

Utilizing Deep Learning for Crystal System Classification in Lithium - Ion Batteries

Yibo Yin¹, Guokun Xu², Ying Xie³, Yang Luo⁴, Zibu Wei⁵, Zhengning Li⁶

¹Computer Science, Contemporary Amperex Technology USA Inc, Auburn Hills, USA

²Computer Science, Beijing Foreign Studies University, Beijing, China

³Computer Science, San Francisco Bay University, Fremont, USA

⁴Computer Science, China CITIC Bank Software Development Center, Beijing, China

⁵Computer Science, University of California, Los Angeles, Los Angeles, USA

⁶Computer Science, Georgetown University, Washington, D.C. USA

¹epark00007@gmail.com; ²nicolashsu@hotmail.com; ³floraxlr999@gmail.com; ⁴luoyangdxx@163.com; ⁵zibuwei@ucla.edu; ⁶z1132@georgetown.edu

Abstract: *Lithium-ion (Li-ion) batteries are pivotal in energy storage, powering diverse applications from portable electronics to electric vehicles. Optimizing Li-ion battery performance relies on understanding the crystal system properties of constituent materials, notably cathodes. This paper proposes a novel approach using Deep Neural Networks (DNNs) for multi-class classification of Li-ion silicate cathode crystal systems. Previous research underscores crystal chemistry's importance and the potential of machine learning in Li-ion battery materials. However, existing methodologies face challenges in accurately capturing material complexities. Our DNN-based model aims to address these limitations, offering improved predictive performance for crystal system classification.*

Keywords: Lithium-ion batteries; Crystal system properties; Deep Neural Networks.

1. INTRODUCTION

Lithium-ion (Li-ion) batteries have emerged as a cornerstone technology in the field of energy storage, powering a wide array of devices from portable electronics to electric vehicles. A crucial aspect in optimizing the performance and design of Li-ion batteries lies in understanding the crystal system properties of their constituent materials, particularly cathodes. The crystal system of these materials influences their structural stability, electrochemical behavior, and overall performance within the battery system. This paper focuses on elucidating the crystal system properties of Li-ion silicate cathodes, which play a pivotal role in determining the battery's class. The crystal system classification encompasses three major classes: monoclinic, orthorhombic, and triclinic. Each class represents distinct structural arrangements of atoms within the cathode material, thereby impacting its electrochemical properties and performance.

In recent years, extensive research has delved into the diverse aspects of crystal system properties in Li-ion batteries, providing crucial insights into material behavior and performance. Notably, foundational work such as that referenced in [1] has explored sulfate-based polyanionic compounds, elucidating their synthesis, crystal chemistry, and electrochemistry aspects. Subsequently, studies referenced in [2] have further expanded our understanding by addressing additional facets of crystal system properties in Li-ion batteries. Moreover, the emergence of machine learning techniques has significantly advanced this domain, as evidenced by [3], which successfully utilized these methods to discover solid Li-ion conducting materials. Additionally, contributions from studies referenced in [4] have played a significant role in enhancing our understanding of crystal system properties, covering a wide range of topics such as machine learning prediction, materials design, and state prediction in Li-ion batteries.

However, existing research exhibits certain limitations. While studies such as those referenced in [5] and [6] have developed machine-learning-based tools for cathode optimization and materials discovery, challenges remain in accurately capturing the complex nature of Li-ion battery materials. Furthermore, the need for more sophisticated modeling approaches, as highlighted in [7-8], underscores the ongoing quest for predictive models capable of addressing the intricacies of crystal system properties in battery materials. Additionally, contributions from studies referenced in [9] and [10] have further enriched our understanding of crystal system properties, covering topics such as machine learning prediction, materials design, and state prediction in Li-ion batteries.

To address these limitations, we propose a novel approach based on Deep Neural Networks (DNNs) [11] for

predicting the crystal system class of Li-ion silicate cathodes. We define the problem as a multi-class classification task, where the input features include physical and chemical properties such as formation energy, band gap, and volume, along with categorical descriptors like formula and space group. The DNN architecture will leverage the expressive power of deep learning to learn intricate patterns and relationships within the dataset, enabling accurate classification across the three crystal system classes: monoclinic, orthorhombic, and triclinic. By integrating both numerical and categorical features, our model aims to capture comprehensive information about the cathode materials, leading to improved predictive performance compared to traditional machine learning approaches.

