

Design of catalysts for electrochemical nitric oxide reduction to ammonia based on stacked ensemble learning

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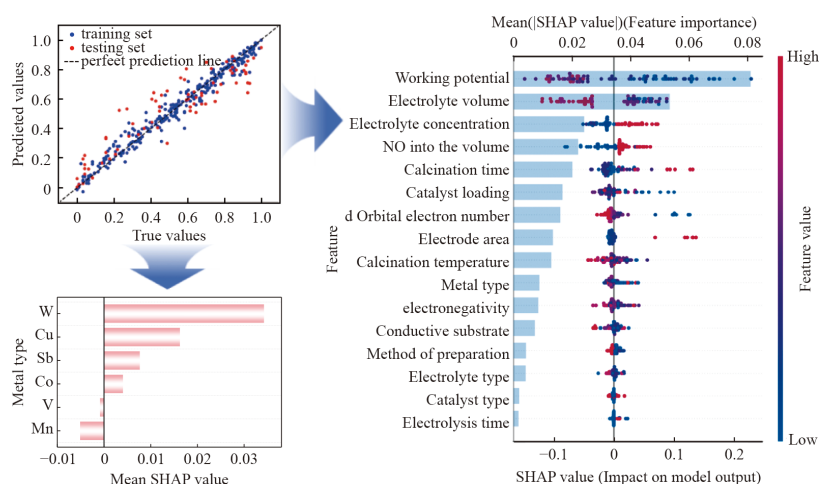
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Abstract: The electrocatalytic reduction of nitric oxide for ammonia synthesis (NORR) is a key green energy conversion technology. Its efficiency relies on high-performance electrocatalysts to enhance both ammonia yield (Y_{NH_3}) and Faradaic efficiency (F_{NH_3}). However, conventional experimental methods for screening high-activity NORR catalysts often entail high resource consumption and time costs. Machine learning combined with SHAP feature analysis was employed to establish a stacked

ensemble model that integrates multiple algorithms, to allow for a systematic investigation of the key descriptors governing NORR performance based on an experimental dataset. Evaluation of eight model algorithms revealed that the Stacked-SVR model achieved an R^2 of 0.9223 and an RMSE of 0.0608 for predicting Y_{NH_3} on the test set, whereas the Stacked-RF model achieved an R^2 of 0.9042 and an RMSE of 0.0900 for predicting F_{NH_3} . The stacked ensemble model integrates the strengths of individual algorithms and demonstrates strong NORR prediction performance while avoiding overfitting. SHAP feature analysis results revealed that the Cu content in the catalyst composition has the most significant impact on catalytic performance. Moreover, the combination of the wet chemical reduction synthesis, a carbon fiber (CF) conductive substrate, and HCl electrolyte is more favorable for enhancing catalytic activity. Additionally, moderately lowering the working potential, controlling the electrolyte volume at low to medium levels, reducing catalyst loading, and increasing electrolyte concentration were found to synergistically enhance both Y_{NH_3} and F_{NH_3} .

Key words: NORR; machine learning; stacked model; ammonia yield; ammonia Faraday efficiency



The electrocatalytic reduction of nitric oxide to ammonia (NORR) represents a technology that supports green and low-carbon development^[1]. Driven by electricity, this process facilitates the conversion of nitric oxide (NO) into high-value ammonia (NH₃) through multi-step proton-coupled electron transfer at catalytic active sites. NORR not only efficiently converts NO, an atmospheric pollutant, into

valuable NH₃, but also reduces reliance on fossil fuels inherent to conventional ammonia synthesis^[2–3]. This process not only efficiently converts nitrate pollutants but also synthesizes clean energy carriers, offering an innovative approach to transforming China's energy structure and promoting nitrogen resource recycling within the framework of the “Dual Carbon” strategy.

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In practical applications, it is often challenging to achieve both high ammonia yield (Y_{NH_3}) and high Faradaic efficiency (F_{NH_3}) simultaneously in the NORR process. Wu et al.^[4] designed a heterogeneous-structured CoB/Co@C catalyst, which achieved an exceptionally high Y_{NH_3} of 315.4 $\mu\text{mol}/(\text{cm}^2 \cdot \text{h})$, while its F_{NH_3} was 73%. In contrast, Liu et al.^[5] developed a bismuth nanoparticle/carbon nanosheet composite (Bi@C) that exhibited a remarkably high F_{NH_3} of 93% at a potential of -0.4 V vs. RHE, demonstrating excellent NH_3 selectivity. However, its Y_{NH_3} was only 27.4 $\mu\text{mol}/(\text{cm}^2 \cdot \text{h})$. Evidently, achieving both high Y_{NH_3} and high F_{NH_3} remains a considerable challenge. Various strategies have been explored to address this issue, such as modulating metal types, optimizing synthesis methods, and modifying conductive substrates^[6–7]. Nevertheless, due to the complex interplay of multiple factors affecting catalyst performance and the vast parameter space, relying solely on experimental screening is both time-consuming and resource-intensive, making it difficult to efficiently identify catalysts with high activity and selectivity.

In recent years, machine learning has rapidly advanced across various fields, becoming an essential tool for addressing complex nonlinear relationships^[8]. In catalyst design, machine learning can quickly identify active sites, optimize reaction conditions, and adjust catalyst components, significantly enhancing research efficiency and reducing costs^[9–10]. Dong et al.^[11] constructed an NH_3 -SCR catalyst database and applied an extremely randomized trees regression (ETR) model to identify the optimal feature combinations, successfully screening 11 potential elemental compositions for highly active low-temperature SCR catalysts. Zhou et al.^[12] established a HER catalyst database using publicly available data and trained three machine learning models to analyze the impact of various input features on overpotential. Through interpretability analysis, they identified the optimal catalyst as a metal combination of Pt and Mo at an optimal Pt-to-Mo ratio of 0.5. These studies demonstrate that machine learning methods integrated with experimental data can effectively address the complexity of catalytic systems, offering a novel approach for the efficient catalyst screening and optimization^[13].

In this study, three individual models, including Random Forest Regression (RF), Extreme Gradient Boosting (XGBoost), and Support Vector Regression (SVR), along with three stacked ensemble models, including Stacked-RF (using XGBoost and SVR as base learners and RF as the meta-learner), Stacked-XGBoost (using RF and SVR as base learners and XGBoost as the meta-learner), and Stacked-SVR (using RF and XGBoost as base learners and SVR as the meta-learner) were employed to predict the Y_{NH_3} and F_{NH_3} of various nitrate reduction reaction (NORR) catalysts, aiming to screen catalyst materials that exhibit both high activity and high selectivity. The evaluation results show that the Stacked-SVR model performs best in

predicting ammonia yield, while Stacked-RF achieves the highest accuracy in predicting Faradaic efficiency. Through SHAP global interpretability analysis, the influence of parameters such as working potential and electrolyte volume on catalytic performance was revealed. Local interpretability analysis further clarified the effects of metal type, synthesis method, conductive substrate, and electrolyte type on catalyst performance. This work provides a data-driven approach for the efficient design and optimization of high-performance NORR catalysts.

