Machine Learning-Based Screening of Li compounds for Solid State Batteries

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Abstract

Solid-state lithium-ion batteries (SSLIBs) offer significant advantages over conventional liquid-electrolyte batteries due to their improved safety, higher energy density, and longer cycle life. However, the widespread adoption of SSLIBs is limited by poor ionic conductivity and high interfacial resistance of the solid electrolyte material. In this work, we develop a machine learning (ML) framework to accelerate the discovery of superionic lithium-based solid electrolytes. We train and compare multiple ML models, including logistic regression, bagging, XGBoost, and neural networks, to classify lithium-containing materials as superionic or non-superionic. These classifications are verified using measured ionic conductivity values and exhibit that advanced ML techniques outperform traditional linear models, with neural networks achieving the highest predictive accuracy (79%) and recall (70%) for superionic materials. SHAP analysis further reveals the importance of Li–anion separation distance and anion coordination features. Furthermore, after pre-screening 21,000+ Li-based compounds using structural descriptors derived from Density Functional Theory (DFT) calculations, we apply Neural Networks, our best-performing model, to screen 560 Li-based compounds, identifying promising candidates with high predicted stability and conductivity. Among the predicted candidates, compounds like $Li_{12}Be_6F_{24}$, $Li_{12}P_{12}O_{36}$, and $Li_3B_3F_{12}$ stand out as promising compounds for application in SSLIBs. Our findings demonstrate the potential of ML to significantly accelerate the discovery of next-generation solid electrolytes, reducing the reliance on costly experimental and computational screening methods.

1. Introduction

Solid-state lithium-ion batteries are promising next-generation energy storage solutions, offering improved safety, longer lifespan, and higher energy density compared to conventional lithium-ion batteries (LIBs) with liquid electrolytes. However, the widespread adoption of SSLIBs is limited by poor ionic conductivity and high interfacial resistance of the solid electrolyte material. A high-performance solid electrolyte must enable fast lithium-ion transport, have minimal electronic conductivity, exhibit a wide electrochemical window, and maintain stability against chemical reactions with electrodes. Additionally, it should possess sufficient mechanical strength to prevent lithium dendrite formation, while being cost-effective and scalable for manufacturing.

Traditional material discovery approaches rely heavily on trial-and-error experimentation, making the search for optimal solid electrolytes slow and resource-intensive. While computational methods such as Density Functional Theory (DFT) have accelerated screening by predicting key properties (Sendek et al., 2019), they remain computationally expensive and may not fully capture complex ionic transport mechanisms. Machine learning (ML) has emerged as a powerful alternative, offering data-driven models that can predict material properties more efficiently. Previous ML-based studies have primarily focused on linear models like logistic regression (Sendek et al. 2017, 2019), which provide interpretability but struggle to capture non-linear relationships in material descriptors.

In this work, we develop an advanced ML framework to identify promising superionic conductors for SSLIBs. We leverage a combination of logistic regression, bagging, XGBoost, and deep learning models (Multi-Layer Perceptron) to improve prediction accuracy. We further enhance interpretability through SHAP analysis, providing insights into key structural features that influence ionic conductivity. Our approach shows promising results compared to traditional models, highlighting the potential of ML to aid in solid electrolyte discovery.

By leveraging high-throughput data from the Materials Project Database (MPD) (Jain et al., 2013) and applying advanced ML techniques, we develop a methodology for materials screening that balances scalability and interpretability. While our work contributes to refining computational screening strategies, further validation and improvements are needed to fully capture the complexities of ionic conductivity. Nonetheless, our findings offer useful insights into structure-property relationships that may help guide future

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experimental research on solid-state electrolytes.