2. RELATED WORK

In recent years, there has been a surge in research focusing on crystal system properties for Li-ion batteries, driven by the increasing demand for efficient energy storage solutions. Several studies have explored various aspects of crystal chemistry, electrochemistry, and prediction methodologies to enhance the performance and design of battery materials. Rousse and Tarascon [1] investigated sulfate-based polyanionic compounds for Li-ion batteries, emphasizing the synthesis, crystal chemistry, and electrochemistry aspects. Their work provided valuable insights into the structural characteristics and electrochemical behavior of these compounds, laying the foundation for further research in this area. Machine learning techniques have emerged as powerful tools for predicting crystal systems and optimizing battery materials. Shandiz and Gauvin [2] applied machine learning methods to predict the crystal system of cathode materials, demonstrating the potential of computational approaches in accelerating material discovery and design.

Sendek et al. [3] utilized machine learning to discover solid Li-ion conducting materials, highlighting the effectiveness of data-driven approaches in identifying promising candidates for battery applications. Similarly, Kauwe, Rhone, and Sparks [4] conducted data-driven studies to explore Li-ion battery materials, leveraging computational methods to uncover structure-property relationships. Houchins and Viswanathan [5] developed an accurate machine-learning calculator for optimizing Li-ion battery cathodes, providing a practical tool for materials design and optimization. Liu et al. [6] further demonstrated the utility of machine learning in materials design and discovery for rechargeable batteries, emphasizing the importance of computational approaches in overcoming traditional experimental limitations.

Zhao et al. [7] focused on machine learning prediction of activation energy in cubic Li-argyrodites, employing hierarchically encoding crystal structure-based descriptors to enhance predictive accuracy. Lv et al. [8] highlighted the role of machine learning as an advanced platform for materials development and state prediction in lithium-ion batteries, showcasing its potential in accelerating research and innovation in the field. Prosini [9] investigated crystal group prediction for lithiated manganese oxides using machine learning techniques, contributing to the understanding of crystallographic transformations in battery materials. Kee and Tran [10] provided an introductory overview of machine learning methods and their applications in Li-ion batteries, emphasizing their significance in energy storage and conversion materials research. Overall, the aforementioned studies underscore the importance of crystal system properties in Li-ion battery materials and highlight the growing influence of machine learning techniques in advancing battery technology. These contributions collectively contribute to the ongoing efforts to develop efficient and sustainable energy storage solutions.

Large Language Models (LLMs) have demonstrated their efficacy across diverse domains, showcasing their versatility and applicability. Xie et al. (2024) introduced a Conv1D-based approach for multi-class classification in legal citation text, emphasizing the potential of LLMs in legal research [13]. Similarly, Xiong et al. (2024) employed LLMs, specifically BERT-RCNN fusion, to enhance sentiment analysis of COVID-19 tweets, highlighting their effectiveness in social media analysis [14]. Furthermore, Liu et al. (2024) explored the use of attention mechanisms in news recommendation systems, indicating the broad utility of LLMs in personalized content delivery [15]. Su et al. (2024) conducted a systematic literature review, shedding light on the role of LLMs in forecasting and anomaly detection, offering insights into their application in predictive analytics [16]. Additionally, Zhao et al. (2024) proposed BERTFusionDNN to improve e-commerce recommendations, underscoring the significance of LLMs in enhancing customer experiences [17]. In another context, Qiao et al. (2023) analyzed the application of machine learning in financial risk early warning and regional prevention and control, further showcasing the diverse applications of LLMs [18]. Moreover, Ni et al. (2024) introduced Smartfix, leveraging machine learning for proactive equipment maintenance in industry 4.0, highlighting the role of LLMs in industrial applications [19]. These studies collectively emphasize the broad scope of LLMs and their potential across various fields of research and industry.

3. ALGORITHM AND MODEL

3.1 DNN MODEL

As shown in Figure 1, we propose a Deep Neural Network (DNN) approach for multi-class classification of crystal system properties in lithium-ion batteries. Our method leverages the expressive power of DNNs to learn intricate patterns and relationships within the dataset, enabling accurate classification across three crystal system classes: monoclinic, triclinic, and orthorhombic. The input features consist of physical and chemical properties of Li-ion silicate cathodes, including 'Formation Energy (eV)', 'E Above Hull (eV)', 'Band Gap (eV)', 'Nsites', 'Density (gm/cc)', and 'Volume'. Additionally, categorical descriptors such as 'Formula', 'Spacegroup', and 'Has Bandstructure' are incorporated to capture structural and compositional characteristics of the cathode materials. The output of our model is a multi-class classification task, where the predicted classes correspond to the crystal system classification: monoclinic, triclinic, and orthorhombic.