1 Machine learning computational methods

1.1 Database construction

In this study, a comprehensive database of metal nitric oxide reduction reaction (NORR) catalysts was established by retrieving and integrating 422 data points from publications since 2020, sourced from the Web of Science. For the selection of input features, four types of descriptors were established based on the influence of fundamental physicochemical properties of catalysts on reaction performance^[14–15]. These include composition descriptors (metal type, catalyst type), synthesis descriptors (preparation method, calcination temperature, calcination time), reaction descriptors (working potential, electrolyte volume, electrolyte concentration, etc.), and electronic structure descriptors (electronegativity, d -band electron count). The output features are Y_{NH_3} and F_{NH_3} . The raw data and the proportions of each metal type are detailed in Table S1 and Figure S1 in the Supplementary Materials. It can be observed that commonly used metals for NORR catalysis each account for approximately 17% of the total entries in this database.

1.2 Machine learning models

A total of eight machine learning algorithms were employed in this study to predict Y_{NH_3} and F_{NH_3} . The algorithms included three independent models, namely RF^[16], XGBoost^[17] and SVR^[18]. In addition, three stacked ensemble models were applied, including Stacked-RF, Stacked-XGBoost, and Stacked-SVR. Model stacking improves generalization performance by combining predictions of multiple base learners through a meta-learner^[19–20]. To evaluate the prediction performance of the stacked ensemble models, the Convolutional Neural Network (CNN) and Artificial Neural Network (ANN) models were also employed to predict Y_{NH_3} and F_{NH_3} . Five-fold cross-validation was applied in this study to produce out-of-fold predictions from the base learners, and three heterogeneous independent algorithms, namely RF, XGBoost, and SVR, were selected and combined in pairs to build the base learners. Differences in the underlying principles of these algorithms lead to complementary predictions, thereby enhancing the diversity and robustness

of the ensemble models. The predictions generated by the base learners serve as the primary outputs, which are subsequently trained and integrated by the meta-learner to produce the final predictions, as illustrated in Figure S2. The stacking ensemble leverages the complementarity of heterogeneous base learners to reduce the structural bias intrinsic to single models. Furthermore, the integration of diverse learning patterns effectively suppresses overfitting and enhances the model's ability to capture complex relationships within the feature space, yielding more robust and reliable predictive performance.

1.3 Machine learning methods

All algorithms were implemented using Scikit-learn in a Python 3 environment. The dataset was randomly partitioned into training and test sets with 80% allocated for training and the remaining 20% for testing^[21]. The training set was primarily used for model training and hyperparameter optimization, while the test set was reserved for evaluating the actual predictive performance of the algorithms. During the model training phase, a combination of grid search and five-fold cross-validation was employed for model optimization^[22–23]. The training set was randomly divided into five mutually exclusive subsets of equal size. In each iteration of the five-fold cross-validation, one subset was designated as the validation set, and the remaining four subsets were employed for model training and grid search. The hyperparameter combination that achieved the highest cross-validated R^2 score was selected as the optimal set. For the stacked ensemble models, the base learners directly adopted the optimized hyperparameter configurations from the independent models, while the meta-learner was optimized using the same method as the independent models. The aforementioned 20% test set was strictly isolated and was not involved in any model training or parameter selection processes. It was used exclusively for the final model evaluation. The coefficient of determination

(R^2) and root mean square error (RMSE) were adopted as metrics to assess the predictive accuracy of the models^[24], as shown in Equations (1) and (2). Additionally, the SHAP (SHapley Additive Explanations) explainer^[25] was introduced to perform interpretability analysis on the model predictions.

$$R^2 = 1 - \frac{\sum_{i=1}^N (\hat{y}_i - y_i)^2}{\sum_{i=1}^N (y_i - \bar{y})^2} \quad (1)$$

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (y_i - \hat{y}_i)^2} \quad (2)$$

where y_i represents the actual value of the i -th sample, $\hat{y}_i$ denotes its predicted value, $\bar{y}$ is the mean of the actual values of all samples, and N represents the total number of samples.

2 Results and discussion

2.1 Data preprocessing

The machine learning workflow of this study is illustrated in Figure 1, which primarily comprises six components. During data preprocessing, Pearson correlation analysis was employed to assess linear relationships between different features^[26]. The Pearson coefficient takes values in the range of -1 to 1 , with coefficients approaching 1 or -1 representing strong positive or negative linear correlations, respectively, and values near 0 indicating weak or no linear correlation. Figures 2(a) and 2(b) present the Pearson correlation coefficient heatmaps for Y_{NH_3} and F_{NH_3} , respectively. The results demonstrate that the correlation coefficient between any two selected features remains below the threshold of 0.85 , confirming the absence of significant multicollinearity among the chosen features^[27]. This effectively prevents feature redundancy from interfering with model training.