2. Related Work

Early ML applications in solid electrolyte discovery relied on logistic regression models, such as those by (Sendek et al. 2017, 2019), which screened over 12,000 lithium-containing materials. Their approach used structural descriptors — Li–Li bond count (LLB), sublattice bond ionicity (SBI), anion framework coordination (AFC), Li–anion separation distance (LASD), and Li–Li separation distance (LLSD) — to predict ionic conductivity. While their model effectively reduced the search space by over 99%, its assumption of linear relationships between features and conductivity limited its accuracy and generalizability, particularly given the small training dataset (38 materials). To address these limitations, more advanced ML methods have been explored. Tree-based models like Extreme Gradient Boosting (XGBoost) capture non-linear relationships among structural descriptors, offering improved predictive performance. Ensemble methods, such as bagging, enhance robustness and reduce overfitting. Moreover, neural networks improve the model accuracy, even with limited available data. Despite these advancements, many effective ML methods are yet to be explored. Most studies focus on single-model approaches rather than leveraging ensemble learning or hybrid architectures for better accuracy. In addition, only a few incorporate uncertainty quantification, making it difficult to assess model confidence and guide experimental validation effectively. Our work addresses these gaps by implementing advanced ML techniques, including bagging, XGBoost, and MLP neural networks, trained on the dataset used by Sendek et al. We incorporate SHAP feature importance analysis to quantify uncertainty and improve interpretability. By tackling these limitations, our approach provides a more robust, accurate, and interpretable framework for accelerating the discovery of high-performance solid-state lithium-ion conductors.

3. Methods

The code and data used for this project can be found in GitHub (Megan Hyatt, 2025).

3.1. Li Sample Set

We sourced our dataset from the Materials Project Database (MPD) using Pymatgen (Shyue Ping Ong, 2013), a Python package for analyzing materials properties. This dataset includes Density Functional Theory (DFT)-calculated atomistic and electronic structure information for over 21,000 lithium-containing materials. Key extracted parameters include atomic structures, electronic band gap, and thermodynamic stability.

3.2. Feature Engineering

We computed five structural descriptors useful for determining superionicity of atomic structures using Pymatgen:

- **LLB**: Li–Li bond count per Li atom
- **SBI**: Sublattice bond ionicity (electronegativity difference between Li and neighboring anions)
- **AFC**: Average anion coordination number
- **LASD**: Mean Li–anion separation distance
- **LLSD**: Mean Li–Li separation distance

3.3. Train Set

Training data is manually extracted from the Li-based compounds listed in the MPD with DFT-verified ionic conductivity values according to tables 1 and 2 in (Sendek et al. 2017, 2019). Our feature matrix is a compilation of the 5 structural features described above, and our classification vector is a Boolean vector that distinguishes between superionic structures ($\sigma \geq 10^{-4} S/cm$) and non-superionic structures ($\sigma < 10^{-4} S/cm$).

3.4. Test Set

Two test sets are implemented in this paper. The first test set is based on a verifiable set of materials and their corresponding experimentally-measured ionic conductivities listed in the Liverpool Ionics Dataset (Hargreaves et al., 2023). Filtering through this dataset for materials that also exist in the MPD provided us with a test set containing 28 elements. Our feature matrix is a compilation of the 5 structural features described above for these 28 elements, and our classification vector used to determine performance metrics for each of our ML models is a Boolean vector that distinguishes between superionic structures ($\sigma \geq 10^{-4} S/cm$) and non-superionic structures ($\sigma < 10^{-4} S/cm$) listed in the Liverpool Ionics Database.

The second test set contains all Li-based compounds in the MPD, which we pre-filtered for prerequisite requirements that determine feasibility for use as a solid electrolyte (outside of ionic conductivity) including:

1. **Band gap $>$ 1 eV**: Materials with a band gap lower than 1 eV can conduct electrons too easily and thus would not be effective ion conductors.
2. **Energy above hull $=$ 0 eV**: Materials need to be thermodynamically stable to operate as an effective ion conductor, so Ehull should be equal to 0 eV.
3. **Does not contain transition metals**: Transition metals are susceptible to reactions with lithium at the low voltage of the anode and thus may not operate as stable ion conductors.

This filtering process shrinks the size of our dataset from 21,756 to 560 Li-based materials.

