See Deeper: Identifying Crystal Structure from X-ray Diffraction Patterns

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***Abstract*—X-ray diffraction is a commonly used experimental science to detect the atomic and molecular structure of crystalline material. The process is called X-ray crystallography (XRC). Traditionally, it is done by human experts with some conjecture about what structure the crystalline material is likely to be. However, the study of crystal structure using X-ray diffraction patterns is applicable in many domains, such as chemistry, physics, biology, *etc*. It is tedious to have manual crystallography of X-ray diffraction patterns to determine a crystal structure with a massive amount of dataset. With the advent of high computational resources, deep learning techniques have taken classification to its peak. Convolution Neural Network (CNN) maps an input image into a high dimensional space and produce a low-cost function for image classification. In this paper, we deploy a variation of the Convolution Neural Network to predict crystal structure from X-ray diffraction patterns. We compare our approach with a wide range of conventional as well as modern Machine Learning based classification techniques for the structure prediction of a crystal. We report a cross-validation accuracy of 95.6% and Micro F1-score of 0.949 with our model for the proposed task which is significantly better than the other reported baseline methods.**

***Keywords*-X-ray Diffraction Pattern; Crystal Structure Classification; Deep Learning; Convolutional Neural Network; Image Processing;**

I. Introduction

The structure classification of a crystal is a fundamental task for many domains, physics, chemistry, biology, *etc*. In biology, identifying the structure of a protein can help discover a drug, analyze protein-protein interaction, and classify protein into its respective family. Since the wavelength of X-rays is on the atomic scale, X-ray Diffraction (XRD) is a primary implement for probing the structure of nanomaterials. XRD offers unparalleled precision within the quantification of nuclear spacing and it is a technique for determining crystal states in thin films. However, it is tedious to have a manual analysis of X-ray diffraction patterns to determine the structure of a crystal with a rapidly growing amount of dataset. In this paper, we present a technique to perform the automatic classification of crystal lattice structure into seven classes, *viz., Triclinic, Monoclinic, Orthorhombic, Tetragonal, Hexagonal, Rhombohedral, Cubic*.

A crystalline material has its atoms arranged in periodic lattices with long-range symmetry. As a result, it scatters a beam of X-rays in distinct patterns. Analysis of an X-ray diffraction pattern engendered by a fabric yields extensive information about the structure of the fabric, mainly, about the type of periodic lattice, its atoms are arranged. Elemental analysis of X-ray diffraction patterns is usually performed manually, often requiring human input and a few guesswork at what the structure of unknown material is susceptible to be. It becomes hard to utilize X-ray diffraction data in the manual analysis of large datasets. Automating the method of structure classification from X-ray patterns would be a step toward high-throughput computational searches for materials with desirable properties - one of the goals achieved by Image Processing.

In this paper, we present a neural network-based approach to predict a crystal structure using X-ray diffraction pattern. Most modern convolutional neural networks (CNN) used for object recognition are built using the same principles: alternating convolution and max-pooling layers followed by a small number of fully connected layers. The a-CNN is a new architecture that consists solely of convolutional layers and yields state of the art performance on several object recognition datasets [1]. The all-Convolution Neural Network (a-CNN) is a deep neural network that can learn a high dimensional representation of images, which leads to more accurate classification of images [2]. X-ray diffraction pattern can also be interpreted as an image. In this paper, we present a-CNN with a bit variation for predicting the structure of a crystal. There have been several attempts to automate the process of crystal structure prediction. Agatonovic-Kustrin et al., 2000 [3] built a Naive Bayes model, which gave an accuracy of approximately 33% and it leveled up utilizing other relegation algorithms like Neural Networks which produced an accuracy of approximately 63% percent. The reason for such low results is the interdependence of the features. Recently, Yang et al., 2019 [4] interpreted various image patterns using Average Class Activation Maps (CAMs), which we have implemented to provide a deeper insight into the results obtained. A number of the precedent works have taken the reciprocal lattice parameter as a feature, but we have got processed the info and go different ways into multiple features [5], [6]. We have introduced an incipient convolutional layer instead of the traditional neural network layers which take up the layers