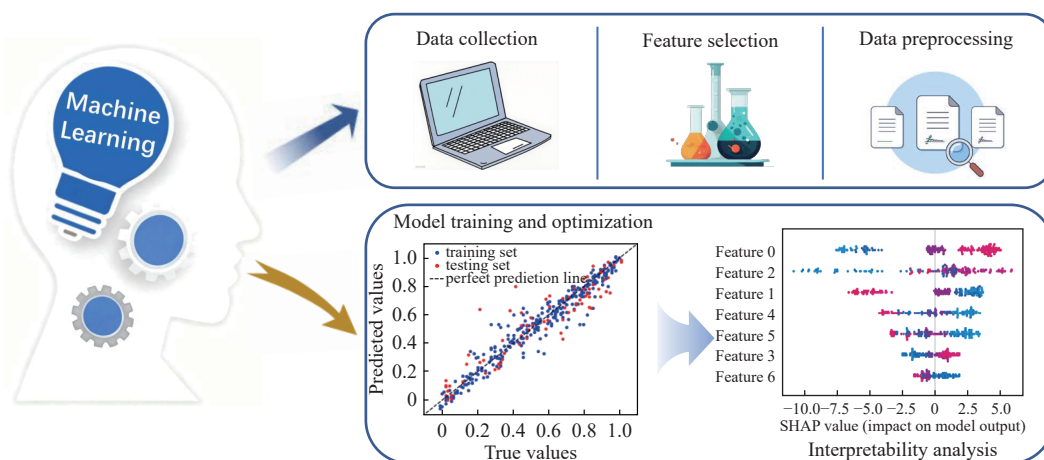


Fig. 1 Machine learning workflow diagram

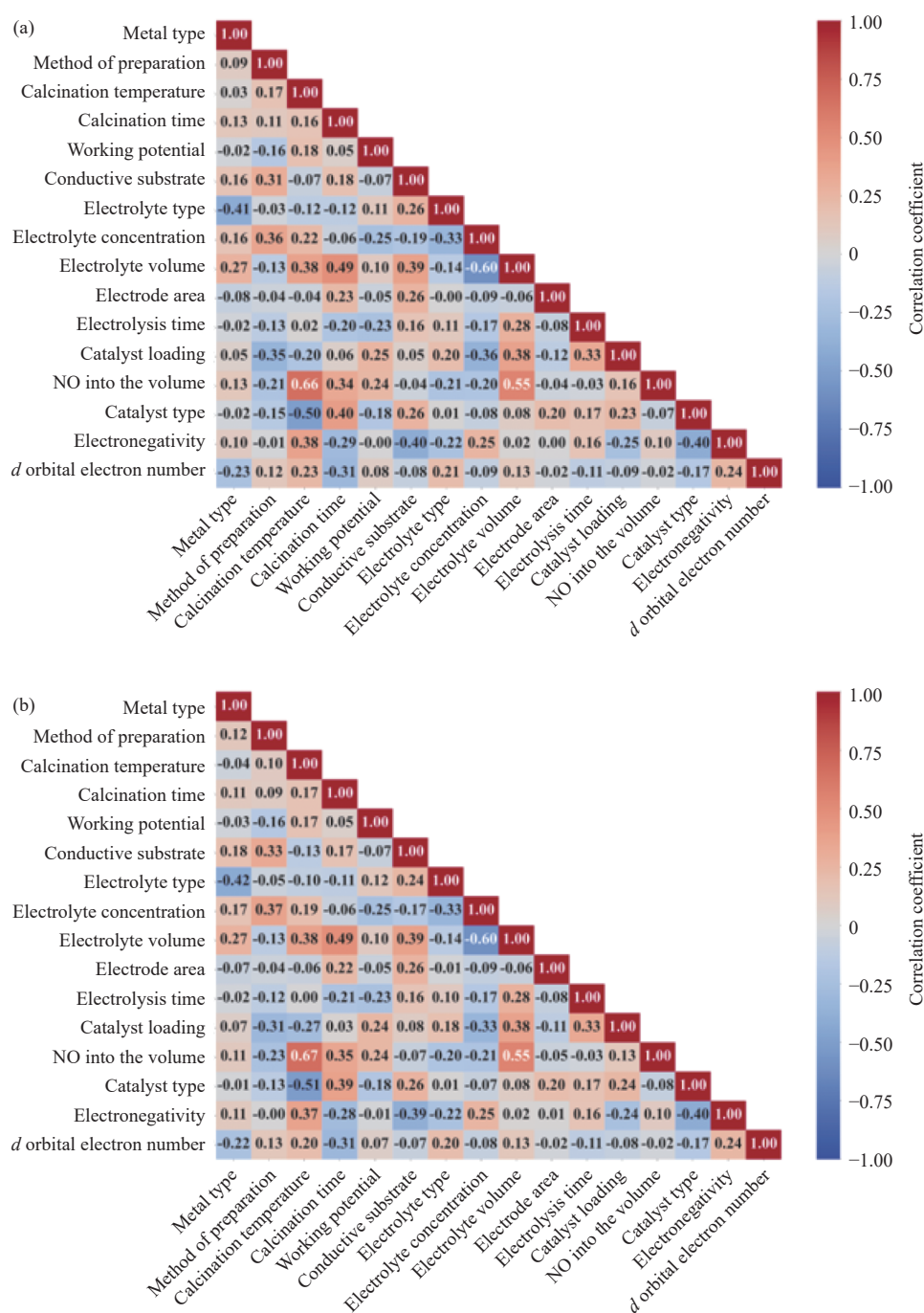


Fig. 2 (a) Heatmap of Pearson correlation coefficient matrix for Y_{NH_3} ; (b) Heatmap of Pearson correlation coefficient matrix for F_{NH_3}

Given the substantial variation in the numerical ranges of experimental parameters, all features were normalized to the [0,1] range to improve model convergence speed and learning accuracy^[28]. The distribution of the normalized feature parameters is shown in Figure 3. Ensuring data completeness is essential for robust machine learning training. Accordingly, a differentiated imputation strategy was adopted to address missing feature values within the database and to preserve data integrity for subsequent modeling. For features with a small proportion of missing values, mean imputation was used to preserve the data distribution. For key features with a higher proportion of

missing values, such as calcination temperature and calcination time, a machine learning approach based on the XGBoost model was employed to impute the missing data.

This imputation strategy allows for more precise estimation of missing values, effectively preserving the intrinsic information within the dataset and consequently improving the efficiency and accuracy of model training^[29]. A total of 14 feature parameters, including metal type, catalyst type, preparation method, and working potential, were selected to construct the input features for the calcination temperature prediction model. For the calcination time prediction model, 15 input features,

including the previous 14 features and the calcination temperature, were used. The detailed feature parameters are listed in Table S2. The results indicated that the calcination temperature model achieved R^2 values greater than 0.8400 and RMSE values under 0.1030 for both the training and test sets. The calcination time model, with R^2 values exceeding 0.9980 and RMSE values under 0.0110, demonstrated excellent fitting and generalization performance, allowing for precise imputation of missing calcination temperature and time values. To evaluate the impact of data imputation on model performance, the models were also trained using the original dataset without imputation. The results of this comparison are displayed in Figures S3–S6. The comparative analysis demonstrated that data imputation using XGBoost led to a significant performance improvement over models trained without imputation. Specifically, the R^2 for Y_{NH_3} prediction increased from 0.8270 to 0.9223, and the R^2 for F_{NH_3} prediction increased from 0.8174 to 0.9042, highlighting the superior generalization ability of the imputed model. To further validate the reliability of the imputed data for subsequent analysis, Pearson correlation analysis was performed on the fully imputed dataset, which is presented in Figure S7. The analysis revealed that no substantial multicollinearity issues were introduced among the feature variables after the imputation process, ensuring the reliability of the dataset for further analysis.