4. Model Architecture and Training

4.1. Baseline Logistic Regression Model

We first implemented a logistic regression model replicating (Sendek et al., 2019), which predicts the probability of a material being a superionic conductor using:

$$P_{\text{superionic}}(x) = \left(1 + e^{-\sum_i \Theta_i x_i}\right)^{-1}, \tag{1}$$

$$\begin{aligned} \text{where } \sum_i \Theta_i x_i = 0 \times \text{LLB} + \ 0.39 \times \text{SBI} \\ - 0.91 \times \text{AFC} + \ 0.67 \times \text{LASD} \\ - 0.18 \times \text{LLSD} + 0.22. \end{aligned} \tag{2}$$

where $n = 5$ is the number of structural descriptors in the model, θ_i are the regression coefficients calculated by our base LR model, and $x_0 \equiv 1$ for all materials to allow for an intercept term θ_0 in the sum. The output of this logistic regression is a scalar value between 0 and 1 indicating the probability that material i belongs to the superionic class:

$$\tilde{\sigma} = \begin{cases} 1, & \sigma \geq 10^{-4} \text{ S cm}^{-1} \\ 0, & \sigma < 10^{-4} \text{ S cm}^{-1} \end{cases} \tag{3}$$

4.2. Advanced Machine Learning Models

To improve prediction accuracy, we implemented advanced models:

- **XGBoost**: A gradient boosting algorithm effective in handling non-linear interactions and structured tabular data.
- **Bagging (Bootstrap Aggregating)**: Trained on bootstrapped datasets to reduce variance, using Logistic Regression as the base estimator.
- **Neural Networks (MLP Classifier)**: A multi-layer perceptron trained with hyperparameter tuning to capture complex structure–property relationships.

4.3. Hyperparameter Tuning

Hyperparameter tuning was conducted using GridSearchCV, optimizing parameters through five-fold cross-validation.

Model	Hyperparameters
XGBoost	learning_rate: 0.1, max_depth: 3, subsample: 1, n_estimators: 100
Neural Networks	hidden_layer_sizes: (20,), activation: tanh, alpha: 0.0001, learning_rate: constant, max_iter: 500

Table 1. Hyperparameter tuning for XGBoost and Neural Network models using grid search.

The best-performing parameters were selected based on maximizing the F1 score. The key parameters are shown in Table 1.

4.4. Evaluation Metrics

We assessed model performance using standard classification metrics:

- **Precision**: Fraction of correctly predicted superionic materials.
- **Recall**: Fraction of actual superionic materials identified.
- **F1 Score**: Harmonic mean of precision and recall, used as the primary metric.
- **Accuracy**: Overall prediction correctness.
- **AUC–ROC**: Area under the receiver operating characteristic curve; used to evaluate prediction quality across all classification thresholds.

4.5. Feature Importance Analysis

To enhance interpretability, we used SHAP (SHapley Additive exPlanations) to quantify feature contributions. SHAP values provided insight into which structural descriptors most influenced ionic conductivity predictions.

4.6. Implementation of Best Classification Model

We implement the best-performing classification model to screen a test set of 560 Li-based compounds, identifying promising candidates with high predicted stability and ionic conductivity for further analysis.

5. Results and Discussion

5.1. Quantitative Performance Comparison

We evaluated four machine learning models: Logistic Regression (LR), XGBoost, Bagging (with LR as the base model), and Neural Networks based on their ability to classify lithium-containing materials as superionic or non-superionic conductors. The dataset included 28 experimentally characterized materials, with 10 superionic and 18 non-superionic instances. Performance was assessed using accuracy, precision, recall, F1 score, ROC-AUC, and confusion matrices.

Logistic Regression, serving as the baseline, achieved 64% accuracy and an ROC-AUC of 0.74. However, it struggled with class imbalance, correctly identifying 94% of non-superionic materials but only 10% of superionic ones. The F1 score for superionic classification was just 0.17, reflecting its difficulty in detecting positive cases.

XGBoost improved overall performance, achieving 71% accuracy with more balanced precision (78% for non-

superionic, 60% for superionic) and a significantly higher F1 score of 0.60 for superionic materials. The confusion matrix showed that XGBoost correctly identified 6 out of 10 superionic materials, demonstrating a better balance between recall and precision. Its ROC-AUC of 0.79 further confirmed its superior ability to distinguish between classes.

