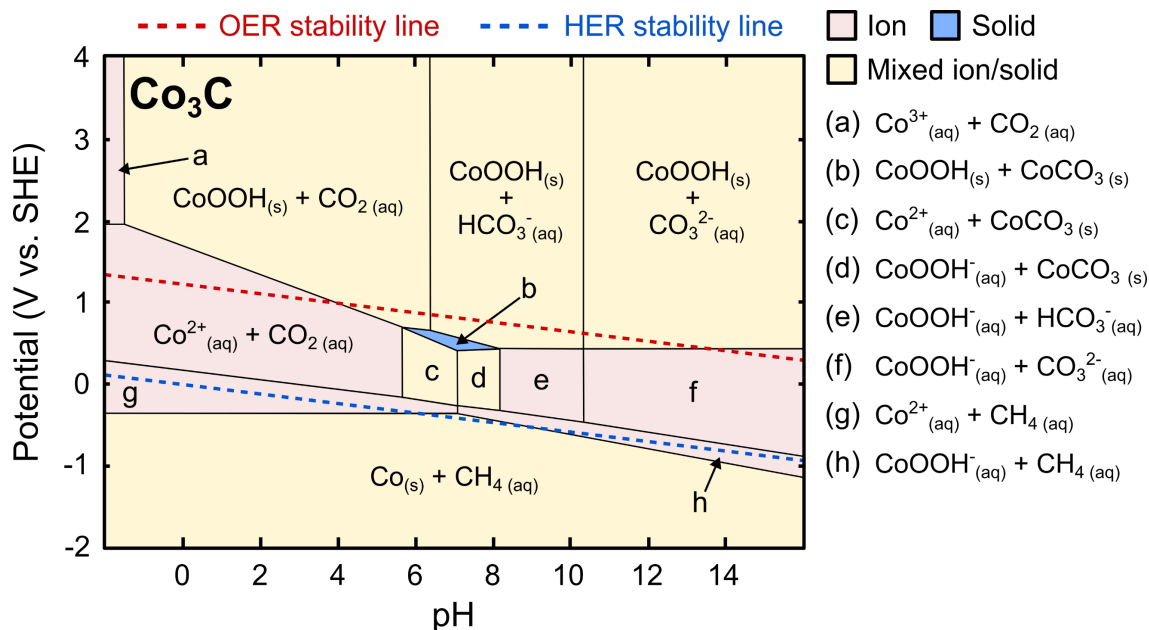
Third, we carefully screened our transition metal (TM) X-ide datasets to ensure a fair comparison under homogeneous testing conditions. We removed certain entries because they used different electrolytes (e.g., NaOH) or due to incomplete data provided in the study. Consequently, our datasets became relatively small after this screening. More complex models with multiple trees could overfit and underperform on new, unseen data. Thus, employing a single-tree model promotes simplicity and reduces the risk of overfitting, making it preferable when working with small datasets.

We utilized the *plot\_tree* function from the XGBoost library to visualize the resulting trees, shown in **Figures 5a, S13, and S14**, for TM selenides, phosphides, and sulfides, respectively.

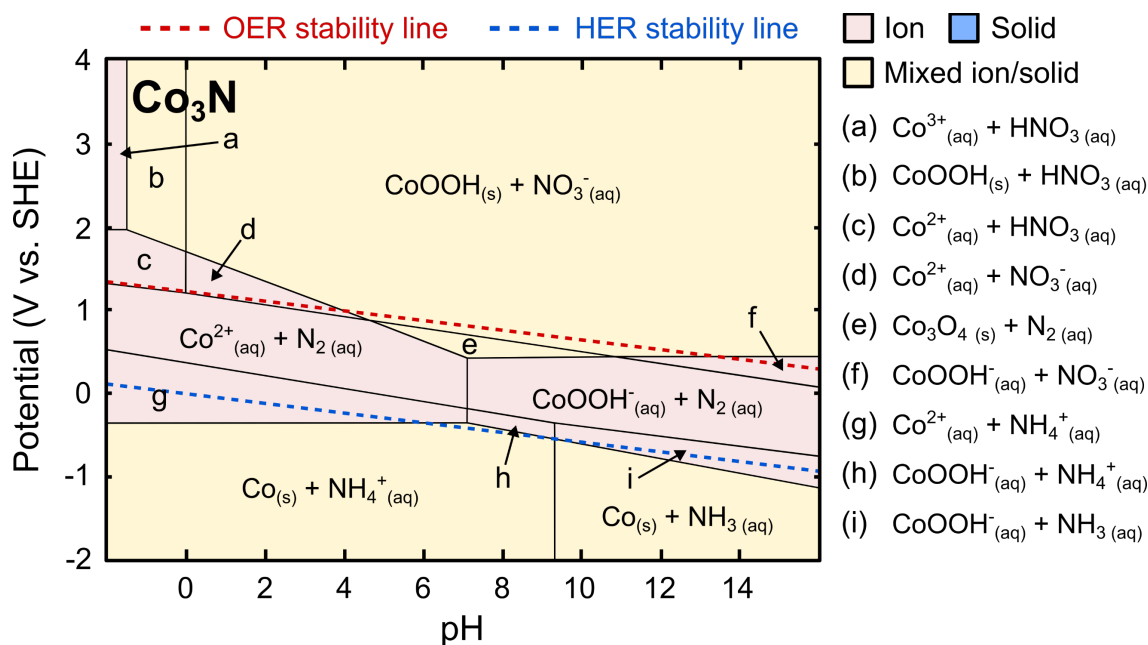
## Supporting Figures, Tables, and Schemes