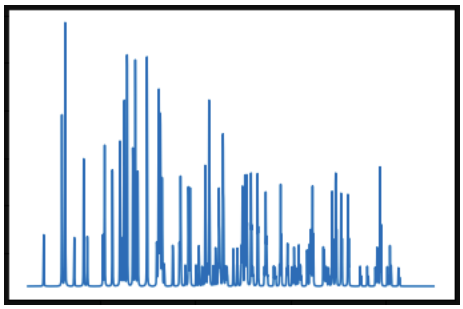

Figure 1. Input XRD Pattern

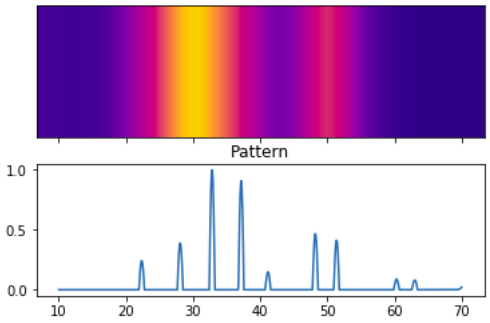

Figure 2. Output CAM for single class

connected to the neurons of the layer afore [7]. We keep the pixel relationships preserved by learning image features utilizing small areas of computer files. The detection of the lattice system is often distributed with more preponderant precision just if we increase the number of convolutional layers [8].

Considering the previous attempts, we compare a-CNN with conventional as well as state-of-the-art classification approaches, *viz.*, Naive Bayes [9], KNN [10], Logistic Regression, Random Forest [11], Support Vector Machine (SVM) [12], Gradient Boosted Regression Trees (GBRT) [13], and Multi-Layer Perceptron (MLP). Results (Section V) show that a-CNN outperforms other methods by a big margin for the classification of the structure of a crystal. Besides, we show that due to its favorable architecture for image classification, a-CNN [14] can outperform a generic Deep Neural Network (MLP) model for the structure prediction task. The input to our program is an XRD pattern (Figure 1) that consists of scattered X-ray intensity as a function of a variable called the reciprocal lattice vector (denoted as q). We discretize this pattern by slicing up the q-space into a variable number of pieces and utilize the intensity at each q as a feature. The output is a relegation (Figure 2) of the given XRD pattern into one of the seven rudimentary crystal systems. This project is the first attempt to classify the structure of a crystal using a variation of the CNN model to the best of our knowledge. We achieve an accuracy of 95.6% and an F1-score of 0.949 for the crystal structure classification.

Our contributions in this paper are as follows.

- Performed augmentation of thin filmed X-ray diffraction patterns.
- Developed a high accuracy model for lattice classification from X-ray diffraction.
- Present a detailed version of the CNN classification model by using class activation maps.

The rest of the paper is organized as follows. Section II provides a relative study of the works done for structure classification using XRD. Further, section III elaborates on the dataset used in the paper. Section IV describes the experimental setup. Section V demonstrates the results obtained, and section VI concludes the paper.

II. Literature Survey

The recent experiments are conducted upon the high voltage X-ray tubes and the X-ray beam is passed through the crystal to be experimented with, and then it's observed under photographic plate [15], [16]. The arrangement of different molecules is analyzed using the lattice constant, and then the crystal lattice system is declared. It becomes difficult when the exact crystal reciprocal lattice constant cannot be determined, or very little data is available to check through the arrangements available. Our model uses each split of the X-ray diffraction pattern as its features. The lattice constant has different methods of evaluation based upon the lattice structure, which in turn is determined from the symmetry of the unit cells of the crystal using the parameters of the cell.