Metric	LR	XGB	Bagging	NN
Accuracy	0.64	0.71	0.68	0.79
Precision (0)	0.65	0.78	0.67	0.83
Precision (1)	0.50	0.60	1.00	0.70
Recall (0)	0.94	0.78	1.00	0.83
Recall (1)	0.10	0.60	0.10	0.70
F1 Score (0)	0.77	0.78	0.80	0.83
F1 Score (1)	0.17	0.60	0.18	0.70
ROC-AUC	0.74	0.79	0.69	0.77

Table 2. Comparison of classification metrics for Logistic Regression (LR), XGBoost (XGB), Bagging, and Neural Networks (NN).

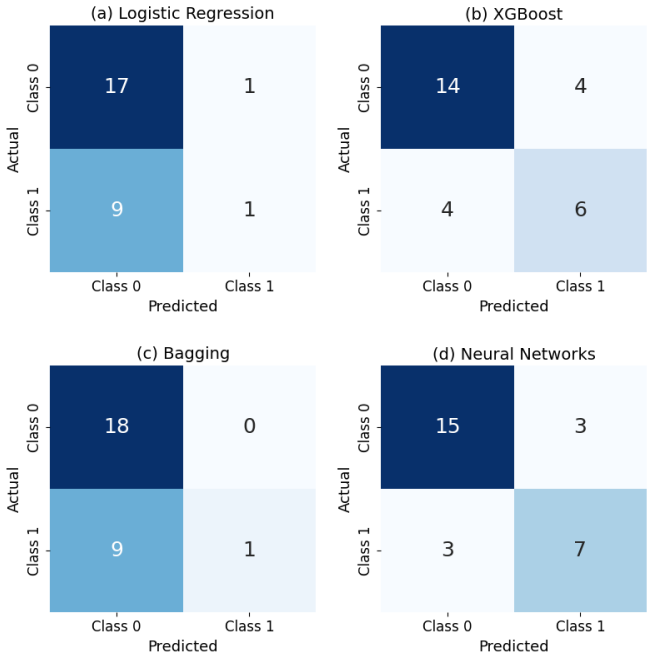

Figure 1. Confusion matrices for different classification models: (a) Logistic Regression, (b) XGBoost, (c) Bagging, and (d) Neural Networks. Each matrix illustrates the distribution of predicted vs. actual class labels.

Bagging offered only marginal improvements over standalone LR. While recall for superionic materials increased slightly to 20%, overall accuracy remained 64%, and the ROC-AUC dropped to 0.69. These results suggest that bagging provides limited benefits when applied to a weak linear classifier in this context.

Neural Networks outperformed all other models, achieving 79% accuracy with a recall of 70% for superionic materials. The F1 score for superionic classification reached 0.70, the highest among all models. The confusion matrix showed that it correctly identified 7 out of 10 superionic materials while maintaining strong performance on non-superionic cases. Its ROC-AUC of 0.77 indicated robust predictive capability, though slightly lower than XGBoost.

5.2. Ablation Study and Model Analysis

To understand the impact of model complexity, we compared Logistic Regression to the more advanced XGBoost and Neural Network models. The results show that simpler linear models struggle to capture the non-linear relationships essential for predicting ionic conductivity.

XGBoost improved classification by using gradient-boosted decision trees, capturing complex interactions between structural descriptors. Its ensemble nature also helped mitigate overfitting despite the small dataset. However, Bagging did not yield significant improvements, suggesting that linear classifiers such as LR lack the expressiveness needed for this task.

Neural Networks further enhanced prediction quality by learning non-linear patterns without requiring extensive feature engineering. Their strong performance suggests that deep learning approaches hold promise for modeling ionic transport, provided sufficient training data is available.

5.3. Limitations and Failure Cases

While XGBoost and Neural Networks improved predictive performance, several challenges remain. The models were trained on very limited data of 38 samples, and the test set included only 28 materials. This restricts model generalization and increases the risk of overfitting. With many more non-superionic samples as compared to superionic ones, the models struggled with recall for positive cases. Even the best model (Neural Networks) correctly identified only 7 out of 10 superionic materials, leaving room for further improvements.