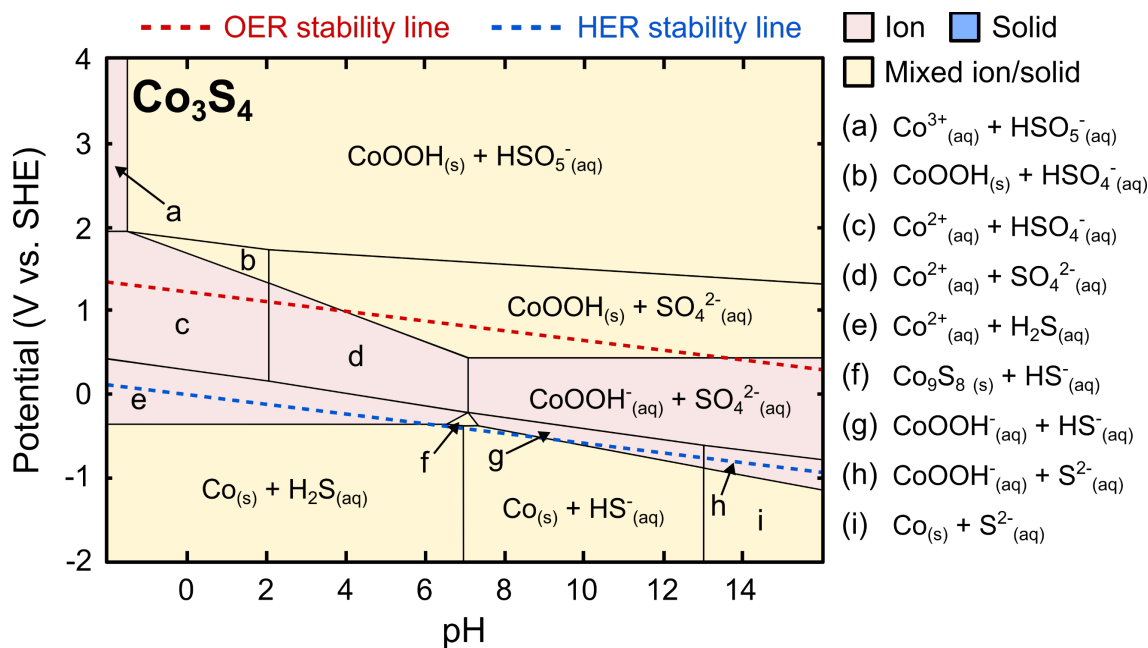
**Figure S1.** Pourbaix diagram for  $\text{Co}_3\text{B}$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



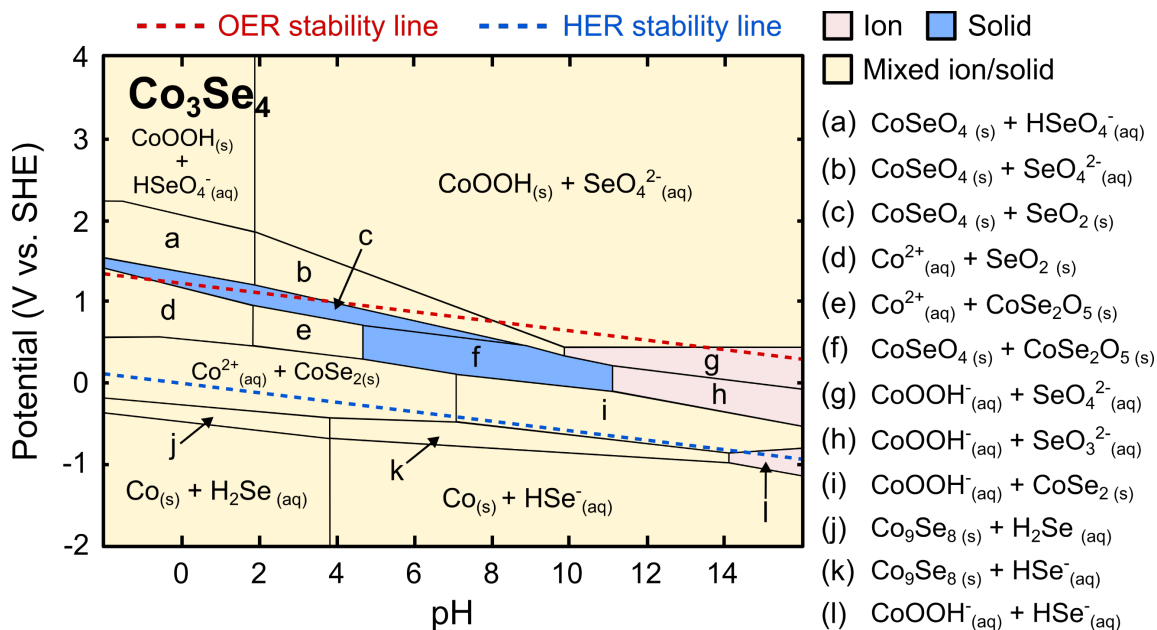
**Figure S2.** Pourbaix diagram for  $\text{Co}_3\text{C}$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



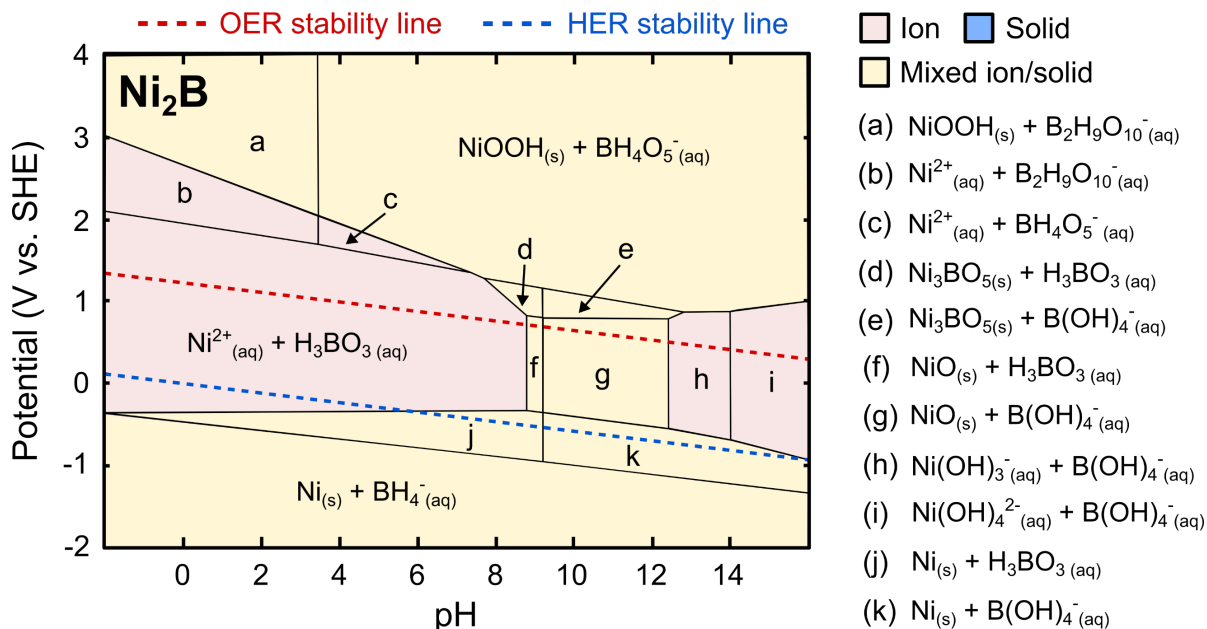
**Figure S3.** Pourbaix diagram for  $\text{Co}_3\text{N}$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