A few recent XRD-related modelling studies used a conventional Artificial Neural Network (ANN) with feature engineering techniques, such as manual featurization [17], Principal Component Analysis (PCA) [18], Partial Least-Squares Regression (PLSR) [19], and various other statistical approaches [20]. Conventional machine learning methods are not able to model features that human cannot recognize. However, Park et al., [21] showed the efficacy of deep neural networks for the classification of powder X-ray diffraction (XRD) patterns in terms of crystal system, extinction group and space group. In this paper, we have taken up the reciprocal lattice constant as input parameter and then generated the XRD pattern by plotting the normalized intensity *vs.* 2-times θ as per physics definitions [22]. We have evaluated the models as provided by other authors, and it gave shallow accuracy results because not much importance was given to the fact that how to increase the number of features or handle the issue of redundant features. We uniquely addressed the issue.

In this paper, a two-dimensional ($2D$) implementation of an all-convolutional neural network (a-CNN) is presented [23]. We implemented and identified a-CNN as the significantly more accurate and interpretable classifier for this problem in comparison to the various baseline methods. On

the other hand, we have implemented class activation maps to give a specific context of the root causes of classification failures or the extent of success.

III. Dataset

One of the most prominent sources for X-ray diffraction patterns is the Inorganic Crystal Structure Database (ICSD). We downloaded 12000 known crystal structures from the ICSD for the training and testing of the models presented in the paper.[1] We deployed an open-source tool called *Platon*[2] to generate a diffraction pattern for each structure using the principles of physics. It can also be used as a library just like Pandas. The simulated training data set consists of 164 compounds extracted from ICSD with a similar composition, expected crystal symmetry, and space group as the synthesized materials of interest. All the possible single, double, ternary, and quaternary combinations of the elements of interest were extracted during database mining. The fundamental crystal descriptors extracted from the material database are used to simulate X-ray diffraction powder patterns with random crystalline orientations.

Data Augmentation: Experimentally generated thin-film X-ray diffraction patterns vary much from simulated, idealized, randomly oriented X-ray diffraction patterns. Due to expansions and contractions in the crystalline lattice, X-ray diffraction peaks shift along the 2θ axis according to the specific size and location of the different elements present in a compound, while maintaining similar periodicity based on crystal space group. To increase the size and robustness of the limited training data set and to account for these fundamental differences between real thin-films and simulated X-ray diffraction powder patterns, we perform a three-step data augmentation procedure based on physical and chemical crystal domain knowledge. They are Peak Scaling, Peak Elimination, and Pattern Shifting [24].

Each training example is a pattern of X-ray Intensity (I) as the function of a reciprocal lattice vector (q) over a fixed range of q, accompanied by an output label indicating which crystal system the pattern corresponds to. The model we use describes each such pattern by discretizing it in q to generate a vector of dimension equal to the number of discrete q-steps chosen. It can be explained in a much better way as the series of peaks in an X-ray diffraction pattern by a discrete function given below.

$$(2\theta) : I \rightarrow R+ \tag{1}$$

This maps a set of discrete angles I to positive real numbers $R+$ corresponding to peak intensities. Now we augment the data through the following sequential process of transformations σ^1_{Sc} , σ^2_S , and σ^3. Periodically random peak scaling

[1]https://databases.library.jhu.edu/databases/database/JHU04574
[2]ff

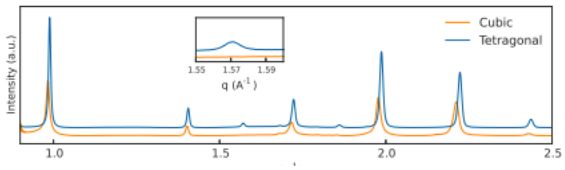

Figure 3. Two crystal systems XRD patterns

is applied. A subset of random peaks at periodic angles S is scaled by factor c, that

$$\sigma^1_{SC} = c * \sigma|_S + f|_{I \setminus S} \tag{2}$$

We perform global average pooling in the last convolutional layer. The size of the data set is critical for obtaining high accuracy and F1-score. The a(all)-CNN accuracy was computed for various combinations of augmented experimental patterns to explore augmented data set size. It augmented simulated data set sizes (number of augmented X-ray diffraction spectra originating from the 164 simulated ICSD spectra). As the size of the temporary and augmented data sets increase, the mean accuracy quickly approaches the asymptotic accuracy. The critical augmented data set size seems to be around 800 augmented spectra. The model's accuracy is more sensitive to the augmented experimental data set size, likely because most of the data set variance comes from the experimental X-ray diffraction patterns, which causes the accuracy to get up monotonically.