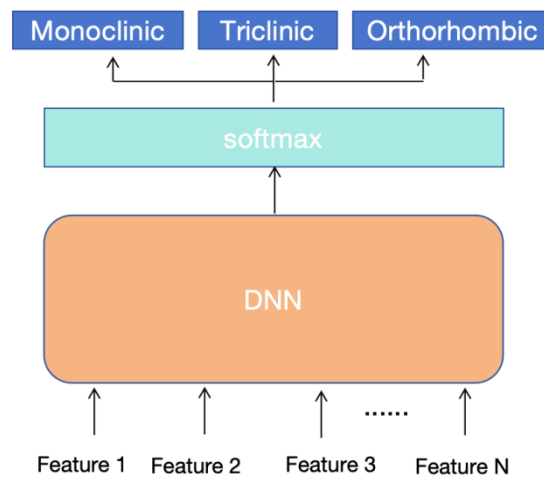


Figure 1: DNN

Our DNN architecture comprises multiple layers of densely connected neurons, allowing the model to learn complex representations of the input features. We employ activation functions such as ReLU (Rectified Linear Unit) [12] to introduce non-linearity and improve the model's capacity to capture intricate relationships in the data. The DNN embedding for input features, denoted as E_{DNN} , is obtained by passing the input features through the DNN model.

$$E_{DNN} = DNN(x) \tag{1}$$

Following feature embedding, the data is passed through a classification layer comprising fully connected layers and a softmax activation function. This layer outputs probabilities for each class (monoclinic, orthorhombic, triclinic), enabling the model to predict the crystal system with confidence.

$$P(\text{Sentiment} = c | E_{DNN}) = \text{Softmax}(FC(E_{DNN})) \tag{2}$$

where represents one of the classes (monoclinic, orthorhombic, triclinic).

3.2 Prospects of Large Language Models (LLM)

Large Language Models (LLMs) such as GPT [41], GPT-2 [42], and GPT-3 [43] offer a promising approach to tackle the challenges associated with crystal system classification in lithium-ion batteries. With their advanced natural language processing capabilities, LLMs are adept at analyzing textual data related to material properties,

experimental findings, and research literature in battery technology. Utilizing pre-trained LLMs like GPT-3 enables the extraction of valuable insights from vast amounts of unstructured textual data, including scientific papers, patents, and technical reports.

Research [20-21] advances legal text classification and enhances sentiment analysis of COVID-19 tweets by leveraging Large Language Models (LLMs). Furthermore, [22] contributes to improving e-commerce recommendations, while [23] focuses on enhancing e-commerce chatbots, both utilizing LLMs to uncover insights and enhance customer experiences. Several studies [24-28] explore diverse applications of machine learning, including financial risk analysis, equipment maintenance, and resource allocation, as well as image classification and object detection. They often integrate LLMs to achieve better performance and efficiency. Studies [29-30] investigate anomaly detection and predictive modeling, respectively, utilizing Large Language Models to analyze multi-omics data and optimize personalized education recommendations. In addition [31], addresses resource allocation in network functions virtualization environments, while [32] explores automatic recognition of static phenomena in retouched images, demonstrating the versatility of LLMs across various domains [33-34]. Delve into enterprise financial risk prediction and particle filter SLAM for vehicle localization, highlighting LLMs' capability in tackling complex challenges. Research [35] examines semi-supervised learning in image classification, whereas [36] focuses on implementing computer vision technology for medical image analysis, showcasing the wide range of applications for LLMs in machine learning and AI. Furthermore, studies [37-38] investigate acoustic encoders and the fusion of labeled and unlabeled data in image classification, showcasing LLMs' potential in enhancing model performance. Finally [39-40], underscore the emerging synergies between Large Language Models and traditional machine learning methods, particularly in the realm of e-commerce recommendations.

4. EXPERIMENTS

4.1 Datasets

The dataset provided comprises essential physical and chemical attributes of Li-ion silicate cathodes, crucial for predicting the class of a Li-ion battery. These batteries are classified based on their crystal system, which includes three primary categories: monoclinic, orthorhombic, and triclinic. The dataset features include 'Formation Energy (eV)', 'E Above Hull (eV)', 'Band Gap (eV)', 'Nsites', 'Density (gm/cc)', 'Volume', 'Formula', 'Spacegroup', 'Has Bandstructure'. Split into training and test sets in an 8:2 ratio, this dataset offers a comprehensive foundation for developing predictive models to classify Li-ion battery crystal systems, facilitating advancements in energy storage technology.