2.2 Model training

For the development of high-precision machine learning models, three classical heterogeneous standalone models (RF, XGBoost, and SVR), along with a Convolutional Neural Network (CNN) and an Artificial Neural Network (ANN), were first chosen to predict NORR performance. On this basis, three stacking models (Stacked-RF, Stacked-XGBoost and Stacked-SVR) were also constructed and used to predict NORR performance. Different standalone algorithms have distinct advantages in capturing data characteristics and relationships. Integrating diverse standalone models allows for the compensation of their individual limitations, thereby enhancing the overall predictive accuracy and robustness of the ensemble model. To ensure an objective evaluation of model performance, the coefficient of determination (R^2) and root mean square error (RMSE) were adopted as evaluation metrics for systematic comparison^[30]. The model optimization criterion aims to maximize the R^2 and minimize RMSE on the prediction dataset, offering a quantitative measure of the model's explanatory power and prediction error.

Figure 4 presents the prediction results of Y_{NH_3} by different machine learning models, where the horizontal axis represents the true values and the vertical axis represents the predicted values.

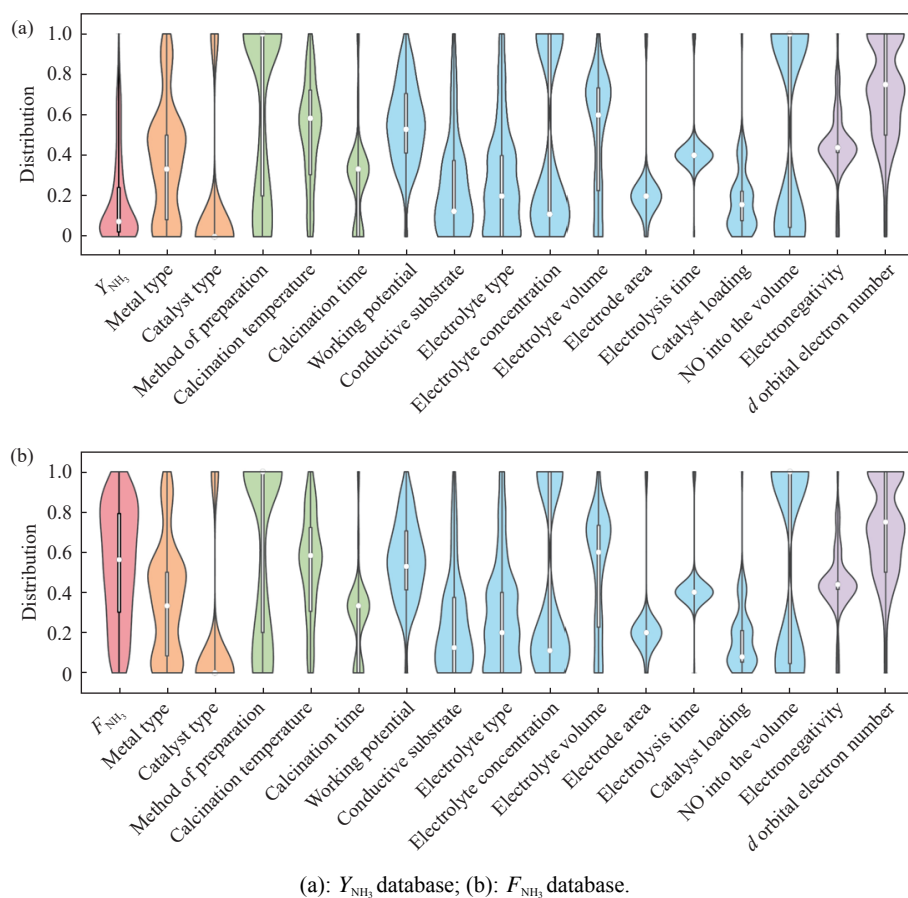
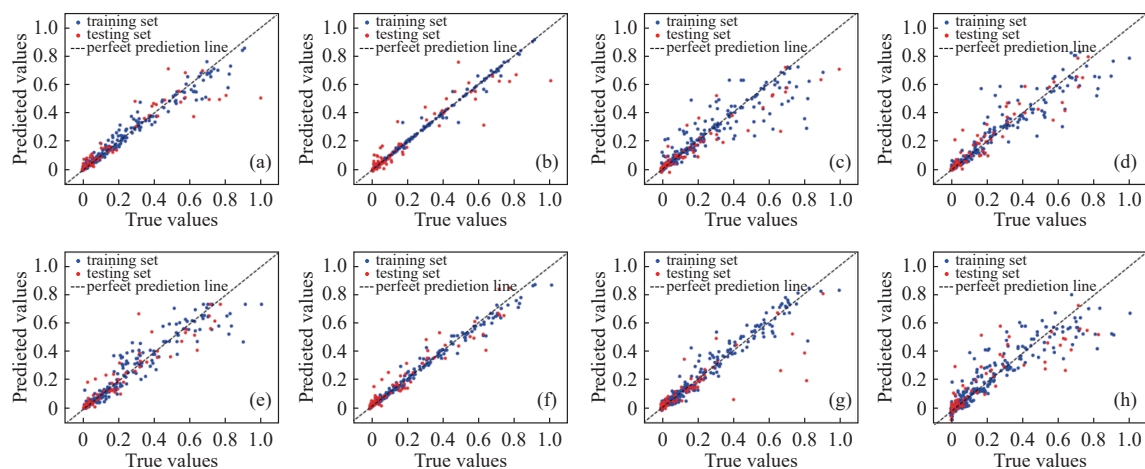


Fig. 3 Distribution of feature parameters after normalization



(a): RF; (b): XGBoost; (c): SVR; (d): Stacked-RF; (e): Stacked-XGBoost; (f): Stacked-SVR; (g): CNN; (h): ANN.

Fig. 4 Validation results of different machine learning models for predicting Y_{NH_3}

The closer the data points are to the ideal prediction line, the higher the model's accuracy and reliability. The results indicate deviations between the predicted and true values across all models, yet the stacking models consistently demonstrate superior performance compared to the standalone models. Among the stacking models, Stacked-SVR shows the best prediction performance for Y_{NH_3} .

Figure 5 displays the RMSE and R^2 values of different machine learning algorithms for predicting Y_{NH_3} on both the training and test sets. Lower RMSE values and R^2 values closer to 1 indicate better predictive performance of the models^[31]. Similar performance on both the training and test sets indicates that the model has good robustness and

generalization ability, whereas a large discrepancy between them could signal overfitting^[32]. For Y_{NH_3} prediction, both CNN and ANN models exhibit higher R^2 values on the training set compared to the test set, which indicates the presence of overfitting. The R^2 values for SVR are relatively low on both the training set (0.8586) and the test set (0.8422), with RMSE values being relatively high (0.0807 for the training set and 0.0913 for the test set), indicating poor predictive performance for Y_{NH_3} . For the RF and XGBoost algorithms, the training set R^2 values are 0.9701 and 0.9928, with RMSE values of 0.0369 and 0.0181, indicating excellent predictive capability.