IV. Experimental Setup

We compare our approach with various standard classification algorithms (Figure 5) and (Figure 6) for the structure classification of a crystal. This section describes the methods we have implemented.

Naive Bayes: In this method, we discretize each X-ray diffraction pattern in both axes - such that each pattern is represented as a vector of dimension equal to the number of discrete steps chosen in q-space. The intensity at each q is modeled as a multinomial distribution over a finite number of steps. Utilizing these feature vectors, we implement the Naive Bayes classifier to model probabilities of an incipient pattern emanating from each of the seven crystal systems (shown in Figure 3). The fundamental posit that the Naive Bayes method makes is that separate features in the input are conditionally independent of one another, given the output. In other words, the probability of one input feature having a certain value has no bearing on the probability of a different feature, even when the correct output corresponding to that input vector is kenned. The probability at each being a particular lattice system is calculated and multiplied with the previous ones. However, the feature independence assumption is not valid in the case of a crystal structure. Upon performing cross-validation, we achieved an accuracy of 45 percent.

Random Forest: In this case, a multitude of decision trees was responsible for deciding the lattice system. We obtained an accuracy of 64%. This is quite higher than Naive Bayes, though, but the other parameters like F1-scores were too low to accept this classifier for the purpose.

K Nearest Neighbour: In this method, clustering is performed. We have obtained an accuracy of nearly 56% with a moderate F1-score. This clustering mechanism is not useful for detecting the crystal lattice systems as some features may be redundant, while others may not.

Decision Tree: In this method, we have obtained an accuracy of 32%, which is fair enough to be discarded for the classification of lattice systems. This method is similar to predictive modeling approaches, but the features or X-ray diffraction pattern splits cannot be used for prediction purposes.

Support Vector Machine: In this method, an optimal boundary is generated for the classification of the crystal lattice systems based upon the splits of the X-ray diffraction pattern. We have obtained an accuracy of 49%.

Gradient Boosting: In this method, we have obtained an accuracy of 38%. We had to train the model, considering a set of weak machine learning algorithms. Hence, this method is also not feasible to be applied for the classification purpose in this case.

all-Convolutional Neural Network: We propose CNN with some modifications to achieve the best results for the structure prediction of a crystalline material using an X-ray diffraction pattern. In this case, rather than just implementing with regular neural network layers, we have introduced convoluted layers. The quandary of novel material development is inherently a multi-class relegation quandary, in which the classes for training and testing purposes can often be imbalanced as some material families are better characterized than others (*e.g.*, lead-predicated crystals are better represented in material databases than more incipient lead-free perovskites).

Results (Section V) show that the a-CNN classifier performs better than any other classification technique. The CNN architecture implemented comprises three $1D$ convolutional layers, with 32 filters each, and strides and kernel sizes of 8, 5, and 3 units, respectively. The activation function between layers is $ReLu$. A global average pooling layer (acting as a weak regularizer) and a final dense layer with softmax activation are used. The loss function minimized is binary cross-entropy. We use early stopping with a batch size of 128 during training to minimize the loss function and an algorithm called *Adam's* optimizing algorithm. Our model does not have max-pooling layers between convolutional layers, and also lacks a set of dense layers in the final softmax classification layer. These modifications significantly reduce the number of parameters in the neural network, allows faster and simpler training, and are less prone to over-fitting. The a-CNN trained after data augmentation has

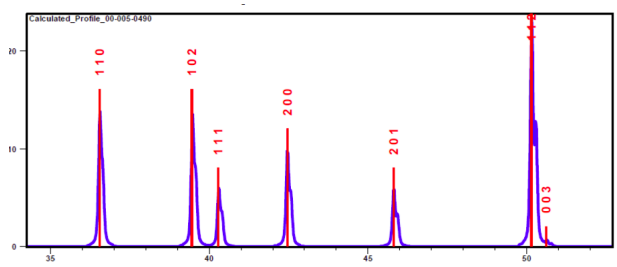

Figure 4. Miller Indices from Peaks

an accuracy of 95% and 86% for crystal dimensionality and space group (Figure 5 and Figure 6) classifications, respectively.