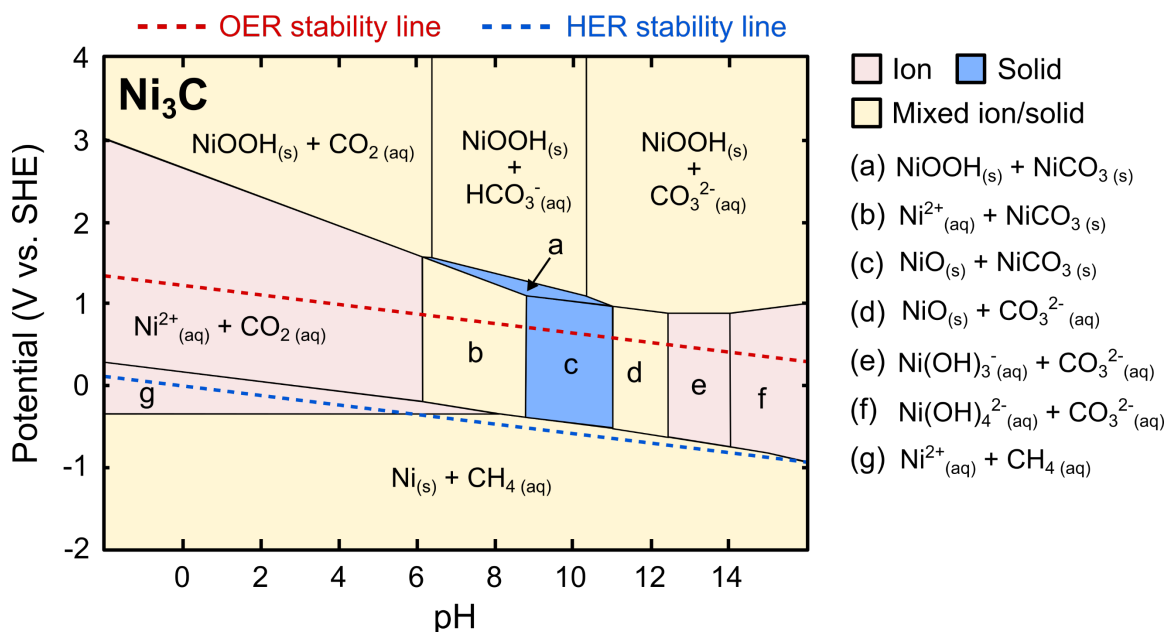
**Figure S4.** Pourbaix diagram for  $\text{Co}_3\text{S}_4$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



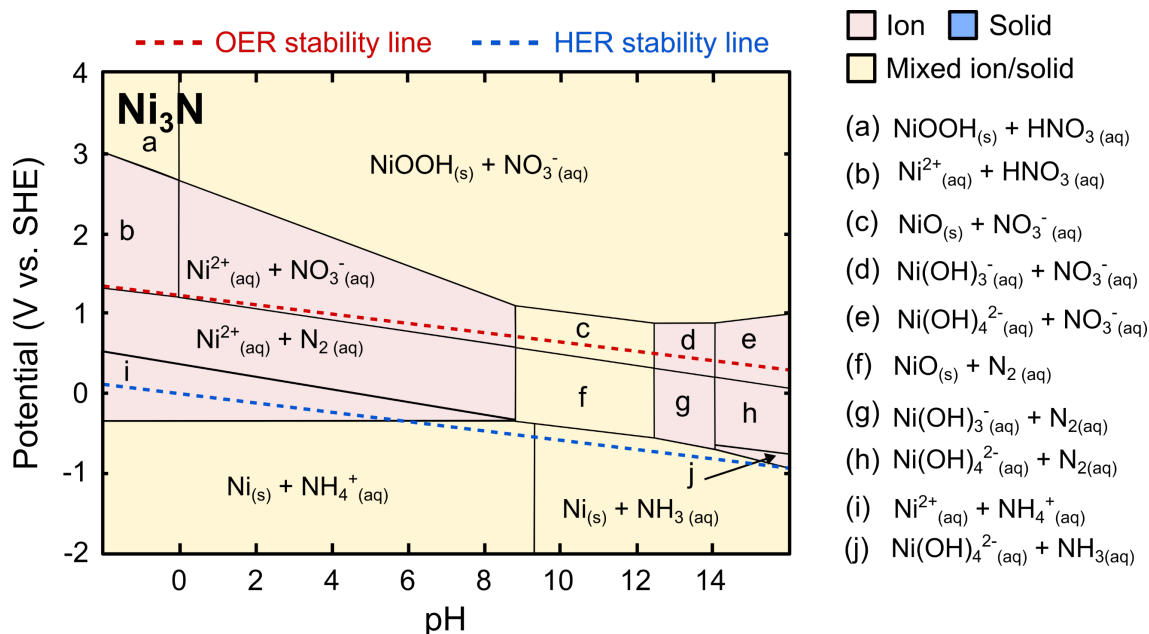
**Figure S5.** Pourbaix diagram for  $\text{Co}_3\text{Se}_4$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



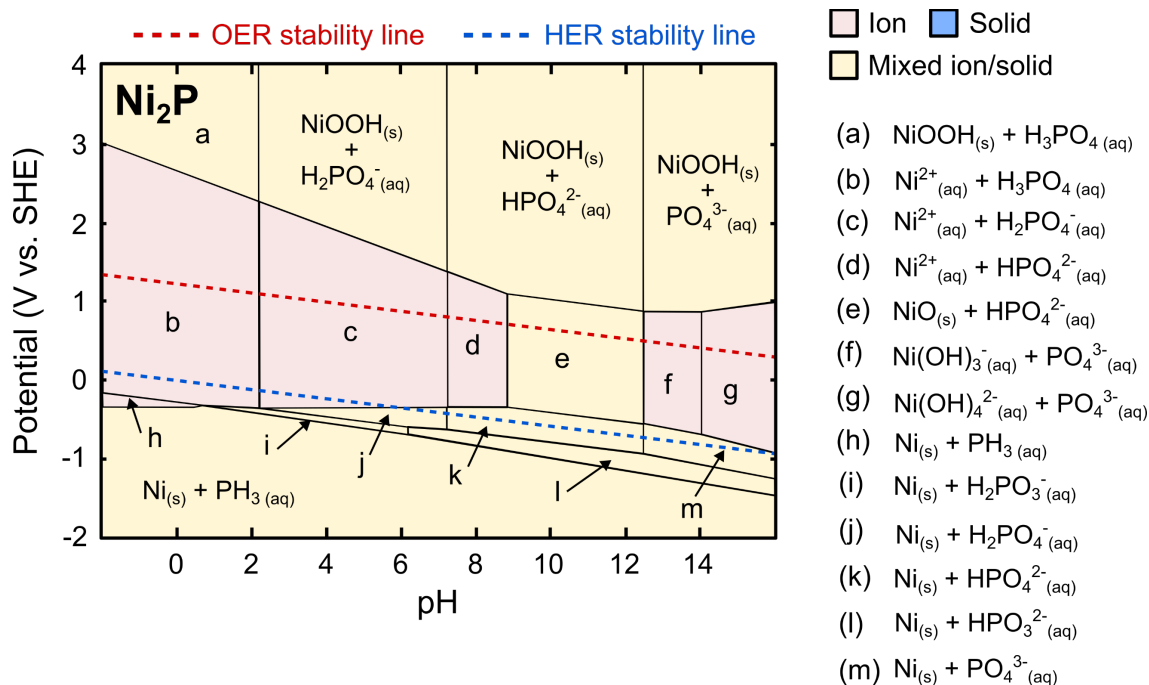
**Figure S6.** Pourbaix diagram for  $\text{Ni}_2\text{B}$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