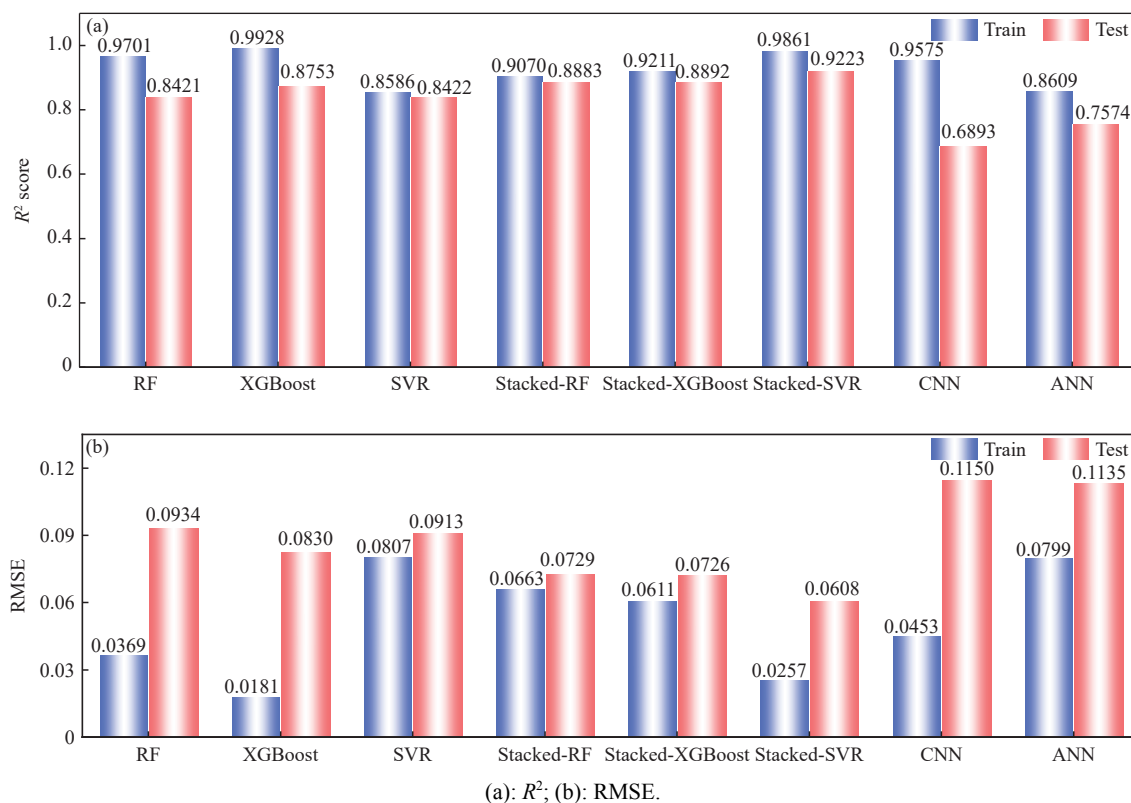


Fig. 5 Different machine learning models for predicting Y_{NH_3} on training and test sets

However, these two algorithms perform poorly in predicting the Y_{NH_3} test set, with R^2 values of 0.8421 and 0.8753 and RMSE values of 0.0934 and 0.0830, suggesting that these algorithms suffer from overfitting. A comprehensive analysis reveals that the Stacked-SVR algorithm demonstrates high accuracy in predicting Y_{NH_3} performance, with an R^2 of 0.9861 and RMSE of 0.0257 on the training set. On the test set, the R^2 is 0.9223 and the RMSE is 0.0608. These results indicate that the Stacked-SVR model effectively integrates the complementary strengths of RF, XGBoost, and SVR, achieving an optimal balance between bias and variance, significantly improving the prediction performance for Y_{NH_3} .

Figure 6 illustrates the predictive performance of different algorithms for the F_{NH_3} on both the training and test sets. Consistent with the prediction results for Y_{NH_3} , the two neural network models show poorer predictive performance compared to the machine learning models, whereas the stacking models consistently outperform the standalone models. Among them, the Stacked-RF algorithm achieves the optimal predictive performance for F_{NH_3} . The results, evaluated with respect to RMSE and R^2 , are displayed in Figure 7. The SVR algorithm exhibits the lowest R^2 and relatively high RMSE on both the training and test sets, indicating its poor predictive capability for F_{NH_3} .

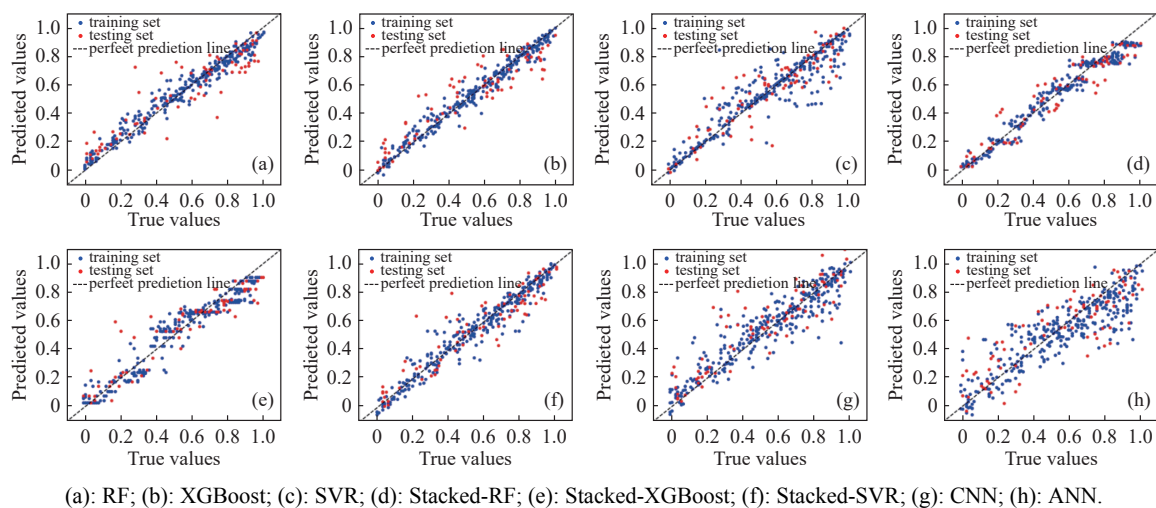


Fig. 6 Validation results of different machine learning models for predicting F_{NH_3}