4.2 Evaluation metrics

The Macro F1-Score [44] serves as a holistic measure for assessing the effectiveness of a multi-class classification model, taking into account both precision and recall across all classes. This metric proves especially valuable in scenarios where there exists an imbalance in class distribution, guaranteeing that the evaluation accurately reflects the model's performance for each individual class. Precision quantifies the accuracy of positive predictions, with Precision Macro representing the average precision calculated across all classes and is calculated as:

$$Precision_{macro} = \frac{1}{N} \sum_{i=1}^N \frac{TP_i}{TP_i + FP_i} \tag{3}$$

Recall assesses the model's capability to identify all positive instances. Recall Macro, on the other hand, represents the average recall computed across all classes and is calculated as:

$$Recall_{macro} = \frac{1}{N} \sum_{i=1}^N \frac{TP_i}{TP_i + FN_i} \tag{4}$$

The Macro F1-Score is determined by computing the harmonic mean of precision and recall, assigning equal importance to every class. It is defined as:

$$F1_{macro} = \frac{2 * Precision_{macro} * Recall_{macro}}{Precision_{macro} + Recall_{macro}}$$

Where:

TP_i is the number of true positives for class i .

FP_i is the number of false positives for class i .

FN_i is the number of false negatives for class i .

N is the total number of classes.

In summary, the Macro F1-Score provides a balanced evaluation of a model's multi-class classification performance, ensuring fairness in assessments amid imbalanced class distributions by considering precision and recall across all classes.

4.3 Results

In our study, we assessed various classification models' effectiveness in predicting crystal system classes for lithium-ion batteries. The models scrutinized encompassed Support Vector Machine (SVM) [45], Logistic Regression (LR) [46], Naive Bayes (NB) [47], and a Voting Model, amalgamating SVM, LR, and NB. Additionally, we proposed a novel Deep Neural Network (DNN) approach for comparison.

Table 1: Model Results

Model	Macro Precision	Macro Recall	Macro F1-Score
SVM	0.37	0.45	0.39
LR	0.54	0.48	0.49
NB	0.29	0.37	0.32
Voting Model	0.35	0.44	0.38
DNN	0.82	0.82	0.81

From the results presented in Table 1, it is evident that the DNN model outperforms the other models across all evaluation metrics. The DNN achieved a Macro F1-Score of 0.81. These results demonstrate the effectiveness of our DNN approach in accurately classifying crystal system properties in lithium-ion batteries. Comparatively, the SVM, LR, and Voting Model yielded lower performance scores across all metrics, indicating their limitations in capturing the complexities of the dataset. Additionally, the Naive Bayes (NB) model showed the lowest performance among all models, with the lowest Macro Precision, Macro Recall, and Macro F1-Score. Overall, our findings underscore the superiority of the DNN model in predicting crystal system classes, highlighting its potential for enhancing battery technology through more accurate classification of material properties.

5. CONCLUSION

In conclusion, our study presents a novel Deep Neural Network (DNN) approach tailored for the classification of crystal system properties in lithium-ion batteries. By harnessing a comprehensive set of physical, chemical, and categorical features, our model adeptly predicts the crystal system classes monoclinic, orthorhombic, and triclinic. The incorporation of deep learning techniques enables the extraction of intricate patterns and relationships within the dataset, facilitating precise classification across multiple classes. Our approach addresses the inherent challenges associated with capturing the complexities of Li-ion battery materials with accuracy. By leveraging the power of machine learning methodologies, particularly deep learning, our model offers valuable insights into crystal system properties, thereby advancing our understanding and optimization of battery performance.

The implications of our study extend beyond theoretical advancements, with practical implications for researchers and engineers in the field. Our developed model serves as a robust tool for designing and optimizing lithium-ion battery materials, enabling the development of efficient and sustainable energy storage solutions. Through accurate classification of crystal system properties, our model empowers stakeholders to make informed decisions in material selection and battery design, ultimately driving progress towards environmentally friendly and cost-effective energy storage technologies. In summary, our study demonstrates the significant potential of machine learning, particularly deep learning, in revolutionizing the field of battery technology. By bridging the gap between theory and practice, our model contributes to the ongoing quest for efficient and sustainable energy solutions, laying the groundwork for future innovations in the realm of lithium-ion batteries.

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