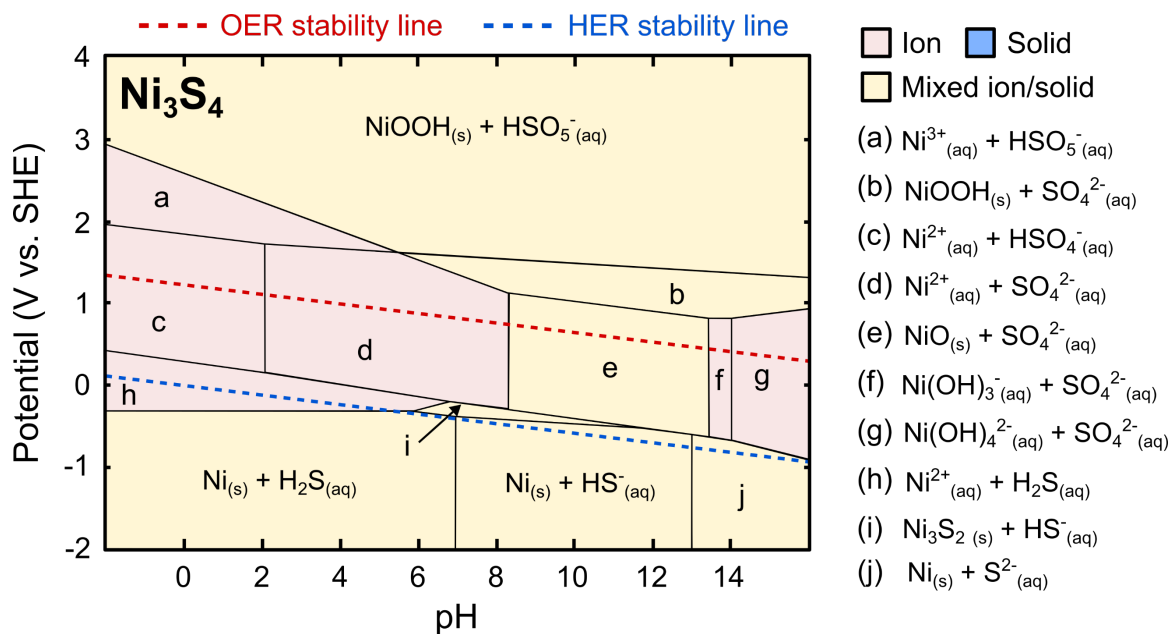
**Figure S7.** Pourbaix diagram for Ni<sub>3</sub>C. The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



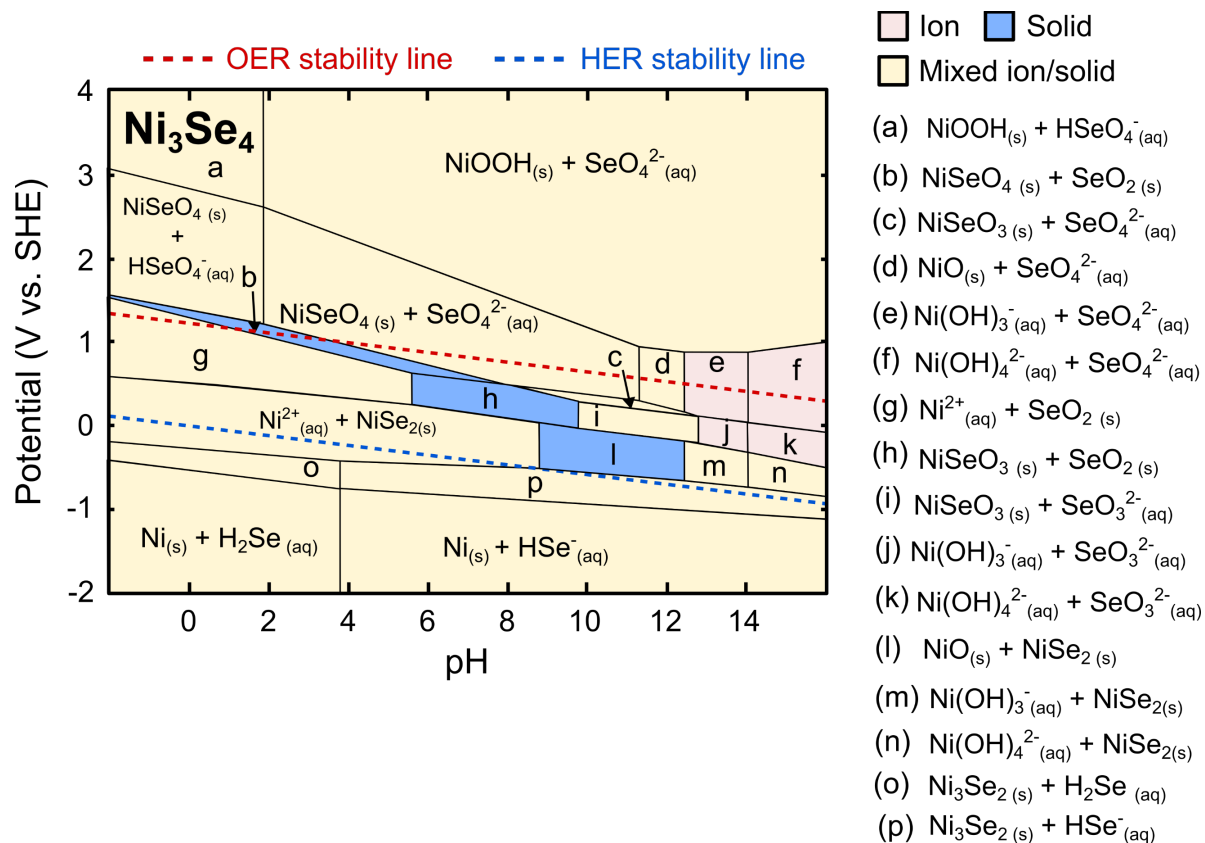
**Figure S8.** Pourbaix diagram for Ni<sub>3</sub>N. The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .