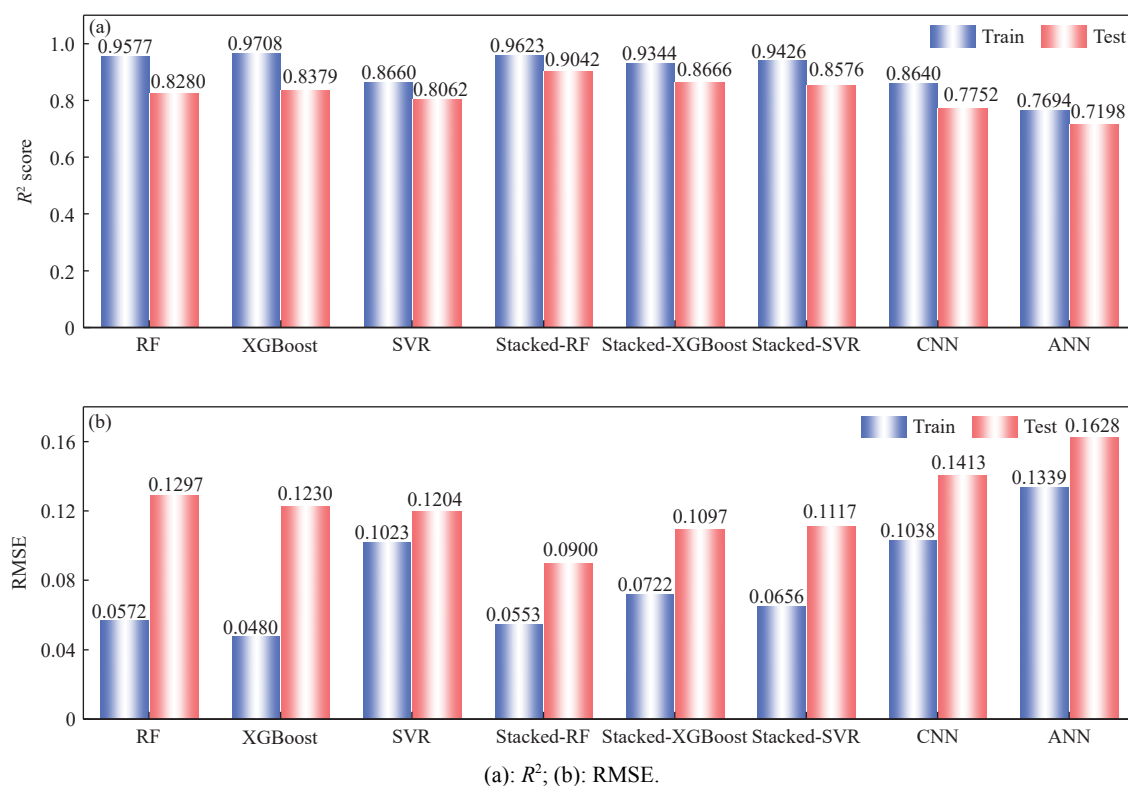


Fig. 7 Different machine learning models for predicting F_{NH_3} on training and test sets

The RF and XGBoost algorithms show high R^2 on the training set (0.9577 and 0.9708, respectively), but a noticeable decrease in R^2 on the test set (0.8280 and 0.8379, respectively), suggesting the presence of overfitting in predicting F_{NH_3} . In contrast, the stacking ensemble model alleviates overfitting by combining the complementary strengths of multiple algorithms. Although the predictive performances of Stacked-XGBoost and Stacked-SVR are superior to those of the standalone models, they are still less accurate than Stacked-RF, which achieves a test set R^2 of 0.9042 and RMSE of 0.0900. This superior performance may be because Random Forest, as the meta-model in Stacked-RF, better captures the underlying data distribution.

Based on the above, the stacking model demonstrates significantly superior predictive performance compared to individual models. By integrating the strengths of multiple base learners, it can more comprehensively capture the complex relationships between data features and the target variable, effectively balance model bias and variance, and minimize the performance disparity between the training and test sets. Furthermore, by intelligently integrating the outputs of multiple base models, the meta-learner enhances overall predictive accuracy, strengthens model robustness, and improves generalization capability.

2.3 Model Interpretability analysis based on SHAP values

Model interpretation is a critical step in machine learning development, aimed at validating the consistency between predictive results and experimental data^[33]. Traditional machine learning models are often regarded as a “black-box”, making it difficult to decipher how individual input features influence the output. In this study, the SHAP method, derived from game theory, was used to quantify the contribution of each feature to the prediction results^[34].

Through global and local interpretation, the influence of input parameters such as preparation conditions and reaction conditions on Y_{NH_3} and F_{NH_3} was systematically analyzed to provide a theoretical basis for understanding complex black-box models. Figure 8 presents a feature importance analysis for Y_{NH_3} and F_{NH_3} based on the SHAP method. In the SHAP summary plot, features are sorted by the mean absolute value of their SHAP values, where a higher rank reflects a stronger overall influence of that feature on the model’s predictive outcomes^[35]. A positive SHAP value suggests that the feature has a positive impact on the prediction, whereas a negative value indicates a negative influence on the predicted outcome^[36].

Figure 8(a) shows that working potential and electrolyte volume have the most significant impact on Y_{NH_3} ($|\text{SHAP value}| > 0.05$). As the working potential decreases, the corresponding SHAP value increases, indicating that lower potentials promote higher Y_{NH_3} . Similarly, the electrolyte volume shows that moderate or lower volumes are more conducive to ammonia synthesis, while excessive volumes inhibit the reaction and result in performance deterioration. In contrast, electrolyte concentration, NO inflow volume, and calcination time exhibit relatively weaker influences. Among these, both electrolyte concentration and NO inflow volume show positive correlations with Y_{NH_3} , as indicated by their increasing SHAP values with higher parameter values, suggesting that elevated reactant concentrations promote ammonia synthesis. Meanwhile, extending the calcination time within an appropriate range also has a positive effect on Y_{NH_3} . On the other hand, F_{NH_3} is another key performance indicator for the NORR reaction. Global interpretability analysis based on SHAP (Figure 8(b)) reveals that working potential, electrolyte volume, and catalyst loading significantly affect F_{NH_3} ($|\text{SHAP value}| > 0.03$). Among them, the working potential exhibits a wide distribution of SHAP values across its range, indicating a nonlinear relationship that is not solely dependent on unidirectional changes in potential.