The XRD pattern can be analyzed as a 2D image that resembles the pattern of the crystal lattice system it belongs to. Hence, peak splitting and scaling are to be performed to determine the crystal lattice from the X-Ray Diffraction pattern. Peak scaling is applied periodically along the 2θ axis to account for different thin-film preferred orientations. The peak scaling performed can be better represented as in Figure 4. It is necessary to gather around the Miller Indices for determining the crystal structure.

V. Results

In our work, we have preprocessed and augmented the dataset. We have performed classification of two types- one based upon the crystal dimensionality and the other based upon the crystal lattice structure. Crystal dimensionality - $0D$, $2D$, $3D$. We represent the X-ray diffraction pattern as either a vector to implement a-CNN and other baseline classifiers. The following classification methods are tested using a vector representation of the X-ray diffraction pattern: Naive Bayes, k-Nearest Neighbors, Logistic Regression, Random Forest, Decision Trees, Support Vector Machine, Gradient Boosting Decision Trees, a Fully Connected Deep Neural Network (Multi-Layer Perceptron), and a-CNN with a global pooling layer.

In this paper, we are dealing with the multi-class classification problem. The classes for training and testing purposes can often be imbalanced as some material families are better characterized than others. For method development in this work, we consider the following metrics: subset precision, defined as the number of correctly relegated patterns among all test patterns, and in turn, F1-score.

We have implemented **Gradient Descent**, **Stochastic Descent** and **Adam** optimizer algorithms to minimize the cost function. The best results were obtained by using the **Stochastic Descent**, and the results were improved upon by using mini-batch gradient descent. Figure 5 and 6 show the cross-validation accuracy for crystal systems and space groups respectively, while Figure 7 presents F1-score obtained using $2D$ vector (crystal dimension) with

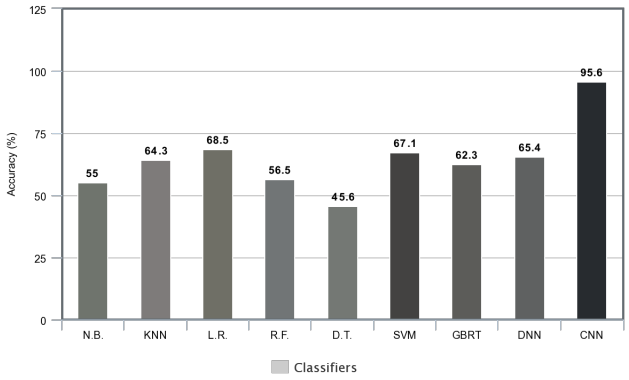

Figure 5. Classification for the crystal systems

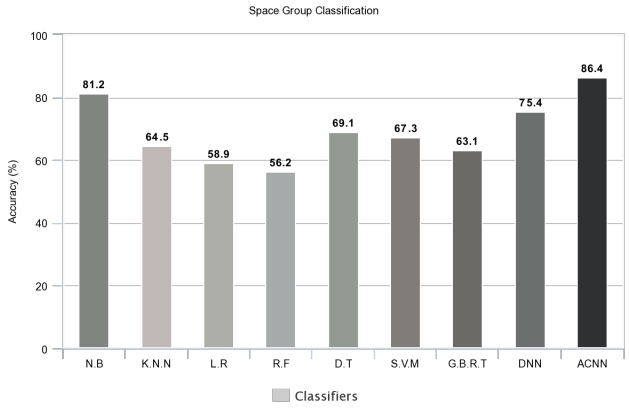

Figure 6. Classification for the space groups

mini-batch (batch-size: 50) gradient descent for CNN. The activation function between layers is not a ReLu in our case. In one of the layers, we have implemented softmax regression (Equation (3)) and also Logistic regression in order to compare the results and get the best. CNN outperforms other classification methods by a big margin for the structure prediction of crystalline material.