**Figure S9.** Pourbaix diagram for  $\text{Ni}_2\text{P}$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .

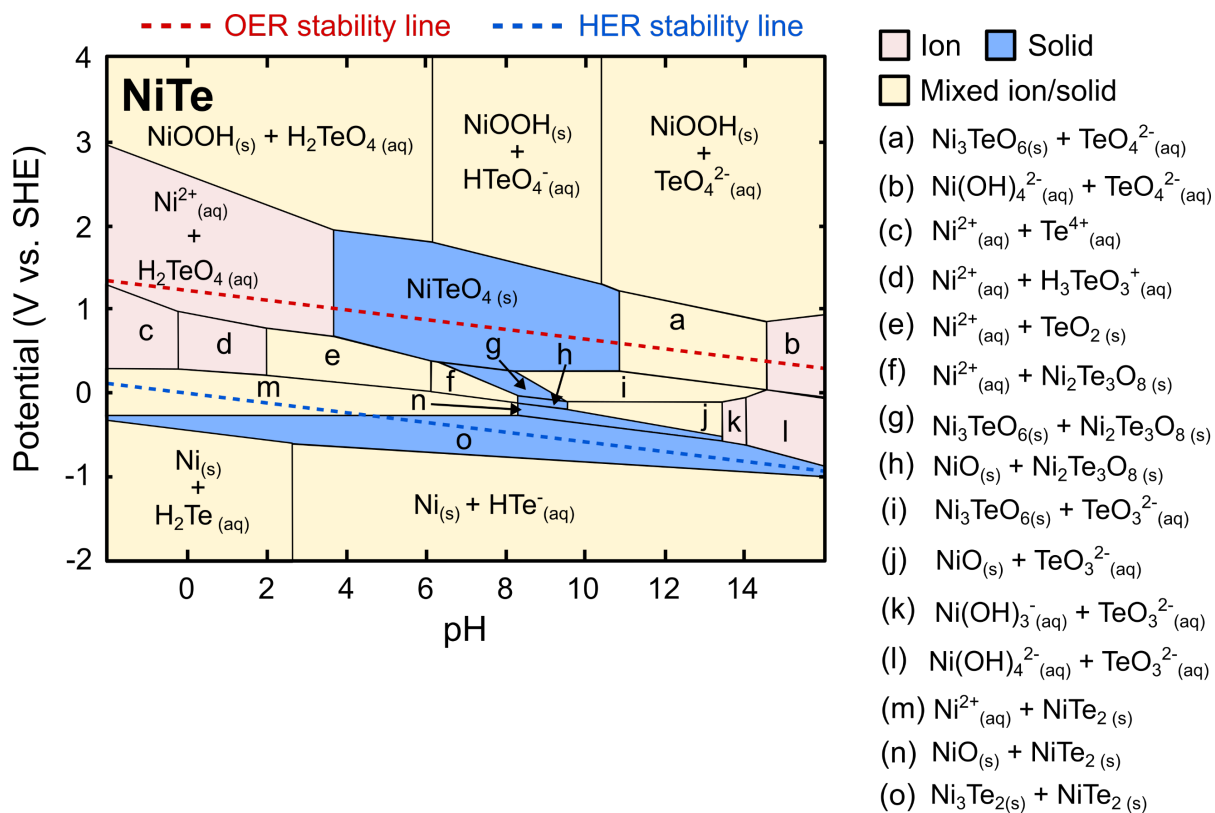


**Figure S10.** Pourbaix diagram for  $\text{Ni}_3\text{S}_4$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol} \cdot \text{L}^{-1}$ .

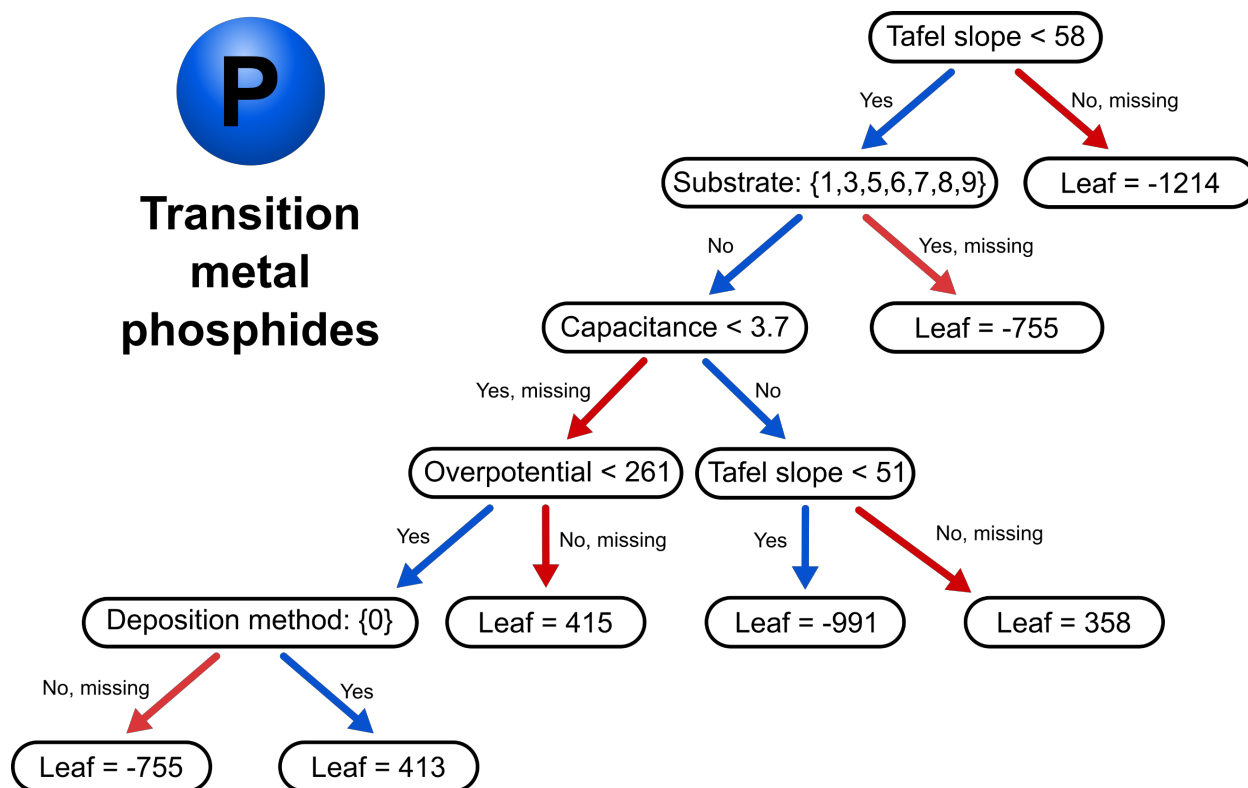


**Figure S11.** Pourbaix diagram for  $\text{Ni}_3\text{Se}_4$ . The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration:  $10^{-5} \text{ mol}\cdot\text{L}^{-1}$ .