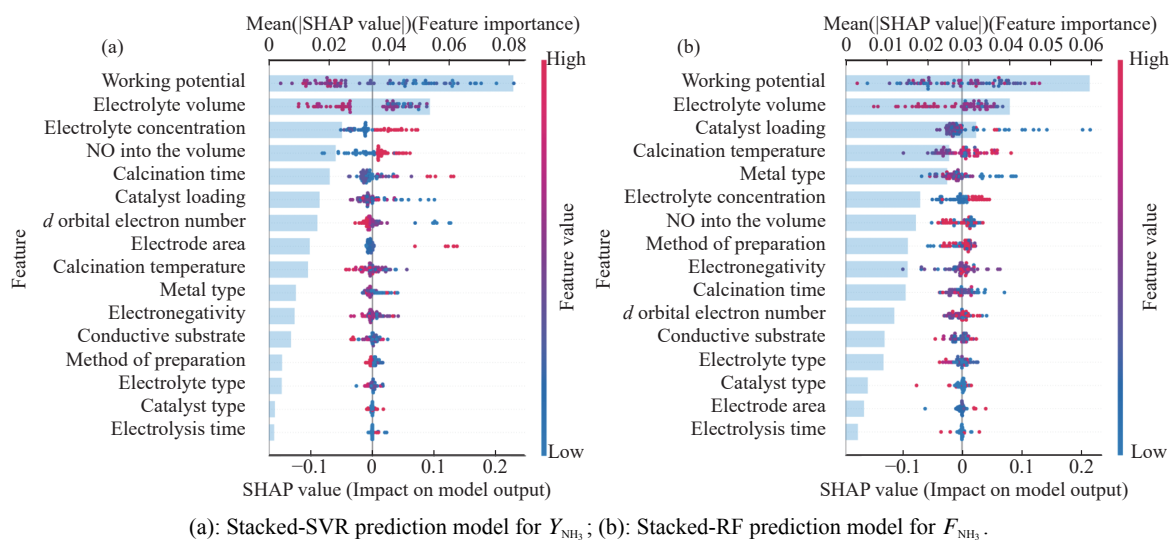


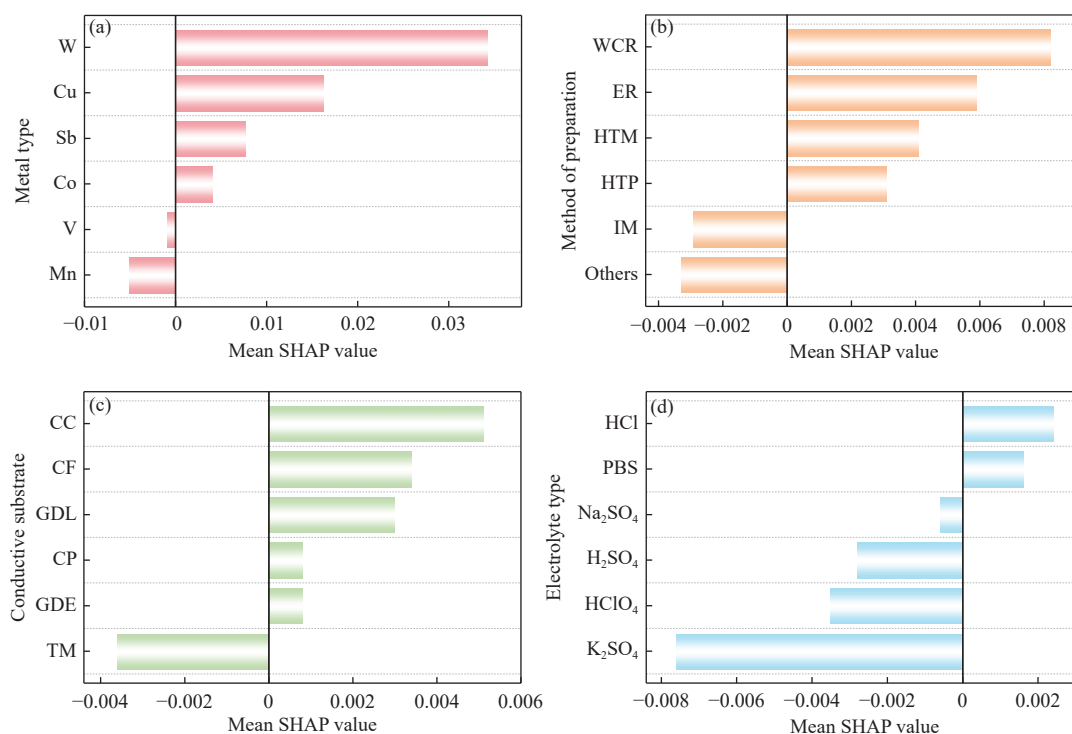
Fig. 8 SHAP-based feature importance analysis plots

A reduction in electrolyte volume is associated with higher SHAP values, suggesting that moderate or smaller volumes favor performance optimization. Similarly, an increase in SHAP value at lower catalyst loading implies that higher dispersion of the catalyst enhances F_{NH_3} . In comparison, calcination temperature, electrolyte concentration, and NO inflow volume serve as secondary influencing factors. Among these, increases in calcination temperature and electrolyte concentration both show positive correlations with F_{NH_3} .

Based on the comprehensive SHAP analysis, working potential and electrolyte volume are identified as the key factors influencing both Y_{NH_3} and F_{NH_3} . Among them, working potential exhibits a significant negative correlation with Y_{NH_3} , indicating that appropriately reducing the potential can effectively enhance Y_{NH_3} . The electrolyte volume should be kept within a moderate or lower range, as this condition promotes the synergistic optimization of both performance indicators. In catalyst design, appropriately reducing the catalyst loading facilitates the exposure and dispersion of active sites, thereby simultaneously improving Y_{NH_3} and F_{NH_3} . Furthermore, increasing the electrolyte concentration has a positive effect on both Y_{NH_3} and F_{NH_3} .