Additional filtering of the test data could also provide more accurate results: our estimate of $E_{hull} < 0.1$ eV was used as an approximate indicator of thermal stability due to limited viable compounds in our training set, when in reality $E_{hull} = 0$ eV would be a more appropriate indicator. Another useful indicator would be to ensure that oxidation voltage at the cathode is high enough to prevent the formation high-resistance interfacial layers that limit ionic current ($V_{ox} > 4$ V for the typical cathode voltage). Finally, screen-

ing materials for their cost and abundance can ensure that results are practical.

5.4. SHAP Feature Analysis

To gain insight into the Neural Network's predictions, we performed SHAP (SHapley Additive exPlanations) analysis. This identified Li-Anion Separation Distance (LASD) as the most influential feature, with higher values strongly correlating with superionic conductivity.

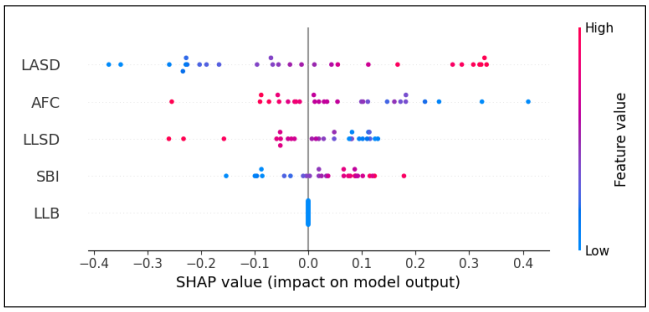

Figure 2. SHAP analysis of the Neural Network model. Li-Anion Separation Distance (LASD) is identified as the most influential feature, with higher values strongly correlating with superionic conductivity.

Other key insights from SHAP analysis include:

- Anion Framework Coordination (AFC) showed mixed effects, suggesting complex dependencies between structural rigidity and ionic transport.
- Li-Li Separation Distance (LLSD) positively influenced predictions, aligning with the hypothesis that greater Li-Li distances aid conductivity.
- Sublattice Bond Ionicity (SBI) contributed variably, depending on the ionicity level.
- Li-Li Bond Count (LLB) had negligible impact, implying it is not a key predictor in this case.

5.5. Analysis of Predicted Superionic Compounds

The neural network model identified a diverse set of lithium-containing compounds as potential superionic conductors. Many predicted compounds exhibited negative formation energies per atom, indicating strong stability. For example, $Li_{12}Be_6F_{24}$ and $Li_{18}Al_6F_{36}$ showed formation energies of -3.36 eV/atom and -3.49 eV/atom, respectively, suggesting they are energetically favorable phases.

The predictions spanned a wide range of chemical compositions and structural motifs, including halides ($Li_4Al_4Cl_{16}$, $Li_4Ga_4Cl_{16}$), oxides ($Li_{12}P_{12}O_{36}$, $Li_8S_8O_{28}$), fluorides ($Li_{18}Al_6F_{36}$, $Li_6Si_3F_{18}$), and mixed anion systems ($Li_2S_2O_6F_2$, $Li_4H_8Cl_4O_{20}$). The ability of the model to generalize across different chemistries suggests it captures fundamental descriptors relevant to ionic conductivity. The model also identified multi-element compounds, such as $Cs_3Li_3H_{12}N_6$ (a cesium-lithium hydride-nitride system) and $Na_8Li_{16}B_8O_{24}$ (a sodium-lithium borate system), adding to the design space for novel solid electrolytes.

Several predicted materials also had exceptionally large bandgaps, a critical property for solid electrolytes to prevent electronic leakage. $Li_3B_3F_{12}$ and $Li_6Si_3F_{18}$ demonstrated bandgap values exceeding 8 eV, highlighting their potential as excellent electronic insulators.

Among the predicted candidates, compounds like $Li_{12}Be_6F_{24}$, $Li_{12}P_{12}O_{36}$, and $Li_3B_3F_{12}$ stand out as promising compounds for application in SSLIBs. These materials should be further examined using DFT molecular dynamics simulations to confirm their ionic conductivity under operating conditions, specifically at room temperature.