**Figure S12.** Pourbaix diagram for NiTe. The plot was adapted from the diagram generated using the Materials Project.<sup>39–42</sup> Ion concentration: 10<sup>-5</sup> mol·L<sup>-1</sup>.



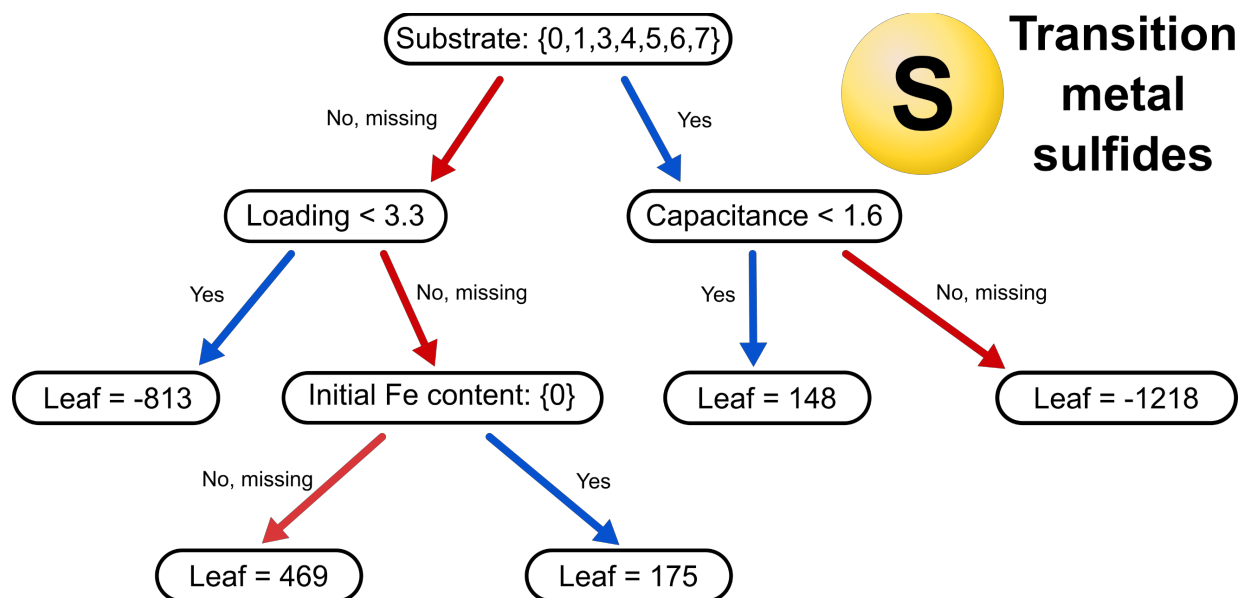
**Figure S13.** Decision tree for the TM phosphide dataset. A blue arrow denotes a catalyst node with identical features, while a red arrow indicates a precatalyst node. The leaves symbolize the purest nodes for each catalyst and precatalyst subgroup. A total of 126 data points, each representing a TM phosphide material from our compilation, were analyzed.

**Supporting note:** The following codes apply for the decision trees and force plots of the TM phosphide, sulfide, and selenide databases:

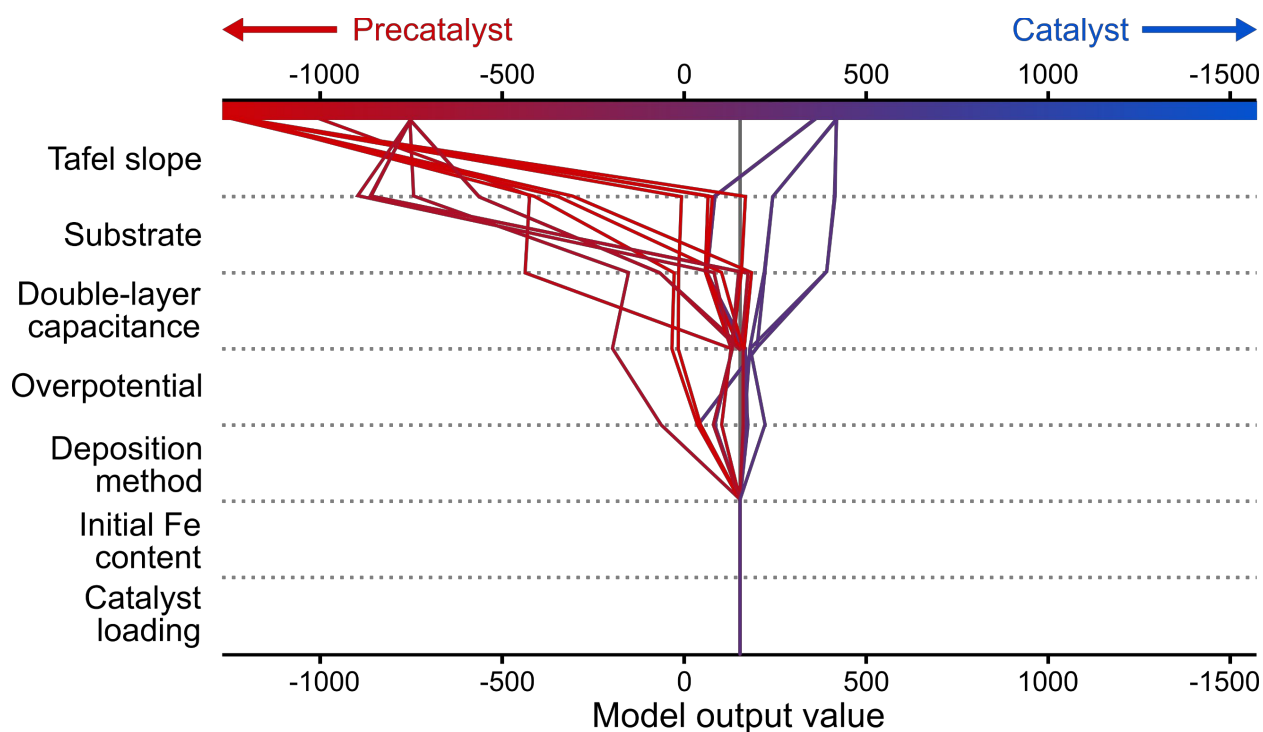
*Substrate type:* "Glassy carbon": 0, "carbon cloth": 1, "graphite": 2, "Au glass": 3, "carbon foam": 4, "Cu foil": 5, "FeNi foam": 6, "Ni foam": 7, "carbon fiber paper": 8, "Pt sheet": 9, "carbon paper": 10, "NiFe alloy": 11, "Fe foam": 12, "Ti mesh": 13, "Cu mesh": 14.