$$\text{Softmax}(x_i) = \frac{\exp(x_i)}{\sum_j \exp(x_j)} \tag{3}$$

Error Analysis: We implemented the extraction of Class Activation Maps from the weight distribution of the average pooling layer. The model's precision is more sensitive to the augmented experimental dataset size, likely because most of the dataset variance emanates from the experimental X-ray diffraction patterns. We have detected a few issues using the Class Activation Maps, which were caused by the phase mixtures in the sample or lack of X-Ray diffraction patterns in the training data set, causing a lack of discriminative or differentiating features. This issue can be resolved by increasing the size of training data or experimental training data of certain classes, and mostly it depends upon the purity of the crystal.

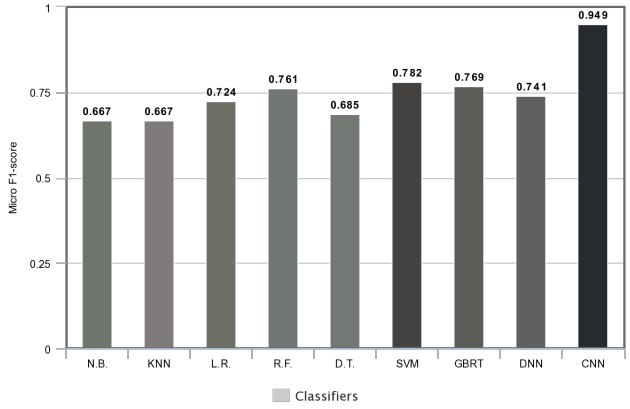

Figure 7. Classifiers with Micro F1-scores

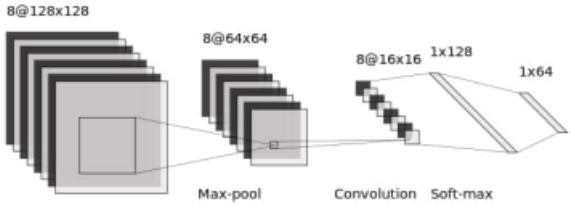

Figure 8. CNN used

Class activation has been performed in the last layer. CNN performs better classification than the k-nearest neighbors' method. In our dataset, the discrimination between thin film and powder spectra may not have been captured properly. In case there are many similarities between the data for a single class, then the accuracy under the k-nearest neighbor classification would be much higher. In general, CNN performs better than K-Nearest Neighbours for classification of time series, which is consistent with our results. It is visible (Figure 8) we have not included any max-pooling layers other than the single global max-pooling. At the last layer, we have implemented soft-max regression as an algorithmic cost function optimizer. Figure 8 shows, the presented CNN is different from the general CNN architecture, as it neither has max-pooling layers between convolutional layers nor a set of dense layers in the final soft-max classification layer. These changes help reduce the number of parameters in the neural network and allow the process of training to be fast enough and avoid the risk of overfitting.

VI. Conclusion

In this paper, we presented a system to classify a crystal structure into seven categories, *viz., Triclinic, Monoclinic, Orthorhombic, Tetragonal, Hexagonal, Rhombohedral, Cubic*. Convolution Neural Network (CNN) maps an input image into a high dimensional space and produce a low-cost function for image classification. We showed that a

variation of CNN using X-ray diffraction pattern as an input outperforms other standard classification algorithms, *viz., Naive Bayes, KNN, Logistic Regression, Random Forest, Decision Tree, SVM, Gradient Boosted Regression Trees, Deep Neural Network* for the crystal structure classification. We achieved a cross-validation accuracy of 95.6% and Micro F1-score of 0.949 with a-CNN for the proposed task, which is significantly better than other reported baseline methods. The structure classification of a crystal is a fundamental task for many domains, specifically the biomedical domain. For example, identifying the structure of a protein can aid the discovery of a drug, analysis of protein-protein interaction, and the classification of protein into its respective family.

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