6. Conclusion

The discovery of high-performance solid electrolytes is a critical challenge for the development of next-generation solid-state lithium-ion batteries (SSLIBs). In this work, we demonstrated various machine learning (ML) models for accelerating the identification of superionic lithium-containing materials. We trained and evaluated multiple ML models, including logistic regression, bagging, XGBoost, and neural networks. These classifications were verified using measured ionic conductivity values and showed that advanced models, particularly neural networks, significantly outperform traditional approaches, achieving high accuracy (79%) and recall (70%) in classifying superionic conductors.

Beyond predictive performance, we applied SHAP analysis to further reveal key structural features that govern ionic conductivity, including Li–anion separation distance and anion coordination. This insight provides valuable guidance for the rational design of new solid electrolyte materials. Additionally, after pre-screening 21,000+ Li-based compounds using structural descriptors derived from Density Functional Theory (DFT) calculations, we used our trained models to screen 560 Li-based compounds, identifying promising candidates for further computational and experimental validation. Among the predicted candidates, compounds like $Li_{12}Be_6F_{24}$, $Li_{12}P_{12}O_{36}$, and $Li_3B_3F_{12}$ stand out as promising compounds for application in SSLIBs.

While our ML-based screening approach demonstrates substantial improvements over traditional methods, improvements can be made. Increasing the train and test set sizes would prevent model generalization andoverfitting, and increasing the ratio of superionic samples would improve recall. Improving indicators of superionicity by filtering the test set compounds without $E_{hull} = 0eV$ and $V_{ox} > 4V$ could also increase accuracy. Finally, screening materials for their cost and abundance can ensure that results are

practical.

Overall, this study underscores the potential of machine learning to transform materials discovery, offering a scalable, data-driven framework for identifying novel superionic conductors. By bridging computational screening with experimental validation, our approach can accelerate the path toward commercially viable solid-state lithium-ion batteries.

References

Hargreaves, C. J., Gaultois, M. W., Daniels, L. M., Watts, E. J., Kurlin, V. A., Moran, M., Dang, Y., Morris, R., Morscher, A., Thompson, K., Wright, M. A., Prasad, B.-E., Blanc, F., Collins, C. M., Crawford, C. A., Duff, B. B., Evans, J., Gamon, J., Han, G., Leube, B. T., Niu, H., Perez, A. J., Robinson, A., Rogan, O., Sharp, P. M., Shoko, E., Sonni, M., Thomas, W. J., Vasylenko, A., Wang, L., Rosseinsky, M. J., and Dyer, M. S. A database of experimentally measured lithium solid electrolyte conductivities evaluated with machine learning. *npj Computational Materials*, 9(1):9, 2023. ISSN 2057-3960. doi: 10.1038/s41524-022-00951-z. URL https://doi.org/10.1038/s41524-022-00951-z.

Jain, A., Ong, S. P., Hautier, G., Chen, W., Richards, W. D., Dacek, S., Cholia, S., Gunter, D., Skinner, D., Ceder, G., and Persson, K. A. Commentary: The materials project: A materials genome approach to accelerating materials innovation. *APL Materials*, 1(1):011002, 07 2013. ISSN 2166-532X. doi: 10.1063/1.4812323.

Megan Hyatt, A. B. Matsci 176: Machine learning-based screening of li compounds for solid state batteries, 2025.

Sendek, A. D., Cubuk, E. D., Antoniuk, E. R., Cheon, G., Cui, Y., and Reed, E. J. Machine learning-assisted discovery of solid li-ion conducting materials. *Chemistry of Materials*, 31(2):342–352, 2019. doi: 10.1021/acs.chemmater.8b03272.

Shyue Ping Ong, William Davidson Richards, A. J. G. H. M. K. S. C. D. G. V. L. C. K. A. P. G. C. Python materials genomics (pymatgen): A robust, open-source python library for materials analysis. *Computational Materials Science*, 68:314–319, 2013. ISSN 0927-0256. doi: https://doi.org/10.1016/j.commatsci.2012.10.028.