*Deposition method:* "Self-supported electrode": 1, "powder-coated electrode": 2.

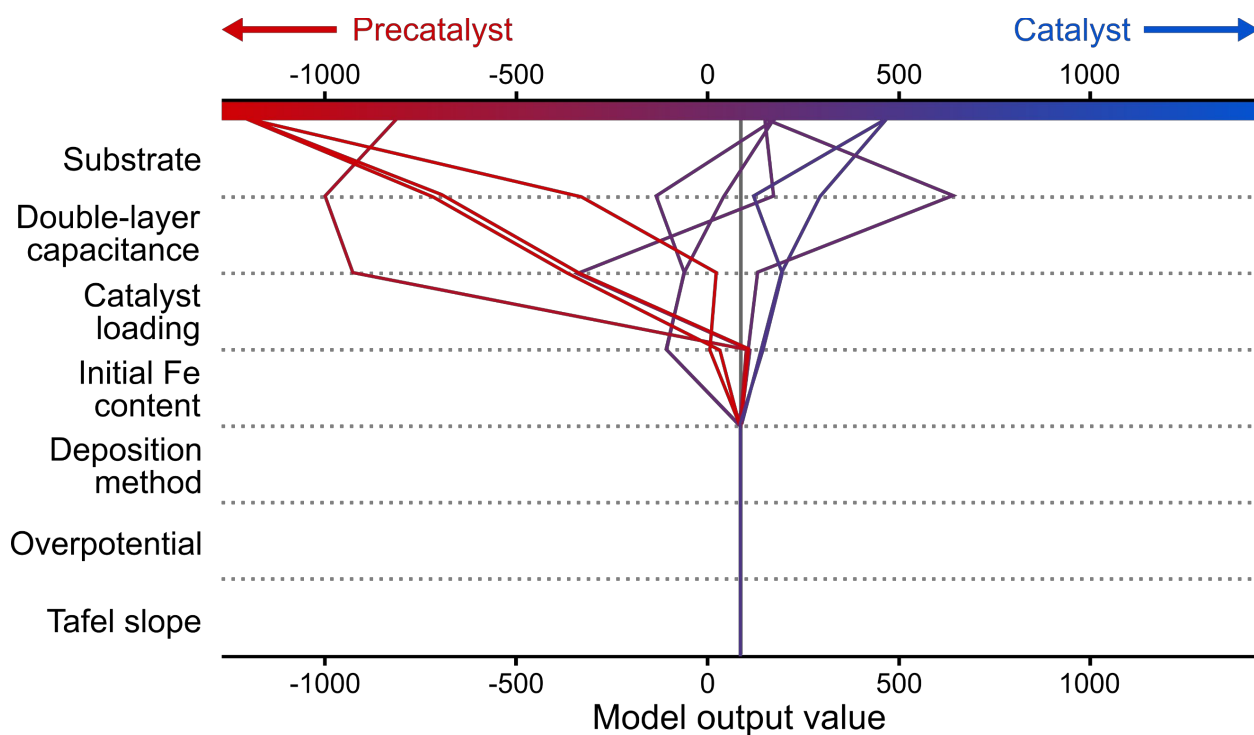
*Initial elemental content:* "Element present": 0, "no element present": 1.



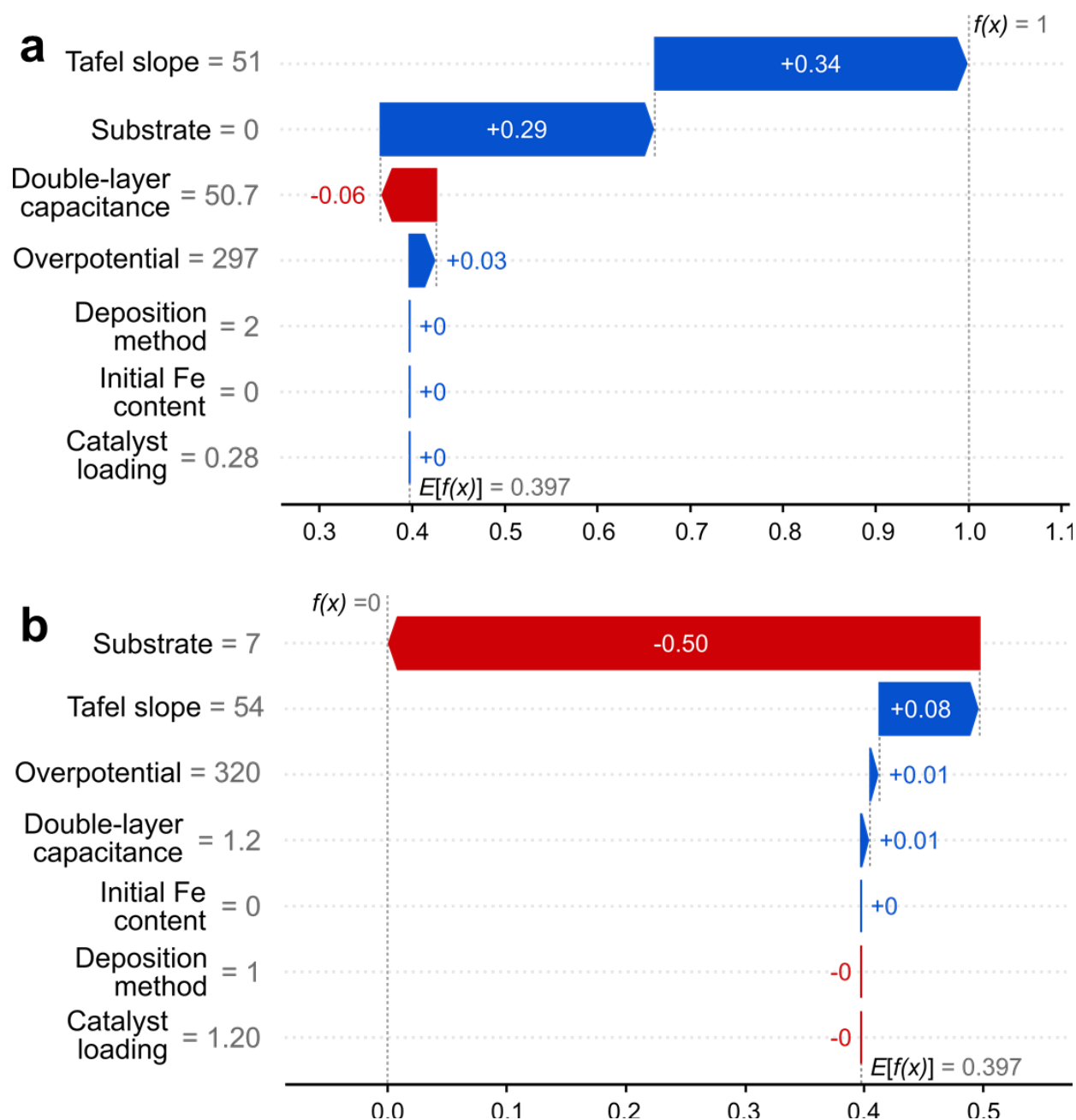
**Figure S14.** Decision tree for the TM sulfide dataset. A blue arrow denotes a catalyst node with identical features, while a red arrow indicates a precatalyst node. The leaves symbolize the purest nodes for each catalyst and precatalyst subgroup. A total of 104 data points, each representing a TM phosphide material from our compilation, were analyzed.