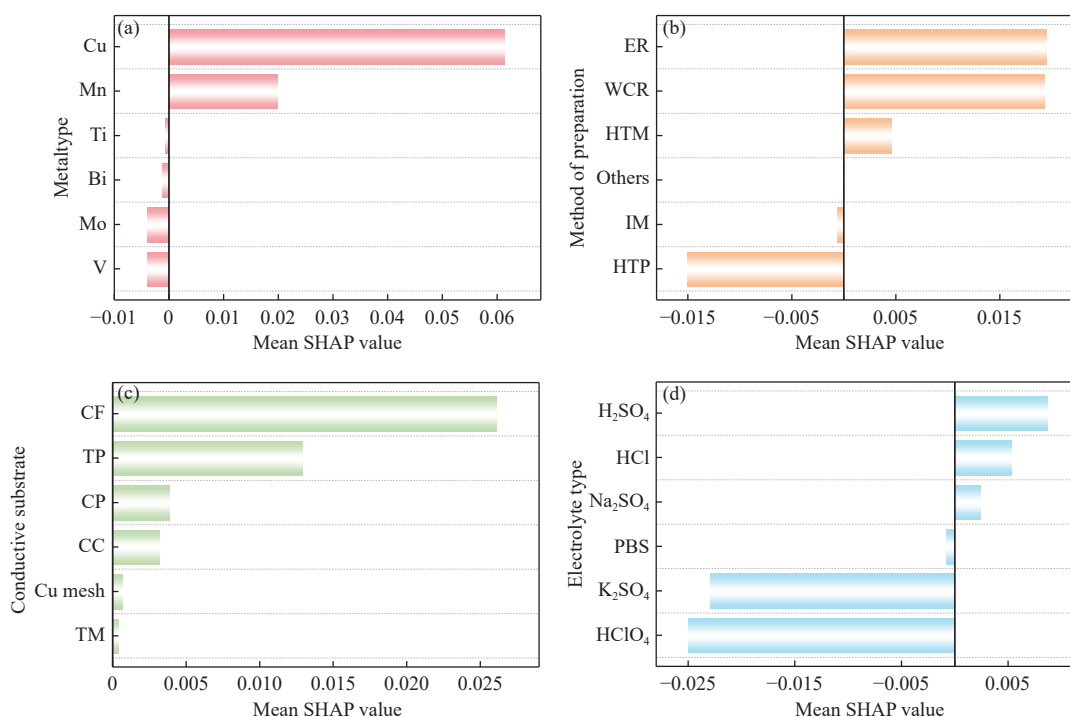
The composition of the catalyst plays a crucial role in its performance for the NORR reaction. Figure 9 and Figure 10 present the results of local feature importance analysis based on the SHAP method. Regarding the selection of metal elements, W and Cu contribute significantly to Y_{NH_3} (Figure 9(a)), while Cu also exhibits a notable positive

effect on F_{NH_3} (Figure 10(a)). Therefore, Cu is identified as the most suitable metal component for the catalyst. Cu-based catalysts significantly accelerate interfacial electron transfer due to their low charge transfer resistance^[37]. Their excellent electron-trapping capability effectively suppresses electron-hole recombination, extends electron lifetime, and improves electron utilization efficiency^[38]. Moreover, the strong interaction between Cu and the support facilitates the transfer of *d*-orbital electrons from Cu to the support, forming electron-deficient Cu species. This significantly enhances the adsorption and activation of NO molecules^[39], thereby improving NORR catalytic performance. As shown in Figure 9(b) and Figure 10(b), the wet chemical reduction (WCR) method has a significant positive effect on Y_{NH_3} and performs comparably to electroreduction (ER) in enhancing F_{NH_3} . Considering both stability and overall contribution, WCR is identified as the preferred option. For conductive substrates, carbon fiber (CF) demonstrates superior performance for both Y_{NH_3} and F_{NH_3} (Figure 9(c) and Figure 10(c)), which is significantly better than titanium plate (TP), carbon paper (CP), and carbon cloth (CC). Thus, CF is determined as the optimal conductive substrate. For electrolyte types, HCl positively contributes to both Y_{NH_3} and F_{NH_3} (Figure 9(d) and Figure 10(d)), demonstrating better synergistic effects. Although H_2SO_4 shows a stronger effect in enhancing F_{NH_3} , HCl proves to be more advantageous when both performance indicators are considered simultaneously.



(a): Metal type; (b): Method of preparation; (c): Conductive substrate; (d): Electrolyte type.

Fig. 9 Local interpretation plot of the Y_{NH_3} prediction model based on SHAP values



(a): Metal type; (b): Method of preparation; (c): Conductive substrate; (d): Electrolyte type.

Fig. 10 Local interpretation plot of the F_{NH_3} prediction model based on SHAP values

Based on the comprehensive SHAP analysis, the optimal configuration is determined to be a Cu-based catalyst *via* the wet chemical reduction method, using carbon fiber (CF) as the conductive substrate and HCl as the electrolyte. This optimized combination significantly enhances Y_{NH_3} while maintaining high F_{NH_3} , providing valuable insights for the optimization of NORR processes and the design of high-performance catalysts.

3 Conclusion

In this study, a machine learning-driven screening model for NORR catalysts was established and a stacked ensemble approach was employed to achieve high-precision prediction of Y_{NH_3} and F_{NH_3} . Through feature importance analysis and the SHAP interpretability framework, the influence of various factors on performance was systematically elucidated. The working potential was identified as the most influential factor, and its appropriate reduction significantly improved Y_{NH_3} . Maintaining the electrolyte volume at a moderate or low level was found to be more conducive to the synergistic optimization of both Y_{NH_3} and F_{NH_3} . Furthermore, moderately reducing the catalyst loading and increasing the electrolyte concentration can further enhance the reaction activity.

Based on the local feature importance analysis, the combination of a Cu-based catalyst, prepared *via* the wet chemical reduction method, using carbon fiber (CF) as the conductive substrate and HCl as the electrolyte, was identified as the optimal system. This configuration

enhances overall performance while effectively balancing both Y_{NH_3} and F_{NH_3} . This research provides a reliable data-driven strategy for the development and optimized design of catalysts for the efficient electrocatalytic reduction of nitric oxide to ammonia.

Declaration of Competing Interest

The authors declare no conflict of interest.

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基于堆叠集成学习的电催化 NO 还原合成氨催化剂设计研究

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摘 要: 电催化还原一氧化氮合成氨(NORR)作为绿色能源转化领域的一项核心技术, 开发高效的 NORR 电催化剂是提高氨产量 Y_{NH_3} 和法拉第效率 F_{NH_3} 的关键。传统实验方法在高活性 NORR 催化剂筛选中通常伴随较高的资源消耗和时间成本。本研究采用机器学习与 SHAP 特征分析相结合的方法, 基于单一算法构建堆叠集成模型算法, 依托实验数据库系统分析影响 NORR 反应性能的关键特征。通过评估八种模型算法发现, Stacked-SVR 模型预测 Y_{NH_3} 测试集 R^2 为 0.9223, RMSE 为 0.0608; Stacked-RF 模型预测 F_{NH_3} 测试集 R^2 为 0.9042, RMSE 为 0.0900。堆叠集成模型整合了各独立模型算法的优势, 在避免过拟合的情况下显示出较强的 NORR 预测性能。SHAP 特征分析结果表明, 催化剂组成中 Cu 元素含量对催化性能影响最为显著; 湿化学还原法制备、采用碳纤维(CP)导电衬底和 HCl 电解液的组合更利于催化活性提升; 同时, 适度降低工作电位、控制电解液体积于中低水平、减少催化剂负载量并提高电解液浓度, 可协同促进 Y_{NH_3} 与 F_{NH_3} 的同步提升。

关键词: NORR; 机器学习; 堆叠模型; 氨产量; 氨法拉第效率

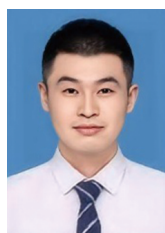
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