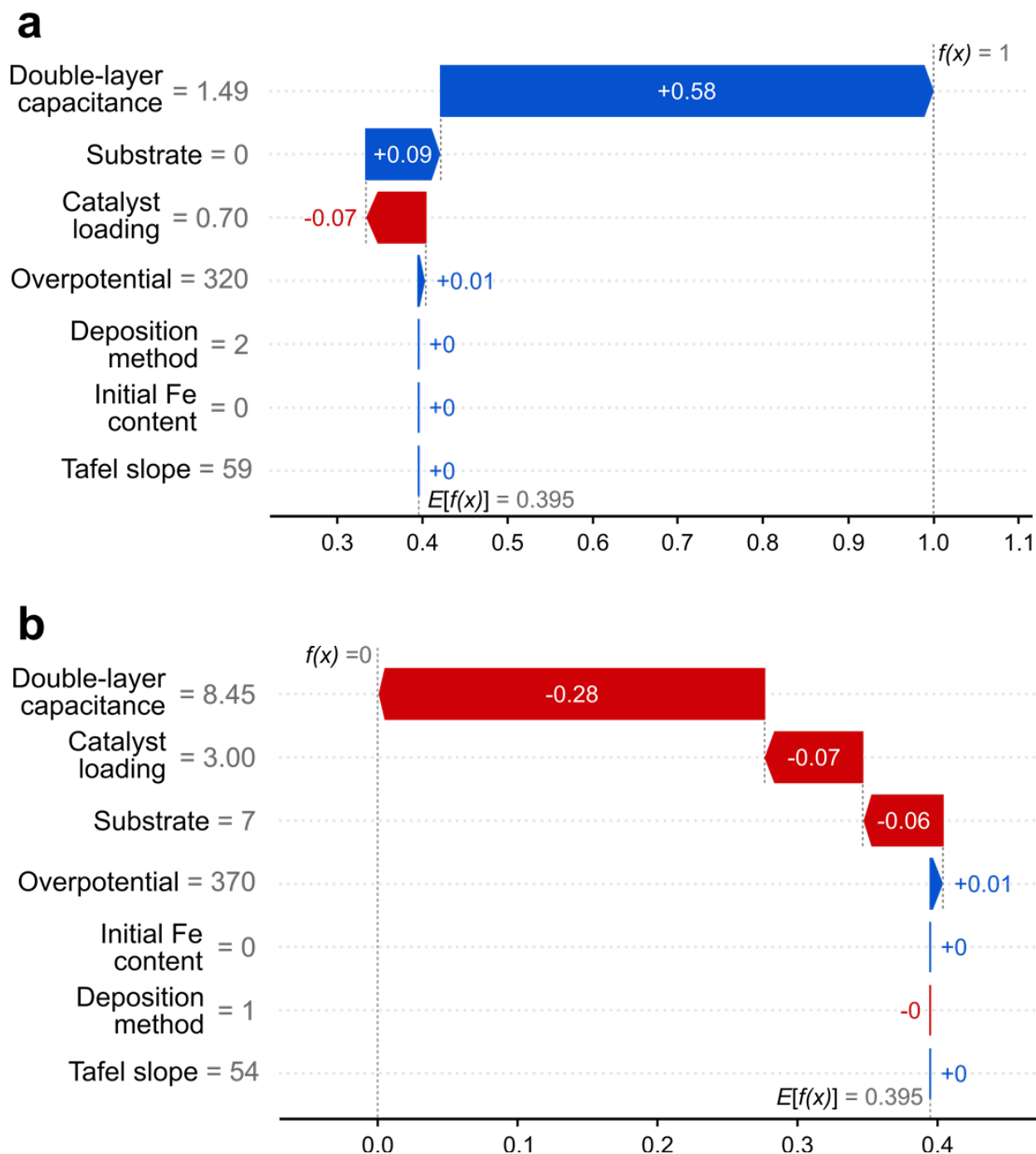
**Figure S15.** Decision plot for the TM phosphide dataset. Each colored line represents a single TM phosphide electrocatalyst from our compilation of the literature. Blue lines represent a catalyst classification, whereas red lines denote a precatalyst. The features are ranked in descending order based on their importance to the model, with the most influential feature at the top. A total of 126 data points, each representing a TM phosphide material from our compilation, were analyzed.



**Figure S16.** Decision plot for the TM sulfide dataset. Each colored line represents a single TM phosphide electrocatalyst from our compilation of the literature. Blue lines represent a catalyst classification, whereas red lines denote a precatalyst. The features are ranked in descending order based on their importance to the model, with the most influential feature at the top. A total of 126 data points, each representing a TM phosphide material from our compilation, were analyzed.



**Figure S17.** Force plots for a single (a) catalyst and (b) precatalyst from the TM phosphide dataset. The bars represent the contribution of specific features to the classification as either a catalyst (blue) or a precatalyst (red). These features are ranked in descending order of importance to the model, with the most influential feature at the top.



**Figure S18.** Force plots for a single (a) catalyst and (b) precatalyst from the TM sulfide dataset. The bars represent the contribution of a specific feature towards the catalyst (blue) and precatalyst (red) classification. The features are ranked in descending order based on their importance to the model, with the most influential feature at the top.

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